JAEA-Review 2008-055



JAEA Takasaki Annual Report 2007

(Ed.) Shigeru TANAKA

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Takasaki Advanced Radiation Research Institute

November 2008

Japan Atomic Energy Agency

日本原子力研究開発機構

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Takasaki Advanced Radiation Research Institute

Japan Atomic Energy Agency

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(Received September 19, 2008)

JAEA Takasaki annual report 2007 describes research and development activities performed from April 1, 2007 to March 31, 2008 with Takasaki Ion Accelerators for Advanced Radiation Application (TIARA, four ion accelerators), and electron/gamma-ray irradiation facilities (an electron accelerator and three ⁶⁰Co gamma-ray irradiation facilities) at Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency (JAEA Takasaki). These activities are classified into four research fields: 1) space, nuclear and energy engineering, 2) environment conservation and resource security, 3) biotechnology and medical application, and 4) advanced materials, analysis and novel technology. This annual report contains 174 reports consisting of 166 research papers and 8 status reports on operation/maintenance of the irradiation facilities described above, and a list of publications, related press-releases, TV programs, patents, and the type of research collaborations as Appendices.

Keywords: TIARA, Ion Accelerators, Electron Accelerator, Gamma-ray Facilities, Nuclear and Energy Engineering, Environment Conservation, Resource Security, Biotechnology, Medical Application, Advanced Materials, Analysis, Novel Technology, Materials for Space, Semiconductors, Inorganic Materials, Organic Materials, Functional Materials, Radiation Chemistry, Radiation Biology, Radioisotope Production, Nuclear Chemistry, Material Analyses, Solid State Physics, Microbeam Technology, Accelerator Technology, Accelerator Operation, Safety Control

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高崎量子応用研究所研究年報 2007

日本原子力研究開発機構 高崎量子応用研究所 (編) 田中 茂

(2008年9月19日受理)

高崎量子応用研究所研究年報 2007 は、同研究所にある TIARA 施設(イオン加速器 4 基) 及び電子・ガンマ線照射施設(電子加速器 1 基、⁶⁰Co ガンマ線照射施設 3 棟)を利用して 2007 年 4 月 1 日から 2008 年 3 月 31 日までの間に行われた研究・技術開発成果をまとめ たものである。この研究年報には、1)宇宙・原子力・エネルギー、2)環境・資源、3) バイオ技術・医学応用、4)先端材料・計測・基盤技術の4分野に分類した166 編の論文 及び8 編の施設の運転・管理状況報告からなる合計 174 編を収録する。また、論文リスト、 新聞発表、テレビ放映、出願特許、及び研究実施形態・利用施設の一覧表を付録として含 む。

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PREFACE

This report covers research and development activities in Takasaki Advanced Radiation Research Institute, JAEA, during the period from April 2007 to March 2008 conducted with TIARA (Takasaki Ion Accelerators for Advanced Radiation Application), electron beam and Co-60 γ -ray irradiation facilities.

One hundred seventy four papers are appeared in this annual report in the fields of

- 1) Space, Nuclear and Energy Engineering,
- 2) Environment Conservation and Resource Security,
- 3) Biotechnology and Medical Application,
- 4) Advanced Materials, Analysis and Novel Technology,
- 5) Status of Irradiation Facilities.

In the field of Space, Nuclear and Energy Engineering, radiation-induced degradation of solar cells and malfunction of semiconductor devices have been examined by using high energy electrons, protons and heavy ions to develop next generation electronic system used in artificial satellites. Single event transient currents induced in semiconductor devices by energetic ions have been investigated experimentally and theoretically for completing prediction models of device malfunction caused by cosmic-rays. Radiation resistant devices have also been developed based on wide band-gap semiconductors like SiC and GaN. Radiation degradation of organic materials, e.g., power supply and data communication cables using in J-PARC, components of the ITER remote maintenance system and plastics used for nuclear fuel treatment, has been tested for aiming at their practical application. As for structural materials used in light water and fusion reactors, microstructural change in pressure vessel steels, fuel ceramics and tritium breeder materials has been intensively studied using TIARA for securing reactor safety. In addition, polymer electrolyte membranes exhibiting high performance suitable for hydrogen fuel cells have been developed by radiation grafting and cross-linking technique using ion beams, electron beams and γ -rays.

In the field of Environment Conservation and Resource Security, metal ion adsorbents have been developed with radiation grafting technique and applied for removing toxic elements like boron and lead in waste water. Radiation grafting technique has also been adopted for surface modification of materials in industrial use, fabricating low loss millimeter-wave planar antennas used in intelligent transport system. Biodegradable carboxymethyl-cellulose (CMC) hydro-gels have been investigated using radiation cross-linking for the application to novel phosphorus absorbents useful for environmental purification. Mixture of CMC and other plant-derived polymers like carboxymethyl-starch and konjac-mannan has been studied to improve the degree of swelling and mechanical strength of gels. R&D of the decomposition and removal of trace amount of pollutants such

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as toxic organic compounds in flue gas or waste water using electron beams and γ -rays have been performed to develop new process technologies for environment conservation.

In the field of Biotechnology and Medical Application, functional analysis of proteins involved in DNA repair and replication has been progressed. By using plant seeds, optimum irradiation condition for ion beam breeding has been explored. The ion beam breeding technique has been applied for many varieties of plants and microorganisms such as flower, crop, vegetable, yeast, alga and fungus. Molecular mechanisms in radiation-induced bystander effects were investigated using heavy-ion microbeams. The foci formation of γ H2AX and pNBS1 in un-irradiated cells was almost completely suppressed by the addition of dimethyl-sulfoxide or γ -hexachlorocyclohexane, suggesting that reactive oxygen species and nitric oxide were initiator/mediators for evoking heavy ion-induced bystander responses. 13N-labeled nitrogen gas for plant study was first applied to the positron imaging and succeeded in obtaining the visualization of nitrogen fixation by soybean nodule. Using fluorescent dye and radionuclide Cu-64 produced by the AVF cyclotron, the difference of images from PET and optical imaging was evaluated as multimodal imaging system for medical use. The in-air micro-PIXE system has been used for investigation of elemental analysis; B, Br, Cd, Pt and other metal signals were visualized and quantified in order to determine their distributions in a cell, organs or tissues using a various kind of disease model.

In the field of Advanced Materials, Analysis and Novel Technology, based on the formation of ion tracks in polymer thin films, fabrication of nano-fibers/wires and ion-track membranes have been performed using high-energy heavy-ion beams. Optical properties of SiO₂ have been studied by several groups with the use of γ -rays and ion beams to investigate radiation defects/damage in materials. Studies on radiation defects/damage induced by high-energy electrons were performed on FeRh, CeO₂, and heavily B-doped diamond thin films. Quantum structures introduced in SiC by H- or He-ion implantation were investigated with a slow positron beam. First-principles molecular dynamics simulation was applied to the SiO₂/SiC interface of Si devices. The in-air micro-PIXE system was refined for two- or three-dimensional measurement of elemental distribution in minute samples such as boron doped steel, metal absorbent resin and anti-cancer reagent. The Proton-Beam-Writing (PBW) technique was applied to fabrication of a Ni micro-structure and Si nano-crystals. The fundamental studies on secondary ions emitted from target materials were performed for various MeV/atom carbon cluster ions including 10 to 100keV C₆₀. A single cluster particle track in polymeric materials was also visually investigated for MeV/atom C or Al cluster ions. Different kinds of monitor systems were developed based on a liquid organic scintillator for neutrons, and characteristics of chemical dosimeter were studied for their application to electron beams. Development of fast single-ion hit system, uniform beam formation and elemental technologies for micro-beam production at the cyclotron as well as development of cluster ion beam production technology at the electrostatic accelerators were in progress.

All the accelerators in TIARA, namely the AVF cyclotron, the 3MV tandem accelerator, the 3MV single-ended accelerator and the 400kV ion implanter, have been operated steadily and safely as well as an electron accelerator and Co-60 γ -ray irradiation facilities. A multi-pole magnet system to form a 2D uniform beam was installed at the downstream of the AVF cyclotron beam line.

Al Mambe

Hideki Namba, Director General Takasaki Advanced Radiation Research Institute Japan Atomic Energy Agency

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1-01 Evaluation of Element Circuits Constructing New Radiation Hardened SOI FPGAs

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Semiconductor devices with Silicon On Insulator (SOI) technology are utilized for high-performance and low-power electronics such as computer servers for high-end commercial markets. SOI is a layered structure consisting of a thin layer of silicon mounted on an insulating substrate. The typical cross section image is shown in Fig. 1. SOI technology is also very attractive as semiconductor devices for space application. SOI devices are expected to have an advantage over equivalent bulk technology devices for Single-Event Effects (SEEs) caused by heavy ions and high-energy protons because the sensitive volume for SEEs is much smaller than that of bulk Si devices. In JAXA, now we continuously perform the design and evaluation of radiation hardened logic circuits utilizing 0.15 µm fully depleted SOI process technology¹⁾. This study is indispensable in order to develop next generation semiconductor devices for space application such as (MPUs) high-speed Microprocessors and Field Programmable Gate Arrays (FPGAs).

We started the development of the radiation hardened FPGAs with SOI technology last year. This year, element circuits constructing FPGAs were designed and evaluated. In this report, we describe the evaluation results of two kinds of element circuits (the configuration bit cells and the FreeRAMs). The schematic diagrams of the configuration bit cells and the FreeRAMs circuits are shown in Fig. 2. The configuration bit cells are consisting of single port SRAMs and used for holding the information of user logic circuits in

FPGAs. The FreeRAMs are consisting of dual port SRAMs and used for memory. For each circuit, two types were designed and installed in the test chips. The difference of these types is the gate separation distance for redundant transistors (Type. A < Type. B). All these circuits were designed by the circuit techniques reported before²⁾. Test chips were fabricated with a 0.15 μ m fully depleted SOI commercial process at OKI (Tokyo, Japan). Figure 3 shows the picture of test device. In this chip, the configuration bit cells and the FreeRAMs compose the 8 kbit and 4 kbit memory array, respectively.

As for the configuration bit cells, Ar, Kr and Xe ion irradiation test results are shown in Fig. 4. Two types of SEUs (Single Event Upsets) were observed. One is the single transmission gate hit mode (STG), the other is the double hit mode (DH), which are shown in Fig. 2. In the case of the FreeRAMs, no DH mode was observed but STG mode SEU was considerably observed. Some multiple bit errors were also observed. These results indicate that some mitigation techniques are necessary to improve the SEU sensitivity of FPGAs. We plan to continue this study in order to construct the radiation hardened SOI FPGAs for space.

References

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- A. Makihara et al., IEEE Trans. Nucl. Sci., Vol. 53, 3422 (2006).
- 2) H. Shindou et al., TIARA annual report 2006 (2007) 6.



Fig. 2 Schematic diagrams of element circuits for FPGAs.







1-02 Comparison of the Experimental Results with Simulated Results of Charge Induced in MOS FET by Heavy-ion Irradiation

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One of the most detrimental effects on semiconductor devices in radiation environments is Single-Event Effect (SEE). When a high-energy heavy ion strikes a device, electron-hole pairs (e-h) are generated along the ion-track and they can create sufficient transient current to cause incorrect actions such as Single-Event Upset (SEU). Recently, the Silicon-On-Insulator (SOI) technology has been applied to high SEE tolerant devices, because it has been believed that the charge collection is suppressed by the existence of a buried oxide layer¹). However, anomalous charge collection in the SOI device was reported²). Therefore, it is important to make clear the generation mechanism of current through an insulation layer due to heavy-ion irradiation.

In this paper, the transient current induced through the gate oxide of Metal-Oxide-Semiconductor Field Effect Transistors (MOSFETs) by single ion is investigated, and we discuss the origin of current induced through an oxide layer by heavy-ion irradiation.

Aluminum gate p-channel (p-) MOSFETs were fabricated on n-type Si substrates. The gate oxide layers were grown by thermal oxidation at 1050 °C in dry O_2 ambient. Subsequently, the samples were annealed in N_2 ambient at 1050 °C for 30 min. The thickness of the gate oxide layer (d_{ox}) is 42 nm, and the channel length (L) and width (W) are 100 and 300 μ m, respectively. Heavy-ion irradiation tests were carried out using a Transient Ion Beam Induced Current (TIBIC) measurement system in conjunction with a Single Ion Hit (SIH) system.

Figure 1 shows the gate transient current induced in p-MOSFET by 15 MeV oxygen (O) ions. The gate voltages (Vg) between -3 and -10 V were applied during irradiation. The drain and source electrodes were connected to the



Fig. 1 Transient current and collected charge induced in a MOSFET at Vg= -3 to -10 V by 15 MeV O ions. Drain and source electrodes are connected to the substrate electrode during irradiation.

substrate (GND) electrode. The peak height increases with increasing bias voltage and the collected charge decreases with increasing bias. Also, the transient current with both positive and negative peaks is observed, and the total charge obtained by the integration of the transient current becomes almost zero within 100 ns after irradiation. This is the same result obtained from MOS capacitors irradiated with heavy ions, and this behavior can be interpreted in terms of the displacement current. Therefore, the result obtained for MOSFETs in this study can be also explained in terms of the displacement current.

Simulation of the transient current induced by oxide-mediated displacement current was carried out using the Synopsis TCAD version 10. The DESS package is used to simultaneously solve the Poisson and e-h continuity equations under cylindrical symmetry in this study. The estimation of the carrier density as a function of depth (the ion track structure) was carried out on the basis of the Kobetich-Katz theory³⁾. In this simulation, the value of d_{ox} was applied to be 40 nm. The donor concentration in the n-type substrate was 1.0×10^{15} cm⁻³ and the acceptor concentration in the p-type drain and source regions was 1.0×10^{17} cm⁻³. Figure 2 shows the comparison of the experimental results with the simulated results. A good agreement between the calculated and the experiment results is shown in the figure. This suggests the validity of the simulation technique used in this study.

References

- O. Musseau et al.: IEEE Trans. Nucl. Sci., NS-43 (1996) 60.
- P.E. Dodd et al.: IEEE Trans. Nucl. Sci., NS-48 (2001) 1893.
 E. J. Kobetich and R. Katz : Phy. Rev. 170 (1968) 391.



Fig. 2 Comparison of the experimental results with simulated results of transient current and collected charge induced in for p-MOSFETs at Vg=-10 V by 15 MeV O ion irradiation.

1-03 Total Dose Effects on Heavy-Ion Induced Gate Current in MOS Structure

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In order to clarify the charge collection mechanism through a buried oxide layer in SOI structure, the single heavy-ion induced transient gate current in MOS structure has been investigated. In this study, we discuss the total ionized dose (TID) effects on the transient current. We measured the transient current in MOS capacitors before and after gamma ray irradiation. From the results of total dose irradiation test, it was found that the peak of transient current was changed by the build-up charge in oxide layer, but the current was also dominated by displacement current after total dose irradiation.

宇宙環境下で半導体デバイスを使用する場合、重イオ ンなど高速荷電粒子の照射により誘起される電流に起因 した、シングルイベント現象が問題となる。能動領域の薄膜 化が可能なSOI (Silicon on Insulator) デバイスは、埋め込 み酸化膜の存在により能動領域が限定されているために 高い耐放射線性が予想されているものの、予想を大きく上 回る電荷収集が観測されるとの報告りもあり、酸化膜を有 する構造における重イオン誘起電流の発生メカニズム解 明が非常に重要である。我々はこれまでに、MOSFETにお ける重イオン誘起ゲート電流の解析より、酸化膜を介する 電流は変位電流が支配的であるとの結論を得た²⁾。一方、 MOS構造の放射線吸収線量が増加すると、酸化膜中の 固定正電荷密度、及び酸化膜リーク電流が増加すること が知られており(トータルドーズ効果)、これらによる照射誘 起電流の変化が予想される。そこで本研究では、γ線照 射によるMOS構造の重イオン照射誘起ゲート電流の変化 について評価を行った。

p形およびn形バルクSi基板を用いて、酸化膜厚 100 nm, Alゲート電極直径100 μm のMOSキャパシタ(p-MOSおよびn-MOS)を作製した。試料に対し、日本原子力 研究開発機構において、吸収線量率6.3 kGy(SiO₂)/h, 1 時間の⁶⁰Co-γ線照射を行った。なおγ線照射時は、酸化 膜電界が0.3 MV/cmとなるよう電極に正の電圧を印加した。 照射前後において、同機構の重イオンマイクロビームシス テムおよびSingle Ion Hitシステムを用いて単一酸素イオン (15 MeV)の照射を実施し、イオン照射誘起ゲート過渡電 流を測定した。また、γ線照射前後において容量-電圧特 性およびリーク電流特性の測定を行った。

 γ 線照射前後において、重イオン照射誘起電流を測定 した結果、両MOSキャパシタにおいて、同一電圧印加時 の過渡電流ピーク値が、 γ 線照射により変化することがわ かった. Fig. 1に、 γ 線照射前後における重イオン誘起電 流ピーク値のゲート印加電圧(V_g)依存性を示す。結果より、 同一電圧印加状態における過渡電流ピーク値は、p-MOS において増加し、n-MOSでは減少することを確認した。な お、照射によるリーク電流増加量は1 pA以下であった。こ れは、 γ 線照射によるピーク電流値変化が、リーク電流増 加によるものでは無いことを意味している。一方、 γ 線照射 によりC-V特性が負方向にシフトすることがわかった。これ は、照射による酸化膜中での正電荷捕獲に起因する。照 射によるミッドギャップ電圧のシフト量(正電荷捕獲密度を 反映) ΔV_{MG} を評価した結果、p-MOSにおいて-15.2 V, nMOSにおいて-12.3 Vであった。このような場合、同一酸化 膜電界を得るには ΔV_{MG} を差し引いた分のゲート電圧を印 加する必要がある。そこで、実効的なゲート印加電圧を V_{g} - ΔV_{MG} と定義し、これを基準に照射前後での過渡電流ピー ク値を比較した結果(Fig. 1の横軸を V_{g} - ΔV_{MG} に変化)、照 射前後でのピーク値は精度良く一致することがわかった。

以上の結果より、y線照射によるMOS構造の重イオン 照射誘起電流の変化は、酸化膜中の固定正電荷密度の 増加による酸化膜電界変化に起因するものであり、y線照 射後においても、変位電流が支配的であることを明らかに した。



Fig. 1 Peak of heavy-ion induced gate current in pand n-MOS capacitors before and after gamma ray irradiation.

- T. Hirao et. al.:"Study of single-event current pulses induced in SOI diodes by collimated swift heavy-ions micro-beams," Nuclear Instruments and Methods in Physics Research B 206 (2003) 457-461.
- Y. Takahashi et. al.: "Heavy-ion induced current through an oxide layer," Nuclear Instruments and Methods in Physics Research B 260 (2007) 309-313.

1-04 Angular Dependence of the Single Event Transient Currents due to Nuclear Products in Semiconductors

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Products from nuclear reactions (secondaries) generated by neutrons and protons (nucleons) cause Single Event Effects (SEEs) on an integrated circuit and/or a power device. The basic mechanism of SEE triggered by nucleons is known as follows. When nucleons pass through silicon based devices, the secondaries, (He, C, N, O, F, Ne, Na, Mg, Al, Si, P) with energies up to several tens of MeV are generated through nuclear reactions including elastic scattering. The ionization by these secondaries induces a current on the terminal of semiconductors resulting in a Single Event Transient (SET) current. We have measured and analyzed the SET currents triggered by the normal incidence of an ion with energies up to tens of MeV¹). For secondaries from nuclear reaction, angles of the secondaries must be taken into account. In this study the angular dependence of transient currents induced by an angled ion are studied.

The device examined in this work was a commercial 1.5 GHz (at -3 dB) Si pin photodiode with a diameter of 450 μ m. The p-layer width was about 0.2 μ m, the depletion width was about 15 μ m at -10 V as estimated by the Capacitance-Voltage (C-V) and cross-sectional Transient Ion Beam Induced Current (TIBIC) methods. A bias was set to be -10 V for all measurements. The angles selected here were 0 (normal incidence), 30, 45, and 60 degrees, respectively. The ion species used in this study were Carbon (C), Oxygen (O), and Silicon (Si) with the energy of 15 MeV. The parameters charactering the SET, collected charge, peak current, fall time, and rise time were obtained from this series of the measurement as a function of the incident angle.

Figure 1 shows the typical SET currents when the angled Si ions pass through the sample. As shown, the SET currents are delayed when the angled ion strike. Figure 2 shows the normalized values of the peak current, the fall time, and the rise time as a function of $\cos^{-1}\theta$, where θ is the angle of the incident ion. All values are normalized by the values when the angle of ion incidence is 0 degree. Although not shown here it is found that the total collected charge is identical for all angles. It is well known that the collected charge depends on the inversed cosine if the range of ion is longer than the depletion layer thickness²⁾. On the other hand, the projected ranges of ions studied here are shorter than the depletion layer thickness. Therefore the total collected charge should be identical. While the angular dependence of charge collection was well studied, the angular dependences of the other parameters (the peak



Fig. 1 Transient currents induced in Si pin photodiodes at a bias of -10 V by 15 MeV-Si ion with the angles of 0, 30, 45, and 60, respectively. The transient signals are delayed when the incident angle increases.



Fig. 2 Normalized values of the peak current, the fall time, and the rise time as a function of $\cos^{-1}\theta$. All values are normalized by the values when the angle of ion incidence is 0 degree. The peak current slightly decreases, the fall time decreases, and the rise time increases with increasing angle for all ions.

current, the fall time, and the rise time) have been not yet studied. As shown in Fig. 2, it is found that these parameters are proportional to the inversed cosines. The result indicates the parameters of the SET current caused by nucleon would vary as a function of angle of secondaries.

- 1) S. Onoda et al., JAEA-Review 2007-060 (2008) 9.
- 2) H. Mori et al., Nucl. Instr. Meth. B 206 (2003) 31.

1-05 Effects of Fluence Rate on Radiation Degradation of Solar Cells

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The United States (NASA, NRL), Europe (ESA/ESTEC), and Japan (JAXA, JAEA) have been technically standardize collaborating to characterization and qualification test methodology for space solar cells. As a result of the collaboration, the international standard (ISO) of space solar cell irradiation test methods was published in 2005. However, no quantitative test condition is described in the standard because the effects of irradiation conditions on performance degradation of solar cells have not yet been fully understood. In this study, the effects of irradiation fluence rate on the degradation were investigated.

Two types of space solar cells, namely, InGaP/GaAs/Ge triple-junction (3J) cell and high-efficiency silicon (HES) solar cell were prepared and irradiated with 1 MeV electrons and 10 MeV protons under different fluence rates. The fluence rate was varied in two orders of magnitude. Five fluence rates for the proton irradiation and four rates for the electron irradiation were selected as described in Table I and II. The current-voltage characteristics of the cells before and after the irradiation tests were measured and compared.

rable i Experimental conditions of proton infaulatio	tal conditions of proton irradia	ation
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Fluence Rate	6.6	1.0	1.0	6.6	1.0	
$(p/cm^2/s)$	×10 ⁸	×10 ⁹	×10 ¹⁰	×10 ¹⁰	×10 ¹¹	
Energy (MeV)	10					
Fluence (p/cm ²)	5×10 ¹²					
Cell Size	$2 \text{ cm} \times 2 \text{ cm}$					

Tuble II Experimental conditions of election intadation.						
Fluence Rate	1.7	1.0	4.0	1.7		
$(e/cm^2/s)$	×10 ¹¹	×10 ¹²	×10 ¹²	×10 ¹³		
Energy (MeV)	1.0					
Fluence (e/cm ²)	1×10^{15}					
Cell Size	$2 \text{ cm} \times 2 \text{ cm}$					

Table II Experimental conditions of electron irradiation.

Figures 1 (a) and 1 (b) depict the radiation degradation of the short-circuit current (Isc), the open-circuit voltage (Voc) and the maximum power (Pmax) of the 3J and HES solar cells, respectively, due to 1 MeV-electron irradiation¹⁾. No significant difference was found in degradation in the 3J cell. However, for the HES cell, cells irradiated at higher fluence rates show greater degradation. On the other hand, in the case of 10 MeV proton irradiation, no significant difference was found in degradation of either the 3J cell or the HES cell.

It is known that the formation of radiation defects are different between the cases of electron irradiation (point defects) and proton one (cluster defects), and the type of radiation defect causing solar cell degradation in Si is different from that in III-V compounds. However, the reason of the different dependence of cell degradation on the fluence rate has not been clarified yet.

Meanwhile, the fact that degradation became greater as the electron fluence rate increased in the HES cells implies that a definition of the fluence rate is needed in the test methodology ISO standard for electron irradiation tests of Si cells.

Reference

 M. Saito et al., Technical Digest of 17th International Photovoltaic Science and Engineering Conference, Fukuoka, Japan (2007) 963.



(b) High Efficiency Silicon (HES) solar cell

Fig. 1 Comparison of remaining factors of I_{sc} , V_{oc} and P_{max} for (a) 3J and (b) HES cells irradiated with 1 MeV electrons.

1-06 Development of ERD System to Observe Hydrogen Behavior in Ion Irradiated a-Si:H

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Hydrogenated amorphous silicon (a-Si:H), which is used as thin film transistor or thin film solar cell, has high radiation tolerance^{1, 2)}. Since a-Si:H thin film solar cell also has advantages from the point of view of resource saving, high output per weight, low cost, and so on³⁾, it is expected as solar cell for space applications⁴⁾. In addition, thermal annealing at low temperatures (under 200 °C) restores the electrical characteristics of a-Si:H solar cell even if the degradation happens due to charged particle irradiation. Previously, the low conversion efficiency of a-Si:H sloar cells has been a problem for space use. However, the conversion efficiency of single-junction a-Si:H solar cell is nowadays about 10%⁵⁾. Furthermore, multi-junction solar cell, of which the efficiency is expected to be higher than that of single-junction solar cells, becomes a mainstream in present day. For instance, the target value of the efficiency of a-Si:H/a-SiGe:H/µc-Si:H triple-junction solar cell is 15%⁵⁾.

We plan to clarify the behavior of hydrogen atoms in a–Si:H (trap site of hydrogen atoms, the interaction between defects and hydrogen atoms, etc.) when a–Si:H is irradiated with ions. Under this plan, firstly, a–Si:H will be irradiated with H or Si ions under low temperature condition, and then ERD (Elastic Recoiled Detection) will be carried out at the low temperature. In this paper, we will show the detailed plan for this study, and report the current status of the system.

The samples used in this study are thin film a–Si:H grown on a Fz–Si substrate with $\sim 1 \mu m$ thick by PECVD (Plasma Enhanced Chemical Vapor Deposition). The samples are prepared at the Research Center for Photovoltaics, Advanced Industrial Science and Technology (AIST).

Figure 1 shows a schematic drawing of the planned experimental setup. Since this irradiation chamber connects both the 400kV Ion Implanter and the 3MV Single-Ended Accelerator, the sample can be irradiated with ion beams from both accelerators simultaneously. The sample holder will be equipped with a cryostat, a heater and a temperature controller to control sample temperature in a range from 10 to 800 K.

The planned experimental procedure is as follows. At first, the sample will be irradiated under low temperature conditions with 100 keV-H⁺ or 380 keV-Si⁺. The irradiation fluence will be selected several conditions between 10^{14} and 10^{17} /cm². By the former fluence, a change in the composition of the sample isn't expected, and the other hand, latter fluence can change the composition. Secondly, the

hydrogen distribution in the samples irradiated with ions will be measured by ERD at low temperatures. After that, the ERD measurement will be continuously conducted every $10 \sim 20$ K during increasing temperature. We can investigate the hydrogen reduction ratio from implanted region to the surface/deeper area. Comparing the ERD results with results obtained from other measurements such as Electron Spin Resonance (ESR) and Fourier Transform Infrared spectroscopy (FT-IR), the behaviors of hydrogen and defects acting as hydrogen trap sites in ion irradiated a–Si:H will be clarified.

So far, we have already installed the measurement system of ERD at room temperature. The hydrogen distribution in a crystalline Si sample irradiated with H ions was measured to confirm the performance of the ERD system.



Fig. 1 A schematic drawing of the experimental setup.

- P. Danesh, B. Pantchev, I. Savatinova, E. Liarokapis, S. Kaschieva, and A. G. Belov, Vacuum, 69 (2003) 79-82.
- N. Wyrsch, C. Miazza, S. Dunand, C. Ballif, A. Shah, M. Depeisse, D. Moraes, F. Powolnly, and P. Jarron, J. Non-Cryst. Solids, 352 (2006) 1797-1800.
- Srour JR, Vendura GJ, Lo DH, Toporow CMC, Dooley M, Nakano RP, King EE, IEEE. Trans. Nucl. Sci., 45 (1998) 2624-2631.
- K. Shimazaki, M. Imaizumi, T. Ohshima, H. Itoh, and K. Kibe, Proceedings of 7th International Workshop on Radiation Effects on Semiconductor Devices for Space Application (2006) 49-52.
- 3rd working paper of the Research Center for Photovoltaics, Advanced Industrial Science and Technology (2007).

1-07 Energy Transfer Efficiency and Proton Irradiation Effects of Eu Implanted III-Nitrides for Optoelectronic Devices in Radiation Environments

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Energy transfer efficiency and proton irradiation effects of Eu-implanted III-nitrides were investigated by photoluminescence (PL) measurement. Weak temperature dependence of energy transfer efficiency for Eu-related emission of $15\sim27\%$ was found in AlGaN:Eu. 380 keV proton irradiation effects on the PL intensity of GaN:Eu showed that Eurelated luminescence kept the initial PL intensity after the proton irradiation of 1×10^{14} cm⁻², where the near band edge emission decreased drastically. These results suggest that Eu-doped III-nitrides are strong candidate for optoelectronic devices in radiation environment.

光を用いた信号伝達は、短距離ではボード内配線 やチップ内配線、長距離では宇宙空間における衛星間 通信など様々な応用が考えられる。半導体発光デバイ スは、小型・軽量・低消費電力動作などの特長を有す るが、少数キャリアの再結合による発光デバイスは放 射線損傷の影響を強く受け、寿命、信頼性など解決す べき課題は多い。本プロジェクトでは、耐熱性・対放 射線に優れ、環境温度の変化による発光波長、発光効 率が影響を受けにくい希土類イオンを発光中心とする III族窒化物半導体の多数キャリアベースの発光デバイ ス実現の可能性を検討した。

有機金属気相成長法により成長したAlGaN層に対し、 原子力機構・高崎研のイオン注入装置を用いて、希土 類元素である Eu^+ を、100~350 keVのエネルギーで室 温注入した。イオン注入後、窒素抜けを防ぐためNH₃ を含むN₂雰囲気中にて、1100 °C、2 minのアニールを 行い注入損傷の回復を図った¹⁾。

作製した試料について、He-Cdレーザを光源とした ホトルミネセンス(PL)測定を行った。PLスペクトルに は、Euからの鋭く強いスペクトルの他、350 nm~ 450 nm付近にドナー・アクセプタ対発光や、窒化物半 導体でしばしば報告されるイエローバンド発光が観測 された。得られたスペクトルから、試料に吸収された 励起光のエネルギーのうち、Euイオンからの発光が 得られた割合、即ちエネルギー移送効率を推定し、そ の温度依存性を評価した。Eu注入量0.5×10¹⁵ cm⁻²およ び1×10¹⁵ cm⁻²試料に対する結果をFig. 1に示す。低温領 域において、エネルギー移送効率は約15%、および約



Fig. 1 Energy transfer efficiency for Eu in AlGaN.

27%が得られた。高いEu注入量で効率が小さいのは、 高濃度添加によりEu元素同士の近接し発光が低下す る濃度消光が生じたためと考えられる。移送効率の温 度依存性は、Eu添加量により若干の差異が見られる が、低温での移送効率が室温付近まで保持されており、 チップ温度が大きく変化する環境下でも安定に動作す る発光デバイスの発光層として有用であることを示唆 している。

次に、Eu添加GaNの室温のPL発光特性における 380 keVの陽子線照射に対する効果²⁾について検討した (Fig. 2)。GaNのバンド端発光強度は低陽子線照射量か ら減少しているのに対し、Eu添加試料のEuからの発 光は初期の発光強度を維持していることが分かる。 undoped GaNのバンド端発光強度の陽子線照射量依存 性の解析から、5×10¹⁵ cm⁻² の陽子線照射により発生 したトラップの濃度を4.5×10¹⁹ cm⁻³と見積もった。こ の濃度は、TRIMによるGaN中のEuの濃度10¹⁹~ 10²⁰ cm⁻³と同程度である。これまでの知見から、Euか らの発光強度は10²⁰ cm⁻³程度までのEu添加に対し単調 増加を示すことが分かっており、陽子線照射の影響は 高濃度のトラップ導入まで現れてこないと考えられる。 これは耐放射線性の優れた新しい発光デバイスへの高 いポテンシャルを有することを示している。

- 1) A. Wakahara, Optical Materials, 28 (2006) 731.
- H.Okada et al., Nucl. Instr. Meth. Phys. Res. B 266 (2008) 853.





1-08 Fabrication of n-Channel 6H-SiC MOSFETs with MGy Range Radiation Tolerance

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Silicon Carbide (SiC) is regarded as one of the candidate materials for devices with radiation hardness, owing to its superior radiation resistance as well as excellent physical properties and chemical stability¹⁾. However, since the fabrication process of SiC devices has not yet optimized, the electrical characteristics such as the channel mobility of SiC Metal-Oxide-Semiconductor Field Effect Transistors (MOSFETs) are still lower than the ideal values. In previous studies²⁾, it was reported that the electrical characteristics of SiC MOSFETs were improved by the refinement of fabrication processes. Besides, Ohshima et al.¹⁾ reported that the channel mobility for 6H-SiC MOSFETs with steam-annealed gate oxide did not degrade by gamma-ray irradiation at 200 kGy in spite that H2-annealed ones showed the degradation above 40 kGy. This indicates that the radiation response of MOSFETs is affected by their fabrication processes. Therefore, it is important to reveal the relationship between radiation hardness and fabrication processes.

In this study, we fabricate n-channel 6H-SiC MOSFETs with different fabrication processes, and radiation resistance of these MOSFETs is compared.

The n-channel MOSFETs were fabricated on p-type 6H-SiC epitaxial layers. The net acceptor concentration of the epitaxial films was 2.3×10^{15} /cm³. The source and drain of the MOSFETs were formed using phosphorus (P) ion implantation at 800 °C and subsequent annealing at 1650 °C for 3 min in an Ar atmosphere. Three-fold implantation (60, 90, 140 keV) was carried out to form a box profile of P at a mean concentration of 5×10¹⁹ /cm³. To avoid the degradation of the surface morphology, the surface of samples was covered with carbon films during the annealing. After the annealing, the carbon films were removed by the oxidation at 800 °C for 30 min in dry oxygen. For the remainder of this article, SiC MOSFETs using the carbon coating procedure is referred to as "carbon-coated MOSFETs". For comparison, MOSFETs without this carbon coating procedure were also fabricated (non-coated). The gate oxide was fabricated by pyrogenic oxidation $(H_2:O_2 = 1:1 \text{ at } 1100 \text{ }^{\circ}\text{C} \text{ for } 30 \text{ min})$. The gate length, width and thickness of the MOSFETs are 10 µm, 200 µm and 30 nm, respectively. After gate oxidation, the samples were slowly cooled at a rate of ≈ 8 °C/min until 500 °C in Ar atmosphere. Al electrodes were formed using a lift-off method. Gamma-ray irradiation was performed up to about 3 MGy (SiO₂) at a rate of 8.7 kGy/hour at room temperature (RT). No electrical bias was applied to the gate, the drain and the source during irradiation. The electrical

characteristics were measured at RT in a shielded probe station under dark conditions to minimize external noises.

Figure 1 shows the channel mobility for n-channel 6H-SiC MOSFETs as a function of gamma-ray absorbed dose. Before gamma-ray irradiation, the values of channel mobility for carbon-coated and non-coated MOSFETs are 41 and 44 cm²/Vs, respectively. For carbon-coated MOSFETs, no significant change or slight increase in the channel mobility is observed up to 3 MGy. The value of gamma-ray induced interface traps (ΔN_{it}) for carbon-coated MOSFETs is estimated to be less than 4×10^{11} /cm², which are close to the detection limit even after 3 MGy irradiation. On the other hand, a decrease in the channel mobility is observed for non-coated MOSFETs at dose ranges above 2 MGy. The ΔN_{it} increases with increasing dose, and the value becomes in the order of 1012/cm2 at doses above 1.5 MGy. Since the channel mobility decreases due to the generation of interface traps³⁾, the result obtained in this study can be interpreted in terms of the generation of interface traps due to irradiation.

- 1) T. Ohshima et al.: Mater. Sci. Forum 389-393 (2002) 1093.
- H. Yano, et al.: Mater. Sci. Forum 388-342 (2000) 1109.
- 3) T. Ohshima et al.: J. Appl. Phys. 90 (2001) 3038.



Fig. 1 Channel mobility for n-channel 6H-SiC MOSFETs as a function of gamma-ray absorbed dose. Squares and circles represent the results obtained for carbon-coated and non-coated samples, respectively.

1-09 First-Principles Molecular Dynamics Simulation of SiO₂/SiC Interface of SiC Devices

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Silicon carbide (SiC) semiconductor devices are expected to be used in severe environments such as outer space and/or nuclear power plants. The performance of SiC metal-oxide- semiconductor (MOS) devices to date is low as compared with theoretical performance expected. This is considered to be attributed to defects generated at the SiO₂/SiC interface that degrade the electrical characteristics of MOS devices. However, it is not clear at present what kind of defects affect the degradation of electric characteristics. Although a lot of researchers have been done for analyzing the relation between physical structure near the interface and electrical characteristics in actual MOS devices, the details are not clear at present.

Last year, we obtained the SiO_2 layers with an amorphous characteristics¹⁾ using 444 atoms middle-scale model. However, the defect at the interface was not induced, though it was observed in the actual devices. The aim of this report is to produce the theoretical interface involving the defects at the interface and to know the relation between physical structure and electrical characteristics at the interface by using large-scale simulation.

A simulation for the amorphous SiO₂(a-SiO₂)/SiC interface in the actual SiC MOS devices was carried out on computer using the first-principles molecular dynamics calculation, in which Vienna ab initio Simulation Package (VASP)²⁾ was employed as a calculation code. Prior to the calculation, a large-scale slab model using 1017 atoms was prepared on the basis of an atomic network for beta quartz $(\beta$ -SiO₂) on 4H-SiC crystal. The Si atoms at the top of β -SiO₂ and the C atoms at the bottom of 4H-SiC crystal were saturated by H atoms. By applying the heating and quenching method to the large-scale slab model, a theoretical structure around the a-SiO₂/SiC interface was obtained on computer. Figure 1 exhibits the temperature profiles of the heating and quenching method for the simulation. Three different types of temperature profiles are applied. The total energy of large-scale model is also plotted in the same figure.





The simulation was done for several months to produce the theoretical structure on computer. In order to estimate the distribution of the SiO₂ layers of theoretical structure obtained, radial pair distribution function (RDF) was applied. No long range order was seen for the theoretical structure. On the contrary, the short range order was observed. As seen in Fig. 2, the average bond angle of Si-O-Si increases from 135 deg to 140 deg with the decrease of quenching rate from -2000 K/ps to -500 K/ps. It was found that the average bond angle for the quenching rate of -500 K/ps is close to that of silica glass (145 ± 10 deg). Similarly, average the O-Si-O bond angle and the Si-O bond length in the SiO₂ layers is calculated to be 0.165 nm and 109 deg, respectively.

As for the theoretical structure at the interface, the several kinds of defects such as a Si dangling bond, Si-Si bonds and a Si atom with the coordination number of 5 are produced at the interface. It is not explained well with reasons that these defects are generated at the interface of theoretical structure during the heating and quenching procedures. However the composition ratio of O to Si (O/Si ratio) at the interface is high enough as compared with that in the SiO₂ layers, which indicates following thing. As compared with Si in SiO₂ layers, O links easily with the exposed bonds of Si atoms at the interface involving 4H-SiC substrate during the process. Therefore these defects, especially for the Si-Si bond or Si dangling bond, would reduce the excess oxygen at the interface and improve the O/Si ratio to a normal value of 2. As for the behavior of a Si atom with the coordination number of 5, the structure disappeared when the quenching rate slow down from -2000 K/ps to -500 K/ps. Taking the angle extension of Si-O-Si bond into account, the Si atom with the coordination number of 5 is considered to be unstable as compared with other defects. The slower quenching rate is needed to obtain the result.

- A.Miyashita et al., Materials Science Forum 556-557, (2007) 521.
- 2) G.Kresse et al., Phys. Rev. B54 (1996) 11169.



Distance between Si atoms in SiO₂ layers (nm)

1-10 Mechanisms of Reduction in Hole Concentration in Al-implanted p-type 6H-SiC by 1 MeV Electron Irradiation

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The relationship between the temperature-dependent hole concentration p(T) and trap density can be directly investigated, when the densities and energy levels of traps can be determined using p(T). A graphical peak analysis method (FCCS: free carrier concentration spectroscopy)^{1,2)} can determine the densities and energy levels of acceptors and hole traps from the experimental p(T) without any assumptions regarding acceptor species and hole traps.

By comparing the radiation damage in 4H-SiC with that in Si,^{1,2)} it was found that the reduction in p(T) in Al-doped p-type 4H-SiC by electron irradiation was much larger than that in p-type Si. Since the large reduction in p(T) in Al-doped 4H-SiC by electron irradiation was an unexpected result, the decrease in p(T) by irradiation with different electron energies was investigated. Since 6H-SiC is expected to be a suitable polytype for electronic devices used in high-energy radiation environments, we report on the reduction in p(T) in Al-implanted p-type 6H-SiC by 1 MeV electron irradiation.

In order to form a p-type 6H-SiC layer, Al ions were implanted at 800 °C in a 4.9- μ m-thick n-type 6H-SiC epilayer with a N-doping level of 6.7×10^{15} cm⁻³ on an n⁺-type 6H-SiC substrate. To obtain a 0.5- μ m-thick box profile of Al concentration, fivefold Al ion implantation was carried out using 20, 50, 110, 200, and 340 keV onto the n-type SiC epilayer surface. The samples were irradiated



Fig. 1 Temperature dependence of hole concentration for Al-implanted 6H-SiC unirradiated or irradiated by 1 MeV electrons. The squares, triangles, circles, and diamonds represent p(T) for 0, 1×10^{16} , 5×10^{16} , and 1×10^{17} cm⁻², respectively.

with 1 MeV electrons. p(T) was measured in the temperature range from 210 to 720 K and at a magnetic field of 1.4 T.

Figure 1 shows p(T) for the unirradiated and irradiated p-type 6H-SiC. Using each p(T) in Fig. 1, the density (N_A) and energy level (E_A) of acceptors were determined by FCCS. N_A is decreased by the displacement of Al atoms or their nearest neighbor C atoms because only the Al atom bonding to four C atoms works as an Al acceptor. Since the number of collisions between incident electrons and those atoms in unit volume is proportional to N_A , the following differential equation describing the fluence dependence of N_A is obtained:

$$\frac{\mathrm{d}N_{\mathrm{A}}}{\mathrm{d}\Phi} = -\kappa_{A}N_{\mathrm{A}}$$

where κ_A is the removal coefficient of Al acceptors for 1 MeV electron irradiation and Φ is the fluence of 1 MeV electrons. Therefore,

$$N_{\rm A} = N_{\rm A0} \exp(-\kappa_{\rm A} \Phi)$$

Figure 2 shows the experimental fluence dependence of N_A , from whose slope κ_A is determined as 6.4×10^{-18} cm².

- 1) H. Matsuura et al.: Jpn. J. Appl. Phys. 45 (2006) 2648.
- 2) H. Matsuura *et al*.: Physica B **376-377** (2006) 34.



Fig. 2 Fluence dependence of acceptor density. The circles and the solid straight line represent N_A obtained by FCCS and the fit obtained by the least-square method, respectively.

1-11 EPR Characterization of Defects in SiC

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Silicon carbide (SiC) is suitable for high-power, high-current. high-frequency and high-temperature applications. Moreover, the high radiation-hardness is especially suited to the semiconductor devices used in space. Since point defects affect the electronic properties of SiC significantly even at low concentrations, it is important to identify the radiation-induced defects and to clarify the mechanism of their formation. By using electron paramagnetic resonance (EPR) technique, we have determined the detailed geometric and electronic structures of monovacancies, $V_{\text{Si}}(h/k)$, T_{V2a} , EI5: $V_{\text{C}}^{+}(k)$, EI6: $V_{\text{C}}^{+}(h)$ and HEI1: $V_{\rm C}^{-}(h)$, divacancies, P6/P7: $[V_{\rm Si}V_{\rm C}]^0$ (S=1), and antisite-vacancy pairs, HEI9/HEI10: $[C_{Si}V_C]^+$ and SI5: $[C_{si}V_C]^{-}$, in 4H-SiC.¹⁾ Here, we report the recent progress of our continuing work of the EPR studies of defects in SiC.

The determination of the energy level positions in the energy gap plays a key role for the EPR centers to be linked to PL and DLTS. We have carried out *photo*-EPR, in which the intensity (appearance) of EPR signal is measured as a function of the photon energy of illuminating light, of the vacancy-related defects in *n*-type 4*H*-SiC irradiated with electrons at various temperatures (the example of 350 °C irradiation is shown in Fig. 1).²⁾ Comparing to the *first principles* calculations, the threshold energy (1.05-1.10 eV) observed for $V_{\rm Si}^-$ and $T_{\rm V2a}$ is ascribed to the intra-vacancy transition energy. The detection of $V_{\rm C}^-$ from 1.00-1.05 eV and $V_{\rm C}^+$ from 2.00-2.05 eV confirms the negative-*U* model of $V_{\rm C}^{2-}$ which is stable when $E_{\rm F}=E_{\rm c}$ -1.0-1.1 eV.

Ion implantation, which enables the spatial selectivity and the desired concentration of doping, is critical for the device fabrication. The presence of the residual defects after post-implantation annealing (1650 °C) has been observed in phosphorus ion implanted 6H-SiC.1) Theoretical studies have suggested that complex formation of carbon interstitials or their aggregation to clusters of carbon antisites should be one of possible candidates for the defects which are stable up to high temperatures.^{3,4}) To identify the isolated interstitial (C_i and Si_i) and to clarify the mechanism of the formation of defects through the migration of interstitials, electron irradiations at various temperatures have been carried out. After electron irradiation at 100 K (2 MeV, 5×10^{17} cm⁻²), we have observed a new EPR center labeled LE1 (C_{2v} , S=1/2) which is likely to be arising from interstitial carbon (Fig. 2). EPR spectra HEI5/HEI6 observed in 4H-SiC after electron irradiation at 350-800 °C

have been identified to be di-carbon antisite defect $(C_2)_{Si}$ (two carbon atoms sharing a single silicon lattice site), which is formed by capture of mobile interstitial carbon by pre-existing carbon antisite, from the ${}^{13}C/{}^{29}Si$ hyperfine interactions.

- 1) J. Isoya *et al.*, phys. stat. solidi b, special issue, (in press).
- 2) T. Umeda *et al.*, Proceedings of ICSCRM2007, (in press).
- 3) A Gali et al., Phys. Rev. B 68, (2003) 125201.
- 4) A. Mattausch et al., Phys Rev. B 70, (2004) 235211.



Fig. 1 *Photo*-EPR (60 K) of vacancy-related defects in n-4H-SiC irradiated with electrons (1×10¹⁸ cm⁻²) at 350 °C.



Fig. 2 EPR spectrum of 3C-SiC irradiated with electrons $(5 \times 10^{17} \text{ cm}^{-2})$ at 100 K.

1-12 Study of the Radiation Resistant Characteristics for the **Candidate Materials and Instruments in the J-PARC**

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Introduction

The several irradiation experiments were made to contribute the construction of J-PARC facility by the evaluation of material reliability. Major activity of the irradiation experiment is a study for the characteristics of cable at the high dose rate environment. Other activities are examination of paint and linear motion guide assembly. Result of each study is referred in the specification of those.

PEEK insulation high voltage power cable

Use: DC 7 kV power supply cable for the ion pump installed in the 3 GeV synchrotron tunnel.

Use position and assumption dose: Right under the accelerator beam line and dose of up to 10 MGy.

Feature: The structure consists of PEEK (poly-etherether-ketone) resin insulator, copper conductor and stainless steel mesh armored power cable.

Irradiation condition: Sample dimension of 10 mm in outer diameter and 10 m long winding by race-track shape. Dose rate of 14 kGy/h average and total exposure of up to 30 MGy.

Summarized result: Insulation resistance has no change up to Total dose of 30 MGy turned mechanical 30 MGy. property deteriorative as the fragileness.

Halogen-free and flame retardant conventional power cable

Use: Ordinary power feed cable for electric device.

Use position and assumption dose: Between power supply building and active components in the accelerator tunnel and dose of up to 3 MGy.

Feature: The structure consists of modified low-density polyethylene resin insulator, copper conductor and the new sheath material of polyolefin resin to which a radiation resistant stabilizer was added.

Irradiation condition: Sample dimension of 16 mm in outer diameter and 40 m long winding by race-track shape for a pair of sample. Two kinds of dose rate at 2 kGy/h and 5.4 kGy/h were used for total exposure of 1 MGy and 2.6 MGy respectively.

Summarized result: The theme of the examination was the flame retardant characteristics. Both of 1 MGy and 2.6 MGy irradiated sample passed the vertical flame test of IEEE-383 regulation.

PEEK insulation coaxial RF cable

Use: Data communication cable for the control and monitoring for the accelerator at the high dose rate area.

Use position and assumption dose: Along the accelerator beam line to connect as pre-amp or monitor assembly, and dose of up to 30 MGy.

Feature: The structure consists of PEEK resin insulator, copper conductor and polyolefin sheath kept a coaxial dimension.

Irradiation condition: Sample dimension of 6 mm in outer diameter and 0.5 m in length terminated by SMA coaxial connector. Dose rate of 10 kGy/h average and total exposure of up to 30 MGy.

Summarized result: Major electrical characteristics such as RF decay, delay, phase-shift, VSWR, impedance, capacitance and insulation resistance had no change until dose of 30 MGy. Total dose of more than 20 MGy turned mechanical property deteriorative as the fragileness.

Room temperature available galvanizing paints

Use: Repair paint of the instruments surface coated with zinc at the high dose rate area.

Use position and assumption dose: Around the beam dump component of the accelerator and dose of up to 50 MGy.

Feature: Handy galvanizing paints by spray coating method available on the market.

Irradiation condition: Galvanizing coated iron plate sample and zinc plated iron plate sample were used. Dose rate of 15 kGy/h average and total exposure of up to 50 MGy.

Summarized result: No change of the surface condition until dose of 30 MGy was observed. Slight powder is found at dose of 40 MGy range. Paint adhesion of level 2 was kept on the JIS- K5600 cross cut test after 50 MGy exposure.

Linear motion guide using the Rice-Bran-Ceramic slider

Use: A precise position guide in a mechanical linear motion drive apparatus applied to the accelerator active component at the high dose rate area.

Use position and assumption dose: In the beam monitor drive mechanism typically and varied dose of 1 MGy to 50 MGv.

Feature: Combined with SUS-304 raceway and oil free sliding carriage using the rice-bran-ceramic.

Irradiation condition: Three pieces of assembly with size of 100mm in raceway length were irradiated.

Summarized result: No change of the friction coefficient was observed until dose of 50 MGy. The friction coefficient was kept around 0.15 µm through the whole examination. No damage in the ceramic structure was observed by scanning electron microscopy after 50 MGy exposure.

Acknowledgement

The author appreciates the collaboration companies of Fujikura Ltd., Mitsubishi Cable Industries Ltd., IHI Corporation and PREFACT Co. Ltd.

1-13 Gamma-Ray Irradiation Test to Investigate Acceptable Total Dose of Rad-Hard Operational Amplifier for Remote Maintenance System of ITER

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The ITER, an international fusion experimental reactor, requires remote maintenance for its components because of the severe environment in the reactor, especially the gamma-ray. The radiation environment during the maintenance operation is about 0.5 kGy/h and therefore, the total dose is 1 MGy if the maintenance operation takes three months.

The irradiation test has been performed for components of the ITER remote maintenance system^{1, 2)}. Regarding instrumentation and operational amplifiers for strain gauges, an irradiation test was performed in order to examine which amplifier on the market can be used from the viewpoint of radiation-hardness. It was found from the test that a operational rad-hard amplifier, HS1-5104ARH-Q, functioned without deviation of characteristics from specification until the guaranteed value, 1 kGy, and had continued to function from 2 kGy till 417 kGy with some of characteristics deviating from specification³⁾. The present study aimed at continuation of the irradiation test to investigate the acceptable total dose of the amplifier.

The dose rate in the irradiation test was almost the same value as that in the real environment of the ITER maintenance operation, 0.5 kGy/h. The samples were irradiated for a certain period with bias current and after that they were took out from the cell for the measurement of their electrical characteristics such as input bias current, offset voltage, open-loop gain and so on. This cycle of irradiation and measurement was repeated.

The identical samples used in the previous study (No. 5538, 5539 and 5540) were further irradiated with additional dose of 595 kGy (1.012 MGy in total) and the results were compared with the reference sample which had not been



Fig. 1 Open-loop gain Avol+.

irradiated. All the irradiated samples had functioned until total dose of 1.012 MGy, and some of characteristics were extremely deviating from specification. Figures 1 and 2 show the transitions of the open-loop gains A_{vol} + and A_{vol} - of HS1-5104ARH-Q during irradiation. Definitions of A_{vol} + and A_{vol} - were as follows:

$$\begin{split} A_{vol} + &= (10V\text{-}0V) / (V_{in_10V}\text{-}V_{in_0V}) \\ A_{vol}\text{-} &= (-10V\text{-}0V) / (V_{in_10V}\text{-}V_{in_0V}) \end{split}$$

where

 $V_{in_XV}\!\!:$ input voltage of the amplifier where output voltage is X (V)

The broken and dotted lines in Figs. 1 and 2 show the reference sample and specification, respectively. Regarding the A_{vol} +, the deviation from specification is generally limited even at the end of irradiation. On the other hand, the A_{vol} - decreased drastically after the total dose of 622 kGy although they still remain at around 70 kV/V until 471 kGy. Therefore, the acceptable total dose can be defined as 471 kGy for the amplifier HS1-5104ARH-Q. The reason for the decrement was considered that the transistor in the amplifier was damaged due to irradiation and its gain decreased.

In the future study, an actual circuit for the strain gauge using the amplifier will be irradiated and the behavior of the circuit will be investigated.

- 1) K. Obara et al., JAERI-Tech 99-003 (1999).
- K. Obara et al., J. Robot. Mechatron. Vol. 10, No. 2 (1998) 121.
- 3) N. Takeda et al., JAEA-Review 2007-060 (2007) 18.



Fig. 2 Open-loop gain Avol-.

1-14 Performance Test of AC Servo Motor for ITER Blanket Remote Robot under High Gamma Ray Radiation

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The inner lining (termed a "blanket") of the vacuum vessel (VV) of the International Thermonuclear Experimental Reactor (ITER) will be activated by 14 MeV neutrons generated by fusion reactions. Maintenance of the blanket in the VV must be carried out under high gamma ray radiation conditions (~0.5 kGy/h) and in the presence of contaminated dust such as beryllium, carbon and steel. All interventions inside the VV therefore must be performed remotely. The blanket is composed of about 400 modules, designed for remote individual replacement¹.

Figure 1 shows the robot for maintenance of the blanket module. The robot has 18 degrees of freedom and is driven by an electrical AC servo motor²⁾ with sensor for position measurement. Irradiation tests for AC servo motor were carried out from 1995 to 2001 with the cooperation of JAEA OuBS to develop the electrical insulator and the mechanical lubricant under high gamma ray irradiation³⁾. However, the AC servo motor that was developed is too expensive. It is necessary to reduce the manufacturing cost of this developed AC servo motor in order to satisfy ITER requirements. To reduce costs, a commercially available AC servo motor used in viewing/inspection robots that are designed for fission reactor was selected. The Objective of this irradiation test is to clarify the performance limits of a commercially available AC servo motor under gamma ray

Blanket Robot for remote maintenance Rail support

Fig.1 Robot for remote maintenance for ITER blanket

Table 1 Materials AC servo motor

Parts for AC servo motor	Materials
 Insulator of winding for motor stator Grease for lubricant Cable for power and sensor 	Varnish (Polyester) Rad-hard grease(GK-1) Insulator: Ethylene Tetra Fluc
- Connection for lead line	Sensor: Enlorosuironated polyethylene Sensor: polylefin Motor: Ethylene Tetra Fluoro

irradiation. As a result of these performance tests, we will determine the criteria for estimating the life time of AC servo motor.

Table 1 shows the materials for AC servo motor used in these irradiation tests. Six AC servo motors were subjected to gamma ray radiation at a dose rate of 0.5 kG/h. Intermediate results of the tests indicate that one of six motors has stopped due to radiation damage of electrical insulator in sensor as shown in Fig. 2. The total dose of the non-functioning motor is about 3.47 MGy. The remained five motors continue to work after having been irradiated with a total dose of about 6.3 MGy. Test will continue until the performance limits of the remaining five motors have been established.

- 1) ITER Technical Basis, ITER EDA Documentation Series No. 24, IAEA, Vienna, (2002).
- H. Sugimoto, et al., Design and theoretical concept of AC servo motor system for practice, Sougou- denshi shuppann, (2005) (in Japanese).
- 3) K. Obara, et al., HIGH GAMMA-RAYS IRRADIATION TESTS OF CRITICAL COMPONENTS FOR ITER IN-VESSEL REMOTE HANDLING SYSTEM, JAERI-Tech, 99-003, (1999).



Fig.2 Sensor for position measurement after about 6940hr (total dose : 3.47 MGy)

1-15 Investigation of Effect of Radiation Deterioration on Confinement Capability of Glove-box

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In the mixed-oxide (MOX) fuel fabrication facility, MOX is required to be handled in Glove-box (GB) to sustain confinement of MOX. Since the GB consists in some inflammable components such as plastic panel, in case of fire, the static containment capability of the GB may be deteriorated by pyrolysis or combustion of the components. Deterioration of the static confinement capability of the GB may cause release of MOX to the room involving the GB and to the outside of the facility and exposure of worker and public around the facility. Therefore, to confirm the safety of the facility under the fire, acquisition of fundamental data of the pyrolysis and combustion properties of the components and establishment of quantitative evaluation method of time-course of the deterioration are very important. From this point of view, in JAEA, the thermal properties of the components have been investigated as a contract research from Japan Nuclear Energy Safety Organization (JNES)¹⁾.

Since a large amount of MOX is handled in the actual MOX fuel fabrication process, the components are irradiated by MOX and radiation deterioration of the components could be caused. In this contract research, the effect of the radiation deterioration on the pyrolysis properties of the components has been also investigated. In fiscal 2006, some components were irradiated under an extremely high-dose condition at 880 kGy, using the 6th ⁶⁰Co cell at JAEA Takasaki and the exothermic and endothermic calorific values and mass reduction with pyrolysis reactions as a function of temperature of the irradiated components were measured by Simultaneous Differential Thermogravimetric Analyzer (Shimazu DTG-60H)²⁾³⁾. In fiscal 2007, to observe the effect in detail, the components were irradiated under two irradiation conditions, 0.5-3 kGy as relative low irradiation conditions and 25-880 kGy as relative high irradiation conditions, and the pyrolysis properties of the components were observed by using the DTG apparatus.

Figure 1 and 2 show the mass reduction rates with pyrolysis reactions under air atmosphere condition for the irradiated Polymethylmethacrylate (PMMA) as commonly used as plastic panel of the GB. The mass reduction rate was evaluated by differentiating mass reduction curve, obtained from DTG measurement, with respect to time and dividing by initial sample weight, used in DTG measurement. For the relative low irradiation conditions, the effect of the radiation deterioration on the pyrolysis properties was not recognized. This means the effect for PMMA, used as the panel for a long period in the actual facility, would be negligible. On the other hand, for the

relative high irradiation conditions, one peak of the mass reduction rate, observed for non-irradiation condition, split up into two peaks and temperatures of the respective two peaks shifted to lower and higher temperatures with increasing dose, respectively. The long-chain polymer, such as PMMA, is affected as following two conflicting effects by irradiation, the one is reduction of molecular weight by cutting of backbone chain and the other is rise of that by binding between the molecules and forming three dimensional network structure. Because of these phenomena, the split of the peak of the mass reduction rate would be induced. In future, the more detailed investigation, for example, observation of threshold of dose affecting the pyrolysis properties, is planned.

- 1) H. Abe et al., JAEA-Research 2006-054 (2006.9). [In Japanese]
- 2) H. Abe et al., JAEA-Research 2007-075 (2007.11). [In Japanese]
- H. Abe et al., JAEA-Review 2007-060 (2008.3). [In Japanese]



Fig. 1 Evaluation results of mass reduction rate (relative low irradiation conditions).



Fig. 2 Evaluation results of mass reduction rate (relative high irradiation conditions).

1-16 Direct Alpha-Ray Irradiation Damage on CSM Rubber Applied for Glove Box for Plutonium Powder Treatment

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Facilities as a MOX plant which treats much plutonium consume a lot of expensive rubber gloves applied for glove boxes. Currently, for economical reasons, newly improved long-lasting glove is desired and under development at Plutonium Fuel Development Center, Tokai, JAEA. In order to map out its course of development of the new glove, the deteriorative factors (oxygen, ozone, ionizing radiation etc.) and the mechanism of damage to raw material are needed to be well-known. However, the present status of experimental data of direct and indirect α -ray effects on glove is very poor notwithstanding that plutonium is strong α -ray emitter, and its systematic evaluation has been scarcely conducted hitherto. Therefore, in order to evaluate the direct α -ray effects for the first step, helium ion irradiation experiments were performed at vacuum environment.

Sample material is chlorosulfonated polyethylene (CSM) glove which SANKO CHEMICAL INDUSTRY CO. Ltd. manufactures and Plutonium Fuel Development Center mainly employs. JIS K 6251-7 dumbbell-shaped pieces cut from a CSM glove were irradiated in a vacuum chamber $(4 \sim 7 \times 10^{-7} \text{ Torr})$ with He²⁺ ion beam (26~180 nA, duration of 4~6104 sec.) generated from 3MV tandem accelerator at TIARA. The kinetic energy of the ion was 5 MeV close to the average energy of a rays emitted from plutonium isotopes. Experiments were divided into two parts, low-fluence experiment and high-fluence one, which correspond to 10 kBq/cm^2 contamination and 0.5~5 years exposure, and 2.9 MBq/cm² and 0.2~10 years, respectively. The maximum was 4.7×10^{14} cm⁻² as high fluence. Since the deterioration-promoting effect under stress by γ -ray irradiation is known, two stress parameters on sample were additionally set as "without-stress" and "elongated" for 100%. Four samples were simultaneously irradiated for one condition: fluence and stress. The irradiated samples are sent to visual inspection and two tensile tests: tensile strength and elongation at break.

At the low-fluence experiment up to 9.2×10^{11} cm⁻², α -ray effects were not observed at all for not only surface



Fig. 1 Photographs of without-stress samples at high fluence experiment. (a) After irradiation (b) Enlarged side view of surface after irradiation (c) After tensile test. Numerical value in photograph indicates ion fluence into the sample in the unit of cm⁻².

state but tensile properties.

Figure 1 is the photographs of the irradiated without-stress samples. Figure 1(a) shows remarkable gradation according to fluence. Figure 1(b) shows the discolored and well-defined layer of $30{\sim}40 \ \mu m$ depth corresponding to the ion range. The surface of samples begin to hardening owing to progressed vulcanization, and the samples irradiated up to the fluence of $4.7 \times 10^{14} \ cm^{-2}$ are surface-fragmented, as shown in Fig. 1(c).

Figure 2 shows the results of tensile strength tests. There is no systematic variation at low-fluence experiment. A tendency to decrease with fluence is shown in appearance at high-fluence region. However, the weakening is likely to be attributed principally to temperature increase due to thermal flux. This supposition is drawn from followings; 1) the ion flux proportional to the thermal flux at the irradiation indicated by symbol A was 1.5~2.0 times higher than that of other high fluence irradiation, 2) though the fluence at the irradiation indicated by B and C symbols differs by a factor of ten, there is only a slight difference in strength. Therefore, irradiation damage on tensile strength may be relatively small. Elongated samples exhibit a faster decrease than without-stress ones. This difference may be due to the deterioration-promoting effect under stress by heat load, not by irradiation.

Since there are no changes in the result of elongation at break under any conditions, no correlation between elongation and tensile strength is given.

At the high-fluence experiment, rubber surface was degraded such as discoloration, hardening and embrittlement. Tensile properties variation which was supposed to be ascribed to heat load must be confirmed with additional experiments in radiation-free condition.



Fig. 2 Tensile strength according to ion fluence. Values are the medians of four samples for one irradiation condition. Non-irradiated data are set at 2×10^{10} cm⁻². Tensile tests for high-fluence and low-fluence samples were performed at temperatures of $17 \sim 18$ °C and ~ 29 °C, respectively.
1-17 Gamma-Ray Irradiation Durability of Silica Based Adsorbents for the Extraction Chromatography

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In the framework of the feasibility study on commercialized FR cycle system, Japan Atomic Energy Agency (JAEA) evaluated Am and Cm (An(III)) recovery processes on design basis, and the extraction chromatography technique, which uses SiO_2 support coated with styrene-divinylbenzene polymer (SiO_2 -P) and an extractant on its surface, was evaluated as the promising process for establishing more compact equipments and a less amount of secondary waste than those of the solvent extraction methods^{1, 2)}.

In this study, resistances of the R-BTP/SiO₂-P and the TODGA/SiO₂-P adsorbents to gamma-ray irradiation were investigated to evaluate durability of the adsorbents. Each R-BTP/SiO₂-P and TODGA/SiO₂-P adsorbent was kept in HNO₃ solution, and they were irradiated at room temperature with Co-60 gamma-rays at a dose rate of 3 kGy/h. The maximum accumulated dose was 4 MGy. The adsorbents separated from the acid solution were provided for the batch adsorption/elution experiments, and the concentrations of the TOC in the residual acidic solution were evaluated to estimate the amount of the eluted organic compounds.

Gases were generated in the vial that contained the adsorbents and HNO_3 solution during gamma-ray irradiation, and colors of the adsorbents turned form white to yellow or brown by irradiation.

Figure 1 shows the distribution coefficients of U(IV) onto the R-BTP/SiO₂-P adsorbent irradiated with the gamma-ray. The distribution coefficient decreased with absorbed dose, and degradation by irradiation was apparently observed through the change of appearance of the adsorbent. Concentration of the HNO₃ also influenced on the distribution coefficient. The adsorbents immersed in the 2 mol/dm³ HNO₃ solutions lost almost all the adsorption capacity by irradiation, however the adsorbents mixed with



Fig. 1 Distribution coefficient of U(IV) onto the irradiated R-BTP adsorbents.

0.01 mol/dm³ HNO₃ solution kept above 80% of the initial capacity up to 1 MGy.

The amount of the eluted R-BTP evaluated by TOC in the residual HNO₃ solution was 40% and 0.1% for 1 MGy irradiated adsorbents in the 2 and 0.01 mol/dm³ HNO₃ solution, respectively. Some carbon in R-BTP should have been emitted as CO_2 gas during the irradiation.

Those results imply that the degradation of the R-BTP mainly depends on the concentration of the HNO₃ solution rather than the absorbed dose of gamma-ray.

Figure 2 shows the distribution coefficients of Eu(III) onto the TODGA adsorbents irradiated by gamma-ray. The distribution coefficient decreased with dose, however the decrease scarcely depended on the concentration of the HNO₃ solution. The amount of the eluted TODGA was 11 % and 1 % for 1 MGy irradiated adsorbents in the 4.76 and 0.1 mol/dm³ HNO₃ solution, respectively. Therefore degradation of TODGA by gamma-ray irradiation is considered to be less significant than that of R-BTP.

Present study is the result of "Development of Extraction Chromatography for Am and Cm Recovery" entrusted to JAEA by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

Acknowledgements

We would like to thank Mr. R. Yamagata at Irradiation Service Section in TARRI for his assistance on the experiment.

- Y. Koma, T. Takata, K. Sato, Proceedings of GLOBAL 2003 Atoms for Prosperity: Updating Eisenhower's Global Vision for Nuclear Energy, New Orleans, USA (2003).
- H. Funasaka, M. Itoh, Proceedings of GLOBAL 2007, Advanced Nuclear Fuels and Cycles, Idaho, USA (2007).



Fig. 2 Distribution coefficient of Eu(III) onto the irradiated TODGA adsorbents.

1-18 Study on Stability of Cs·Sr Solvent Impregnated Resin against Gamma Irradiation

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Minimization of radioactive waste from reprocessing process of spent nuclear fuel is strongly desired. We develop an advanced technology for separation of heat generating elements (Cs and Sr) from high level waste to optimize radioactive waste by its characteristics. Recently, some analogues of crown ether or calix-crown extractant were reported on specific selectivity for Sr or Cs, respectively^{1,2)}. In our previous study, some novel solvent impregnated resins (SIRs) were prepared and these SIRs indicated promising ability to separate Cs and Sr from other typical fission products dissolved in nitric acid solution.

An extractant and a modifier were impregnated into porous silica, which is embedded styrene divinyl benzene polymer on the surface. These adsorbents contacting with nitric acid solution were exposed to gamma ray in a vial. After the gamma irradiation, adsorbents were filtrated through a membrane filter and supernatant were measured by LC/MS. The adsorption capacity and distribution coefficients were also examined by using irradiated adsorbents after drying.

Cs SIR has a significant selectivity for Cs by size exclusion. As shown in Fig. 1, the distribution coefficient decreased with dose. After an irradiation of 827 kGy, the distribution coefficient of Cs to that of unirradiated one was depressed to be 1/3 at 30 °C.

No significant degradation compound of extractant was observed by LC/MS analysis. Calix-crown, impregnated into resin, indicated strong stability against irradiation. Thus, it was presumed that decrease of distribution coefficient was due to degradation of alcohol used as a modifier of



Fig. 1 Distribution coefficient of Cs onto Cs SIR from nitric acid solution. SIR was irradiated by contacting with 4 mol/dm³ nitric acid solution. Distribution coefficient was obtained at 30 °C.

adsorbent.

The distribution coefficient of Sr from nitric acid solution onto irradiated Sr SIR at 30 °C was shown in Fig. 2. The distribution coefficient decreased with dose. Some degradation compounds of crown ether, which is impregnated into resin, was observed by LC/MS analysis. It was assumed that crown ether was cloven and dissolved into aqueous phase. The distribution coefficients of typical fission product elements were also evaluated after irradiation. These elements were not adsorbed onto irradiated Sr SIR from 2 mol/dm³ nitric acid solution. Therefore, degradation compounds from Sr SIR have no ability for extraction of above elements and don't prevent selective separation of Sr.

It is concluded that both Cs SIR and Sr SIR maintained their high selectivity for Cs and Sr, respectively, after irradiation. Decreasing ratio of adsorption capacity by irradiation can be estimated from these results. It contributes significantly for conceptual design of separation plant.

Present study is the result of "Development of separation technology of transuranium elements and fission products by using new extractants and adsorbents" entrusted to JAEA by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT). We are very grateful to Mr. R. Yamagata for his valuable cooperation in this study.

References

- P. V. Bonnesen, et al., Solve. Extr. Ion Exch., 18 (2000) 1079.
- 2) J. F. Dozol, et al., EUR 19605EN, (2000).



Fig. 2 Distribution coefficient of Sr onto Sr SIR from nitric acid solution. SIR was irradiated by contacting with 2 mol/dm³ nitric acid solution. Distribution coefficient was obtained at 30 °C.

1-19 Investigation of Stability of Pyrrolidone Precipitants against *γ* Irradiation

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We have been developing a novel reprocessing system for spent FBR fuels based on two precipitation processes^{1,2)}. In this system, only UO_2^{2+} species firstly precipitate in nitric acid solutions dissolving spent fuels by using a pyrrolidone derivative (NRP) with low hydrophobicity and donicity which bring lower precipitation ability. Secondly the residual UO_2^{2+} and Pu (IV, VI) precipitate simultaneously using another NRP with higher precipitation ability. In order to develop such a reprocessing system, it is necessary to evaluate the stability of the precipitants. In this study, γ irradiation to the candidate NRPs was carried out.

N-n-propyl-2-pyrrolidone (NProP), *N-n*-butyl-2-pyrrolidone (NBP) and *N-iso*-butyl-2-pyrrolidone (NiBP) were examined as the candidate NRPs with lower precipitation ability. For NRPs with higher precipitation ability, *N*-(1,2-dimethyl)propyl-2-pyrrolidone (NDMProP), *N*-neopentyl-2-pyrrolidone (NDMProP), *N*-neopentyl-2-pyrrolidone (NNpP), and *N*-cyclohexyl-2-pyrrolidone (NCP) were examined. A 3 mol·dm⁻³ (= M) HNO₃ solution containing 2 M NRP was put into a Pyrex glass tube. Gamma irradiation by the ⁶⁰Co source was performed basically at 13 kGy/h up to 1 MGy at room temperature under atmosphere. An aliquot of the irradiated sample solution was added to 3 M HNO₃ solution containing 1 M UO_2^{2+} at the ratio of [NRP]/[UO_2^{2+}]=1.4. The stability of the precipitant was evaluated by the precipitation ratio of UO_7^{2+} (P.R., %) calculated as,

 $P.R. = 100 \text{ x} ([U]_I - [U]_S)/[U]_I,$

where $[U]_I$ and $[U]_S$ are the concentrations of UO_2^{2+} in the HNO₃ solution before precipitation and those in supernatant liquids, respectively.

For the appearances of the sample solutions of NRPs after the irradiation, almost no changes were observed in NRP with lower precipitation ability. Among NRP





solutions with higher precipitation ability, the solution of NDMProP showed almost no changes as well. However, those of NNpP and NCP were found to split into 2 phases before the irradiation of 1 MGy and 0.2 MGy, respectively. The ease of the splitting was in accordance with the hydrophobicity of NRP. The changes in P.R. versus irradiation dose for NRPs with lower and higher precipitation ability are shown in Figs. 1 and 2, respectively. It can be seen from Fig. 1 that the decreases in P.R. of NRPs with lower precipitation ability are gentle, indicating that all of these precipitants have equivalent and stability enough to be used for multiple precipitation cycles. For NRPs with higher precipitation ability, Figure 2 indicates that all of them have enough stability as well up to ca. 0.5 MGy, and the order of the stability was evaluated as follows judging from the ease of the splitting of the solutions; NDMProP > NNpP > NCP.

Acknowledgment

Present study is the result of "Development of Advanced Reprocessing System Using High Selective and Controllable Precipitants" entrusted to Tokyo Institute of Technology by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT). The authors also greatly thank Mr. R. Yamagata, Department of Advanced Radiation Technology, TARRI, JAEA, for his help at the irradiation facility.

- Y. Ikeda, *et al.*, Proc. Intl. Conf. on Advanced Nuclear Fuel Cycles and Systems (GLOBAL2007), (2007) 1503.
- Y. Morita, *et al.*, Proc. Intl. Conf. on Advanced Nuclear Fuel Cycles and Systems (GLOBAL2007), (2007) 1508.





1-20 Gamma-Ray Irradiation Tests of Magnetic Sensors

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In order to take countermeasures to irradiation assisted stress corrosion cracking of internal structure material in Supercritical Water Cooled Fast Reactor (SWCFR), development of nondestructive detection system for material degradation before cracking has been carried out. This development is aiming at establishing a symptom diagnostic system of material degradation by magnetic measurement by evaluating the relationship between magnetic properties and material degradation, and using electro-magnetic approaches. For this system, magnetic sensors assembled in probes of applicable system to real component are required to have sufficient radiation durability. Extensive irradiation tests of magnetic sensors have therefore been conducted at the gamma-ray irradiation facility in TARRI, JAEA.

Irradiated sensors examined were a Type-I coil sensor, a FG (flux gate) sensor and a MI sensor. The Type-I coil sensor had a core diameter of 3 mm, and was coiled with polyimide insulated wire. All the surface of the FG sensor and the MI sensor were coated with polyimide¹). All the three sensors were irradiated at room temperature. The dose rate was 10,000 Gy/h, and the maximum dose was 10 MGy, which is higher than that assumed to be exposed at the time of maintenance of reactor²).

Examples of the results are shown in Figs. 1 and 2. Inductance and impedance of the detection coil of Type I sensor were not influenced by gamma ray irradiation at the frequencies of 1, 10 and 100 kHz. The changes of the



Fig. 1 Inductance of the detection coil of Type I coil sensor.

measured data before and after irradiation were within 3%. These seem to be within errors. Linearity of both the FG sensor and the MI sensor were obtained up to the dose of 6 MGy. The output voltage excited by change of magnetic field showed a variation of only 3% at the dose from 0 to 6 MGy. This small variation does not seem to affect the magnetic property measurements of materials. Although slight increase of noise was observed, we also succeeded in reducing noise signals by adding magnetic data averaging function to the measurement system and increasing exciting voltage.

We have confirmed sufficient durability of these sensors to gamma ray irradiation with these results.

Acknowledgement

Present study is the result of "Development of diagnostic method of in-core structure material degradation for super critical water cooled fast breeder reactor" entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

- 1) K. Ara et al., J. Magn. Soc. Jpn, 26, 536(2002).
- H. Yoshikawa et al., J. Atom. Ener. Soc. Jpn, 48 (2006).
 461.



Fig. 2 Impedance of the detection coil of Type I coil sensor.

1-21 Irradiation Simulation of Neutron Damage Microstructure in Extra High Purity Fe-25Cr-35Ni Austenitic Stainless Steels

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The ultra high burn-up (>100 GWD/t) of LWR is considered to be an important technology for establishing nuclear power plants as one of the most promising future energy systems from a view point of reducing radioactive waste and greenhouse gas. Cladding materials with the long excellent performance under heavy irradiation are required to these developments. The high chromium and high nickel austenitic stainless steel (25Cr-35Ni-0.2Ti, EHP alloy: Extra High Purity alloy)^{1,2)} was selected as one of candidates that are possible to be made by the present engineering technologies.

Irradiation induced microstructure changes are believed to be the key variables responsible for degradation of materials in LWR. Months to years of irradiation time are required to obtain a change in microstructure of materials at dozens of dpa. Charged particle simulation with accelerated damage rate is often used in such situations as to forecast the behavior of neutron-irradiated materials. By establishing the correlation between charged particle- and neutron-irradiated microstructures of EHP alloy, the results from charged particle-irradiation can be used to provide valuable information on microstructure evolution in EHP alloy in LWR cores.

This work is focused on investigating the microstructure of EHP alloy irradiated with charged particle under condition relevant to LWR cores. The dislocation loop density and size, the void density and size, and irradiation hardening are determined as a function of irradiation temperature, and are compared to those for neutron irradiations of EHP alloy.

Chemical composition of the materials tested is shown in Table 1. EHP alloy was machined from the tube (ϕ 11.3OD × t 0.4 mm) produced by the thermo-mechanical treatment so-called SAR (strained, aged and recrystallized)^{1,2)}. The thickness of specimen is about 0.2mm. The surface of specimen was polished with #2400 emery paper and 0.1 µm diamond paste.

The specimens were irradiated in triple (12 MeV Ni^{3+} , 1.1 MeV He^+ and 380 keV H^+) ion beam mode at temperatures of 300, 350, 400, 450 °C using the triple ion beam facility (TIARA). The temperature of the specimen was measured by an infrared thermometer (THERMAL VISION, Nikon Co.). The displacement damage in the specimen was mainly attributed to Ni^{3+} ion irradiation. The peak dose was about 5 dpa at around 2 μ m depth. The He⁺ and H⁺ ions were implanted in depth ranges from 1.0 to 1.5 μ m using aluminum foil energy degraders. The concentrations of He⁺ and H⁺ ions in the implanted range were 3 appmHe and 15 appmH, respectively, which correspond to LWR condition. The dose was about 2 dpa in the implanted range of He⁺ and H⁺ ions.

A focused ion beam (FIB) microprocessor was applied to prepare thin specimens for microstructure observation. FIB processing technique is using focused Ga⁺ ion beam sputtering. FIB processor could pick up a small piece of thin foil sample from chosen area. Size of samples are about $20 \times 6 \times 0.05 \ \mu\text{m}^3$. It would be efficient to reduce radiation activity when it would be used for irradiated materials. Detail of a micro-sampling system of FIB processor is explained in the reference³⁾.

Figure 1 shows the bright field image at each irradiation temperature. Some defects caused by FIB processing were observed in each image. After removal of FIB defects, microstructure of charged particle irradiated specimens would be characterized by the dislocation loop density and size, the void density and size. Also, neutron irradiated specimens would be analyzed.



Fig. 1 Bright field image at each irradiation temperature.

- 1) K. Kiuchi et al., IAEA TCM, Argentina, Nov.(1999) 15-19.
- 2) K. Kiuchi et al., ICAPP, June 9-13, 2002 (Florida).
- 3) H. Tanigawa et al., J. Nucl. Mater., 297 (2001) 279.

Table 1 Chemical composition of the material tested in weight percent (wt%)

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Materials	Fe	C	Si	Mn	Р	S	Cr	Ni	Ti	N	0	Co
25Cr-35Ni-0.2Ti EHP	bal.	0.0013	< 0.005	0.001	0.001	0.0009	24.55	34.99	0.18	0.0014	0.0011	-
Type 304SS	bal.	0.063	0.49	1.45	0.016	0.012	18.72	10.27	-	-	-	0.03

1-22 Evaluation of Microstructure Change in Extra High Purity Austenitic Stainless Steel under BWR Condition Simulated by Triple Ion Irradiation

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1. Introduction

To prevent material degradation under LWR condition and corrosion, we developed extra high purity (EHP) grade (C + N + O < 100 wppm) and high Cr (~25 wt% Cr) austenitic stainless steel. We have evaluated change of mechanical properties and corrosion behavior after neutron irradiation up to 2 dpa in JRR-3. As a result, the EHP grade stainless steel has good IASCC (Irradiation Assisted Stress Corrosion Cracking) resistance than SUS304. However, it is not clear if the microstructure of EHP grade stainless steel does change or not after heavy irradiation. The purpose of this study is to evaluate irradiation resistance of EHP grade stainless steel for heavy irradiation.

In this paper, triple ion irradiation that simulates BWR condition was carried out for the candidate (Fe- 25Cr- 35Ni- 0.2Ti EHP grade). After irradiation, TEM observations are carried out and evaluate the number density and average diameter of dislocation loops.

2. Experimental procedures

The candidate was cut to a size of $\phi 3 \times 0.2$ mm. Irradiated area was then mechanically and electrochemically polished to a specular finish. Triple ion irradiation simulating BWR neutron irradiation condition was performed using Ni³⁺ (12 MeV), He⁺ (1.1 MeV) and H⁺ (0.38 MeV) ion beams. Aluminum film degraders were used to spread He⁺ and H⁺ irradiated area. Irradiation temperature was 573 K. Irradiation damage and ion-implantation depth were calculated using TRIM code. As a result, He⁺ and H⁺ ion-implantation depth was estimated to be 1.1~1.3 µm and irradiation damage of this area to be 75 dpa. He^+ and H^+ concentrations in ion-implantation area were 3 and 30 appm/dpa, respectively. TEM observation was carried out to evaluate number density and average diameter of dislocation loops. To get irradiated area on TEM specimen, irradiated surface was electropolished to 1.2 µm depth before electropolish on non-irradiated surface using single-jet electropolish method.

3. Results

Figure 1 shows weak beam image of specimen after ion-irradiation observed by TEM. This image shows that irradiation defects such as dislocation loops are introduced by ion-irradiation. Number density and average diameter of dislocation loops that were estimated from weak beam images are 2.04×10^{21} n/m³ and 11.54 nm, respectively. Figure 2 shows number density and average diameter of dislocation loops as functions of dpa. This graph shows that the growth of dislocation loops is saturated up to 10 dpa. This result suggests that EHP grade stainless steel has good irradiation resistance for heavy irradiation.



Fig. 1 Weak beam image (g*/3g*) of irradiated specimen.



Fig. 2 Ion irradiation dependence on number density and average diameter of dislocation loops.

1-23 Effects of Microstructural Evolution in Low-alloy Steels for Radiation Hardening under Electron Irradiation

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Understanding the mechanisms at the origin of radiation hardening and associated embrittlement is of great interest for predicting the evolution of reactor pressure vessel steel during nuclear power plant running. In the present study, to examine the behavior of different solutes under irradiation, radiation hardening behavior in 3 model alloys (Fe-2%Ni, Fe-2%Ni-0.5Cr, and Fe-0.6%Cu) and pure Fe under electron irradiation was investigated.

The alloy ingots of Fe-2%Ni, Fe-2%Ni-0.5%Cr, and Fe-0.6%Cu (in weight percent) were made by means of vacuum induction melting method in order to reduce impurities to very low level. Highly purified (99.99%) iron sheet supplied by Nilaco Co. Ltd., was used as pure Fe. After cold-rolling to sheets of about 5mm thickness, Fe-2%Ni and Fe-2%Ni-0.5%Cr were water-quenched after annealing at 950 °C for 1 hour and tempered at 750 °C for 1 hour, and Fe-0.6%Cu was water-quenched after solution treatment at 825 °C for 4 hour to keep the copper in supersaturated solution. Pure Fe was annealed at 800 °C for 4 hour and air cooled. After heat treatment, specimens with $15 \text{ mm} \times 15 \text{ mm} \times 0.5 \text{ mm}$ in dimension were made from inner part of each sheet and irradiated with 2.0 MeV electrons using a Dynamitron accelerator at JAEA-Takasaki to doses of 3.5×10^{23} e⁻/m², corresponding to calculated atomic displacement doses of 1.2×10^{-3} dpa. The irradiation temperature was kept at 200 °C or less and the dpa rate was



Fig. 1 Increases in indentation hardness as a function of irradiation dose for pure Fe, Fe-2Ni, Fe-2Ni-0.5Cr, and Fe-0.6Cu alloy. Here, calculated Vickers hardness change is indicated at the right side of the vertical axis for the convenience.

 3.0×10^{-9} dpa/s. The indentation hardness test was performed using the SHIMADZU Nanoindenter DUH-211S before and after of irradiation. The applied load was 100 gf and measuring temperature was about 23 °C.

Figure 1 shows the dpa dependence of the increases in indentation hardness of the irradiated materials. As shown in the figure, the change in hardness in Fe-0.6%Cu alloy showed remarkable increase and tends to be saturated at the high dose. This result has good agreement to the previous work¹⁾. By contrast, change in hardness in pure Fe, Fe-2%Ni and Fe-2%Ni-0.5%Cr alloy are much smaller than that in Fe-0.6%Cu alloy. However, attention should be paid to that initial hardness of each material is different as shown in Fig. 2. Moreover, from the difference between the result of Fe-2%Ni and Fe-2%Ni-0.5%Cr alloy, it is revealed that the addition of Cr enhanced the radiation hardening in Fe-2%Ni alloy. Further experiments and investigations by using positron annihilation spectroscopy (PAS) and small angle x-ray scattering (SAXS) measurements will be made in the future in order to clarify the relation between irradiation hardening and microstructural evolution formed by solute segregation and point defect accumulation under irradiation.

Reference

1) T. Tobita et al, J. Nucl. Mater. 299 (2001) 267.



Fig. 2 Comparison of indentation hardness before (unirrad.) and after (1.2×10⁻³ dpa) of irradiation for pure Fe, Fe-2Ni, Fe-2Ni-0.5Cr, and Fe-0.6Cu alloy. All initial hardness is smaller and that after irradiation is larger. Calculated Vickers hardness is also indicated.

1-24 Study on Irradiation Defect Generation Process and Radiation Induced Segregation in Fe-based Model Alloys

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To investigate the fundamentals of irradiation embrittlement mechanisms of Reactor Pressure Vessel steel (RPVs), we have performed electron irradiation experiments using Fe-based model alloys, such as pure Fe, Fe-Cu and Fe-P. The impurities in RPVs materials have been known to have a strong effect on the irradiation embrittlement. The Cu precipitates which grow during irradiation cause irradiation hardening, while the P segregation to the grain boundary weakens the Fe-Fe bonds. Our previous study^{1,2)} shows the changes of electrical resistivity of Fe-Cu and Fe-P model alloys induced by electron irradiations at high temperature to evaluate the irradiation damage mechanisms, such as the behaviours of irradiation defect or Cu clustering in the materials.

In this paper, we measured the changes of hardness of several types of Fe-based model alloys induced by 2 MeV electron irradiations at high temperature (523 K). We tried to obtain the relation between mechanical property and electrical resistivity change.

The chemical composition of these materials is shown in Table 1. The Fe-based model alloys were fabricated as pure as possible to eliminate the effects of other impurities. After the heat treatment, the materials were quenched to keep the Cu and P atoms in supersaturated solid solution at room temperature. Specimens with $5 \times 5 \times 1$ mm in dimension were irradiated with 2.0 MeV electrons at dose rate of 1.3×10^{-9} dpa/s. The hardness was measured by 500gf Vickers hardness tester.

Figure 1 shows electron dose dependence of the electrical resistivity change^{1,2)} and irradiation hardening. In the case of Fe-0.6%Cu, the hardness increases with decreasing electrical resistivity, clearly. However, in spite of large decreasing in electrical resistivity, the hardening of Fe-P was negligible. It means that detection of P segregation effect on mechanical properties by electrical resistivity is impossible.

Figure 2 shows the relationship between irradiation hardening and electrical resistivity change in Fe-0.6%Cu. Where, these Δ HV values were estimated by linear interpolation at the same dose of $\Delta \rho$ dataset. The relationship is described below;

At this temperature and dose, accumulation of irradiation defects (vacancy and interstitial) is negligible¹⁾. Therefore, both electrical resistivity change and hardening were mainly caused by irradiation induced Cu clustering. This formula suggests that irradiation embrittlement of

RPVs caused by Cu clustering may be evaluated by measuring electrical resistivity.

Table 1 Chemical composition of materials (wt %).



Fig. 1 Electron dose dependence of the electrical resistivity change($\Delta \rho$) and irradiation hardening(ΔHV) in Fe-0.6%Cu, Fe-0.1%P and Fe-0.04%P.



Fig. 2 Relationship between electrical resistivity change and irradiation hardening in Fe-0.6%Cu.

- T. Tobita, Y. Chimi, N. Ishikawa, S. Nakagawa and A. Iwase, JAEA-Review 2006-042 (2007) 20.
- T. Tobita, Y. Chimi, N. Ishikawa, S. Nakagawa and A. Iwase, JAEA-Review 2007-060 (2008) 27.

1-25 Corrosion Behavior of Type 316L Stainless Steel Ion-irradiated under Strain Condition

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Irradiation assisted stress corrosion cracking (IASCC) on austenitic stainless steel (SS) is one of the main issues in reactor cores of nuclear power plants. Degradation of materials under irradiation and applied stress are important factors affecting the IASCC initiation. The objective of this work is to obtain the fundamental knowledge on the synergistic effect of strain and irradiation on the corrosion behavior of type 316L SS. The correlation between the corrosion behavior and microchemistry change in irradiated specimens was studied in this paper.

In order to examine the synergistic effect of strain and irradiation, flat plate specimens that were plastically deformed by bending were irradiated by 12 MeV Ni³⁺ ion using the tandem accelerator at JAEA-Takasaki. Plastic strain levels of the irradiation surface region were selected to about 2% tensile strain, 7% tensile strain and 2% compressive strain. Irradiation temperature was 330 °C. The displacement damage levels ranged from 1 to 45 dpa. The corrosion resistance of irradiated specimens was estimated by means of single-loop electrochemical potentiokinetic reactivation (SL-EPR) testing, in which the corrosion property was evaluated using the normalized charge (P_a). Three dimensional atom probe (3DAP) analysis was conducted on the irradiated specimens to investigate the compositional change microchemical in irradiated specimens.

The dependence of the P_a on displacement damage is shown in Fig. 1. The P_a of undeformed (strain free) specimen increased rapidly with increasing dose, and became saturated above 12 dpa. On the other hand, P_a values of deformed specimens proved to be smaller than that of the undeformed specimen. The degradation of corrosion resistance caused by the irradiation was suppressed under the strain state, and the suppressive effect was significant in 2% compressive strain condition, followed by 2% and 7% tensile stain condition.

To identify cause of the difference in corrosion resistance among specimens irradiated under stain free and strain condition, the solute distribution in specimens irradiated up to 6 dpa was investigated using 3DAP. Radiation induced segregation, enrichment of Si, Ni and depletion of Cr, occurred around Frank loops and dislocations1) in the matrix in irradiated specimens regardless of applied strain level. However, the degree of segregation was changed by applied strain during irradiation. Figure 2 shows the strain dependence on compositional change from bulk at the sink such as Frank loop and dislocation. Compositional change of Si and Ni in specimen applied 2% tensile or compressive strain, which showed high corrosion resistance, was obviously lower than that in the strain free specimen. Segregation behavior in 7% tensile strain was similar to that in the stress free specimen. About 2% tensile or compressive strain enhanced the suppression effect on segregation.

The Cr depletion around Frank loops in the matrix was not affected by applied strain level, though the enrichment of Si and Ni might be affected. This enrichment influenced the P_a value, but was thought that the enrichment was not enough to cause large changes of the P_a value.

Present study includes the result of "New evaluation method of material degradation considering synergetic effects of radiation damage" entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT). A part of this study was financially supported by the Budget for Nuclear Research of the MEXT, based on the screening and counseling by the Atomic Energy Commission.

Reference



Fig. 1 Dose dependence of corrosion behavior in single-loop EPR testing on deformed and undeformed specimens. 1) K. Kondo et al., JAEA Takasaki Ann. Rep. 2006 (2007) 23.



Fig. 2 Strain dependence of the radiation induced segregation around dislocations in matrix.

1-26 Damage in Oxide Ceramics Irradiated with 10MeV Ni at High Temperature

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Radiation damage is one of the important causes of degradation of thermal conductivity and serious swelling in nuclear ceramic fuels. Understanding of radiation damage in nuclear fuel is important for controlling fuel properties during burn up and for the prediction of fuel burn up behavior. The objectives of this study are to elucidate the irradiation parameter dominating the radiation damage of ceramics by means of accelerator experiments, and to precisely evaluate their radiation damages. Especially for simulating the process of radiation damage by elastic displacements, low-energy particle (10 MeV Ni) irradiation experiment has been performed. The oxide ceramics target, CeO₂ target, which has the same crystal structure (fluorite structure) with nuclear fuel is adopted. In the present study, the radiation damage behavior especially at very high temperature (800 °C) has been investigated systematically.

Thin films of CeO₂ were prepared on Al₂O₃ single crystal substrates, and were irradiated with 10MeV Ni ions at 800 °C. After the irradiation, fluence dependence of change of X-ray diffraction (XRD) pattern was measured in the wide fluence range up to 2.4×10^{16} ions/cm², and the result was compared with that obtained for irradiations at room temperature and 400 °C. The film thickness was restricted to be about 0.3 µm which is thin enough to rule out the possibility of unrequired implantation effects.

In Fig. 1 evolution of X-ray diffraction peak for different fluence is shown for CeO2 irradiated with 10 MeV Ni at 800 °C. A sudden decrease in peak intensity is observed in the high fluence range of about 1.0×10^{16} ions/cm². This tendency is clearly demonstrated in Fig. 2. The other aspect of XRD pattern for 800 °C irradiation is that new peak appeared at lower angle side of (002) peak. The intensity of the new peak depends on fluence, and the new peak is not induced by just heating the CeO₂ sample to 800 °C. It should be noted here that the new peak is not observed for CeO₂ irradiated at room temperature and 400 °C. The new peak observed after the irradiation at 800 °C suggests that there is a threshold for creation of new crystallographic structure between 400 °C and 800 °C. One possibility for the origin of the new peak is a radiationinduced reallocation of oxygen atoms which can move fast at high temperature.

Present study is the result of "Research of highly accurate evaluation of radiation damage in advanced nuclear reactor fuel ceramics" entrusted to "Japan Atomic Energy Agency" by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).



Fig. 1 Evolution of X-ray diffraction peak assigned for (002) reflection of CeO₂ irradiated with 10MeV Ni at 800 °C. The fluences are 0 (top), 1.0×10¹³ ions/cm², 1.0×10¹⁴ ions/cm², 1.0×10¹⁵ ions/cm², 1.0×10¹⁶ ions/cm² and 2.4×10¹⁶ ions/cm² (bottom).



Fig. 2 Fluence dependence of XRD peak intensity normalized by that of unirradiated sample for CeO₂ irradiated with 10 MeV Ni at room temperature (RT: open squares), 400 °C (open circles) and 800 °C (closed circles).

1-27 Effects of H⁺ Implantation in Multi-ion Irradiation on Microstructural Changes in Li₂TiO₃

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 Li_2TiO_3 is regarded as one of the most suitable candidates for the solid tritium breeder material of D-T fusion reactors. Simulation of the fusion reactor environment and hence the study of a synergistic effect of atomic displacement damage in Li_2TiO_3 are presumed to be approached by a simultaneous irradiation with "triple" ion beams which consist of O^{2+} , He⁺ and H⁺ ion beams. In this study, the results of the FT-IR photoacoustic spectroscopy (PAS) with Li_2TiO_3 samples irradiated with the Multi-ion beam were analyzed for investigation of microstructural changes.

Recently, we have started to examine new Li_2TiO_3 samples which were prepared by ART Kagaku. The reported preparation method and the characteristics of the new samples are quite similar to the previous samples described in ref.1. In addition, the characteristic peaks in FT-IR PAS spectra for non-irradiated Li_2TiO_3 sample were also similar between two samples. As discussed in ref.1, in comparison to the reported spectra of TiO₂ and Li_2TiO_3 , 680- and 780-cm⁻¹ peaks are identified from Ti-O bond, 1570-, 3150- and 3450-cm⁻¹ peaks are from the O-H bond in hydroxyls adsorbed on or near the surface, and others are from C=O bond in Li_2CO_3 formed on or near the surface by reaction with CO₂ in the atmosphere¹.

In the previous study using the previous samples, the amount of the formed hydroxyl (3450-cm⁻¹ peak) is observed to be affected not only by displacement per atom (dpa) but also by the method of irradiation, that is, the single, the simultaneous triple/dual or the sequential triple/dual irradiation. This may suggest that H⁺ implantation after irradiation degrades the generation of hydroxyl (-OH) group near the surface by trapping hydrogen in the irradiation defects or radiolytic products^{2,3)}.

However, on the other hand, the results of examinations using the new samples were observed to have opposite dependence on H⁺ ion implantation. Figure 1 shows dependence of the 3450-cm⁻¹ peak area of the irradiated new samples on dpa by the single (O^{2+}) , dual $(O^{2+}+H^+)$ and sequential $(O^{2+}$ after H^+ or H^+ after O^{2+}) irradiation. This shows that the peak area of the samples irradiated by single O²⁺ ion irradiation increases with dpa by the irradiation. However, that of the sample irradiated by dual irradiation is observed to be larger than by single ion beam irradiation. Further, that of the samples irradiated by sequential (H⁺ after O²⁺) irradiation is observed to be larger than that by dual irradiation, while that by sequential (O^{2+}) after H⁺) irradiation is observed to be smaller than that by dual irradiation and the same as that by single irradiation. In comparison with the results of experiments using the

previous samples, the effects of H^+ ion implantation on the new samples are quite opposite, that is, for the previous samples, the H^+ ion implantations with or after O^+ ion irradiation are observed to degrade the generation of hydroxyl group near the surface²⁾, while, for the new samples, to promote the generation of hydroxyl group near the surface.

Although this significant difference in results using the two types of samples is quite interested, the cause of this difference is not clarified so far. One possible suggestion is some slight differences in preparation processes such as pressure, composition, temperature, and so on, which could not be described clearly in the preparation report, may greatly affect the surface quality of the samples. Further studies are needed for identification of the difference of surface quality between the two types of the samples.

- 1) D. Yamaki et al, J. Nucl. Mater. 329-333 (2004) 1279.
- 2) D. Yamaki et al, JAEA-Review 2005-001 (2006) 212.
- 3) D. Yamaki et al, JAEA-Review 2007-060 (2008) 173.



Fig. 1 The effects of dpa by O^{2^+} irradiation and H^+ implantation on the 3450- cm⁻¹ peak area, which is due to the O-H bond. Notation as "A+B" means simultaneous irradiation of the beams A and B, and "A \rightarrow B" means that irradiation with beam B was performed after closing of irradiation with beam A.

1-28 Gamma-Ray Irradiation Tests of Quartz Optical Fiber Transmission Line for Commercially-Available Multi-kW High Power Yb-doped Fiber Lasers

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An Yb-doped High Power CW Fiber Laser has been used widely to weld, to cut and to peel some industrial materials like stainless-steel, aluminum, and other alloy and pure metals because of the relatively-cheap prices, long-life time, easiness in operation and maintenance. We plan to apply the fiber lasers to disassemble the nuclear reactor system inside and outside of the nuclear reactor vessel, therefore we have to know how gamma rays inside the nuclear reactor vessel shows that irradiation of oxide superconductors with 0.5 MeV C monoatomic ion beam causes atomic displacements via elastic collision and it results in lattice expansion¹⁾. We have recently reported that the cluster effect is absent in lattice expansion for oxide superconductors EuBa₂Cu₃O₇ irradiated with 4 MeV C₈ cluster ions, indicating the absence of cluster effect in terms of defect density²⁾. However, in the present report we report a new insight on the cluster effect observed for some aspects of XRD(X-ray diffraction) pattern of EuBa₂Cu₃O₇ irradiated with cluster ions.

Thin films of c-axis oriented EuBa₂Cu₃O_y (y=7) were prepared on MgO substrates by a sputtering method. $[0.5 \times n]$ MeV C_n (n=1,4,8) ions, all having the same velocity, were irradiated at room temperature using the tandem accelerator at JAEA-Takasaki. The projected range of those ions is about 0.6 μ m, but for the cluster ions they become dissociated during the passage in the sample and have no more spatial correlation as the clusters proceed deeper into the sample. In order to observe the cluster effect, the sample thickness was determined to be 0.1 μ m, which we believe is enough to assure the spatial correlation of clusters during their passage through the sample. The fluence dependence of c-axis lattice parameter was examined by





measuring the positions of XRD peaks. The fluence dependence of XRD peak intensity and FWHM (Full Width at Half Maximum) of those peaks was also measured.

As reported in the previous report²⁾, the XRD peak position corresponding to c-axis lattice spacing shifts to lower angle side in the same manner for the irradiation with 0.5 MeV C, 2.0 MeV C_4 and 4 MeV C_8 when the effects are compared in the unit of carbons/cm². Since the lattice expansion by 0.5 MeV C irradiation has already been known to be attributed to the elastic displacements¹), the lattice expansion for the irradiation with 2.0 MeV C_4 and 4 MeV C_8 can be understood in the framework of the elastic displacements. From the above results, no sign of the cluster effects is detected. However, as demonstrated in Figs.1 and 2, the cluster effect is observed for some aspects of an XRD peak; the fluence dependence of XRD peak intensity, and that of FWHM of XRD peaks.

These cluster effects can be understood in the framework of elastic displacements, and the key parameter for the interpretation for the present result is the energy of PKA (primary knock-on atoms). The irradiation with larger cluster is expected to cause higher maximum PKA energy, since larger cluster has higher mass (with same velocity in this case). Higher maximum PKA energy can create larger defect agglomerates rather than simple defects. The larger decrease in intensity of XRD peak and larger increase in FWHM for irradiation with larger cluster may be related to the higher energy of PKA.

- 1) N. Ishikawa et al., Nucl. Instr. Meth. B 135 (1998) 184.
- 2) A. Iwase et al., TIARA annual report 2003 (2004) 217.



Fig. 2 Fluence dependence of FWHM of (005) peak for 0.5 MeV C, 2.0 MeV C₄ and 4 MeV C₈ irradiation. The fluence is indicated in the unit of carbons/cm² in order to facilitate detection of the cluster effect.

1-29 Preparation of Chemically Stable Hybrid Polymer Electrolyte Membranes by Combining the Radiation Grafting with Silane-Crosslinking

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Hybrid polymer electrolyte membranes (PEM) have been widely studied for their application in intermediate temperature fuel cells. The inorganic particles in the PEM enhance the water retention and thermal stability of the hybrid PEMs. However, we found that the inorganic particles are easily separated from the PEMs after long immersing in water. In our previous study, we developed a covalently crosslinked hybrid PEM combining the radiation grafting and silane-crosslinking techniques.¹⁾ In that study, we grafted p-styryltrimethoxysilane (StSi), which has aromatic ring amenable to sulfonation and a trimethoxysilyl group amenable to silane-crosslinking, to ethylene tetrafluoroethylene (ETFE) film. After grafting, sulfonation and then hydrolysis-condensation were performed to give the PEM proton conductibility and crosslinks. These radiation-grafted and silane-crosslinked hybrid PEMs are very thermally stable, and is comparable to Nafion.

In this study, to further enhance the properties of the proton-conducting hybrid PEM, the effect of changes in the preparation process, such as the timing for hydrolysis-condensation (before or after sulfonation) and the sulfonation method on the chemical stability as well as on the ion exchange capacity (IEC), proton conductivity, water uptake, and methanol permeability of the hybrid PEMs were investigated.



Fig. 1 Water uptake (a) and proton conductivity (b) of the StSi-grafted PEM as a function of IEC.

The grafting of StSi to the ETFE films was carried out by a γ -ray preirradiation method.¹⁻⁴⁾ After grafting, the StSi-g-ETFE films were subjected to sulfonation and hydrolysis-condensation for preparation of the PEM. The sulfonation of the StSi-g-ETFE film in ClSO₃H solution was quite slow; the sulfonation level was always below 100% even after sulfonation for a long time. On the contrary, when high concentration H₂SO₄, the sulfonation level was quickly beyond 100%, indicating that more than one sulfonic acid group was substituted for each aromatic ring.



Fig. 2 Relationship between the durability time toward 3% H₂O₂ aqueous solution at 60 °C and proton conductivity of the StSi-grafted PEMs.

The resulting PEM with sulfonic acid groups has hydrophilic and hydrophobic phases. The mutual relationship among the proton conductivity, IEC and water uptake is shown in Fig. 1. Both the water uptake and proton conductivity of the StSi-grafted PEM increased with increase in IEC, and were independent of the sulfonation method and the crosslink timing. The highest proton conductivity of 0.16 S/cm and highest IEC of 1.86 mmol/g were quite higher than those of Nafion membrane, 0.06 S/cm and 0.91 mmol/g, respectively. The chemical stability of the PEM was tested in a 3% H₂O₂ aqueous solution at 60 °C. The durability time of the StSi-grafted PEMs was investigated as a function of proton conductivity, and the results are shown in Fig. 2. As expected, the durability times of the crosslinked PEMs logarithmically decreased with the increase in proton conductivity, and were independent of the sulfonation method and crosslink timing. Even then, the crosslinked StSi-grafted PEMs were quite stable compared to the St/divinyl benzene (DVB)-grafted PEM having the low proton conductivity of 0.06 S/cm. In addition, the methanol permeability of the crosslinked hybrid PEM was almost six times lower than that of Nafion. Together with the high thermal stability reported in our previous study, we conclude that the properties of the StSi-grafted crosslinked hybrid PEMs make them promising materials for fuel cells.

- J. Chen et al, J. Membr. Sci. 296 (2007) 77; 277 (2006) 249; 269 (2006) 194; 256 (2005) 38.
- 2) J. Chen et al, J. Power Sources, 158 (2006) 69.
- 3) J. Chen et al, J. Appl. Polym. Sci. 100 (2006) 4565.
- 4) J. Chen et al, Polymer 48 (2007) 600.

1-30 Development of Polyetheretherketone-Based Polymer Electrolyte Membranes for High Temperature Fuel Cells

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Polymer electrolyte membrane fuel cells (PEMFCs) are very promising as power sources for automotive applications, where stability at a high temperature and low humidity is demanded, but under such operation conditions, the currently used expensive Nafion membranes cannot maintain their performance stable. For this reason, inexpensive polymer electrolyte membranes have been extensively studied for their suitability in high temperature PEMFCs.¹⁻⁷⁾ In this study, ethyl styrenesulfonate (ETSS) monomer was graft polymerized onto a divinylbenzene-treated polyetheretherketone (PEEK) films, followed by hydrolysis of the ETSS graft chains to yield the SS graft chains, so that the new ssPEEK electrolyte membrane was obtained without the sulfonation damage.



Fig. 1 I-V performances of the ssPEEK- and Nafion-based fuel cells at operation conditions: (a) 80 °C and 100 RH-%, and (b) 95 °C and 40 RH-%.

The ssPEEK membrane was tested for use in a fuel cell. For comparison, the Nafion 212 was also tested under the same condition. Figure 1 shows the I-V performances of the fuel cell with the ssPEEK and Nafion membranes. Under the operation conditions 80 °C and 100 RH-% (Fig. 1a), both ssPEEK and Nafion membranes exhibited good cell performance. On the contrary, under the operation conditions 95 °C and 40 RH-% (Fig. 1b), the performance of the Nafion-based fuel cell was considerably worse than that of the ssPEEK-based cell.

Both the ssPEEK and Nafion-based cells were operated at 95 °C and 40 RH-% to evaluate their long-term membrane durability. As can be seen from Fig. 2, the ssPEEK-based cell could be stably run for more than 250 h, with a cell voltage around 0.6 V. The cell voltage periodically increased by about 0.05 V, due to the membrane electrode assembly (MEA) not being optimized for water measurement, and to the fluctuation of the gas pressure in the cell. On the other hand, the cell voltage of the Nafion-based cell progressively decreased from 0.6 V to 0.3 V during the 250 h of operation. This decrease in cell voltage was due to the degradation of the Nafion membrane, resulting in the formation of pinholes on the membrane allowing for reactant crossover.



Fig. 2 Durability test of the ssPEEK- and Nafion-based fuel cells at a high temperature and low humidity condition 95 °C and 40 RH-%.

The continuously increased crossover could also be verified by the decrease in open circuit voltage (OCV) of the cell from 0.977 V to 0.856 V. Furthermore, the thickness of the Nafion in the active area decreased from 52 μ m to 46 μ m due to the membrane degradation. Therefore, the Nafion is not suitable for application in a fuel cell under a high temperature and low humidity conditions. The new ssPEEK electrolyte membranes are promising and deserve further study.

- J. Chen et al. J. Membr. Sci., 256(2005) 38; 277(2006) 249; 269(2006) 194; 283 (2006) 373; 296 (2007) 77.
- J. Chen et al. J. Appl. Polym. Sci., 100 (2006) 4565; 101(2006) 2661; 103(2007) 1966.
- J. Chen et al. J. Mater. Sci., 41(2006) 1289; 42(2007) 1330.
- 4) J. Chen et al. J. Power Sources, 158(2006) 69.
- J. Chen et al. Electrochem. Solid-State Lett., 9 (2006) G184; 9 (2006), G326.
- J. Chen et al. J. Polym. Sci. A, Polym. Chem., 45(2007), 2624.
- 7) J. Chen et al. Polymer, 48 (2007), 6002.

1-31 Preparation of Fuel-Cell Membranes by Ion Track Technology: Enhanced Reactivity of Ion-Track Grafting

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We have recently been developing proton exchange membranes (PEMs) for polymer electrolyte fuel cells (PEFCs) by our original swift-heavy-ion based technique in addition to the γ -ray or electron-beam crosslinking and graft polymerization¹⁾. Our main focus is the use of cylindrical damage produced along with ion trajectory in polymers, which is called a latent track. The approach using this ion track technology is being taken according to two techniques^{2,3)}. One of these is heavy-ion grafting⁴⁾, i.e., a heterogeneous modification through graft polymerization in the latent tracks only. As shown in Fig. 1, the resulting PEMs are expected to have low fuel permeability and high thermal stability because the substrate matrix without any modifications mechanically prevents excess swelling, while they maintain their proton conductivity. In order to meet this expectation, the largest amount of proton-conductive graft components should be introduced into as small a number of tracks as possible. It was demonstrated here that a proper selection of grafting solvents enhanced reactivity of the ion-track grafting, defined as the degree of grafting.

25-µm-thick poly(ethylene-*co*-tetrafluoroethylene) (ETFE) films were bombarded by 3.5 MeV/n ¹²⁹Xe from the TIARA cyclotron. Just after irradiation, the films were exposed to air to produce peroxide groups for initiating the grafting reactions. The peroxide grafting of styrene was then performed in toluene, tetrahydrofuran, propan-2-ol, and methanol (a styrene/solvent volume ratio was 40/60%) at 60 °C for 24 hours. The fluence was limited to 3.0×10^8 ions/cm², at which almost no ion track overlapped. The degree of grafting was calculated by the following equation:

Degree of grafting (%) = $100(w_g - w_0)/w_0$, where w_g and w_0 are the weights of the styrene-grafted and just pre-irradiated films. The grafted films were sulfonated in a chlorosulfonic acid solution to obtain the PEMs.

Table 1 lists the degree of grafting of styrene into the Xe-irradiated ETFE films in different solvents and ion



Fig. 1 The scheme for PEM preparation by a heavy-ion grafting method; (1) styrene grafting and (2) sulfonation.

exchange capacity (IEC) of the sulfonated PEMs. In a non-solvent system, the degree of grafting leveled off at around 10% after the 24-hour reaction. The use of toluene and tetrahydrofuran, both of which are solvents good enough to swell the grafted polystyrene, was found to retard the reaction under the same conditions. Although the quantitative sulfonation was then completed in a reproducible manner, the IEC of the PEMs was not as high as that of a Nafion membrane (0.9-1.0 meq/g).

On the other hand, poor solvents facilitated the grafting reaction. For example, the degree of grafting in methanol and propan-2-ol appeared to increase by a factor of three and four, respectively, compared to that in neat styrene. According to Odian's study on the y-ray pre-irradiation grafting onto various base polymers⁵⁾, the enhanced reactivity due to dilution with these solvents was related to a reduced termination rate, in other words, a particularly pronounced Trommsdorff-Norrish effect. Additionally, incompatibility of the solvent with the polymer phase should reduce the mobility of the growing polymer chains and extend the radical lifetime; preferential accumulation of the monomer in the polymer phase can increase the local monomer concentration and thus the polymerization rate. As a result, such a high grafting yield enabled us to prepare PEMs endowed with a sufficient IEC and, therefore, proton conductivity.

Table 1 Degree of grafting of styrene in different solvents and IEC of the subsequently sulfonated PEMs.

Solvent	Degree of grafting (%)	IEC (meq/g)
no (neat styrene)	9.4	0.71
toluene	4.5	0.30
tetrahydrofuran	0.20	< ML*
propan-2-ol	42	1.9
methanol	27	1.5

* Measurement Limit

- For review, T. Yamaki et al., "Current Developments of Radiation-Induced Graft in Membranes" (Research Signpost), in press.
- 2) T. Yamaki et al., ECS Transactions 3 (2006) 103.
- 3) T. Yamaki et al., Electrochemistry 75 (2007) 175.
- N. Betz et al., Nucl. Instrum. Methods Phys. Res., Sect. B, 105 (1995) 55.
- 5) G. Odian et al., J. Appl. Polym. Sci. 7 (1963) 245.

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2-01 Preparation of Metal Adsorbent with Sulfo Groups by Radiation-Induced Grafting of Ethyl *p*-styrenesulfonate

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1. Introduction

There are various types of ion exchange resins for different purposes, and these resins have been widely used in various fields. Especially, a graft adsorbent has been attracting attention as a sophisticated ion exchanger because the adsorption rate of the graft adsorbent is 10-100 times higher than that of a commercially available particulate resin¹). In the present study, the preparation of metal adsorbents with sulfo groups was attempted by using radiation-induced graft polymerization of ethyl *p*-styrenesulfonate (ETSS), and the metal adsorption performance was evaluated.

2. Expermental

Polyethylene-coated polypropylene nonwoven fabrics, which were used as the trunk polymer, were purchased from Kurashiki Textile MFG Co. (Osaka, Japan). The average diameter of the nonwoven fabrics was 13 µm. The nonwoven fabrics were irradiated with an electron beam up to 100 kGy in nitrogen atmosphere at dry ice temperature. The irradiated nonwoven fabrics were contacted with the aqueous emulsion solution, which was composed of ETSS, polysorbate 20 (Tween 20) and deionized water, in a deaerated glass ampoule and were kept in water bath at the desired reaction time and temperature. After grafting, the ETSS-grafted samples were further reacted with 1 M potassium hydroxide solution at 80 °C to convert the ester groups of the ETSS graft chain to sulfo groups. The degree of grafting (Dg) was evaluated by the increased weight after grafting, and the degree of saponification (Ds) was estimated by the equilibrium adsorption capacity for Cu^{2+} . The metal adsorption performance of the ETSS-saponified adsorbent was evaluated through the batchwise adsorption tests of Cu²⁺, according to the procedure described by Aoki et al^{1} .



Fig. 1 Effect of reaction temperature on Dg.

3. Results and Discussion

The Dg increased with increasing irradiation dose, ETSS concentration, reaction temperature, and reaction time. The optimum grafting conditions were irradiation dose of 100 kGy, ETSS concentration of 3 wt%, Tween 20 concentration of 0.3 wt%, and reaction temperature at 60°C. In these conditions, the maximum value of Dg became 800% (4.2 mmol-ETSS/g-adsorbent) at the reaction time of 4 h, as shown in Fig. 1.

When the 50% ethanol aqueous solution was used as a saponification solvent, the saponification reaction of the ETSS-grafted adsorbent was finished within 2 h regardless of the Dg. After saponification for 2 h, the Ds reached 88%, 85%, and 82% (2.7, 3.4, and 3.7 mmol-SO₃⁻/g-adsorbent) for the Dg of 200%, 400%, and 700%, respectively. Additionally, when the deionized water was used, the marked saponification reaction occurred after 6 h, and the Ds reached over 80% at 18 h.

To evaluate metal adsorption performance of the ETSS-saponified adsorbent, the adsorption amount of Cu^{2+} onto the adsorbent was monitored and plotted in Fig. 2. The adsorption equilibrium of the ETSS-saponified adsorbent of 200% Dg was established within 80 min, and the adsorption capacity of that adsorbent was 1.4 mmol- Cu^{2+}/g -adsorbent. On the contrary, that of a commercially available particulate resin (DIAION PK216, adsorption capacity was 2.3 mmol- Cu^{2+}/g -resin), which has crosslinked network matrices, was not yet completely attained even after 480 min. Thus, the metal adsorption rate of the ETSS-saponified adsorbent is extremely higher than that of the DIAION PK216.

Reference

S. Aoki, K. Saito, A. Jyo, A. Katakai and T. Sugo, *Anal. Sci.*, **17 Suppl.** (2001) i205–208.



Fig. 2 Kinetic curves of Cu²⁺ onto the ETSS-saponified adsorbent and commercial particulate resin.

2-02 Preliminary Evaluation of Fibrous Boron Adsorbent with Groundwater

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1. Introduction

Boron is widely distributed in the environment as boric acid or borate salt. In Japan, the environmental standard for release of boron has been legally determined at 1 ppm (mg/l). Hence, JAEA tried to remove the boron from waste groundwater at the Mizunami Underground Research Laboratory (MIU), Tono Geoscience Centre, before discharge of the groundwater to the river. A granular resin of boron adsorbent has been used in MIU for boron removal. However, the granular resin adsorbent is effective only for boron removal at relatively slow flow rates around 10 bed volume (BV)/h .Therefore, JAEA's Takasaki site developed a new fibrous boron adsorbent¹ using a graft polymerization to realize the effective removal of boron. The boron adsorption from the waste ground water was preliminary evaluated using the fibrous boron adsorbent.

2. Experimental

The fibrous boron adsorbent was prepared by radiation-induced emulsion graft polymerization of glycidyl methacrylate onto non-woven polyethylene fabric and consequent chemical modification of the epoxy group to glucamine. To evaluate the performance of the boron adsorbent with groundwater at the MIU, the resulting adsorbents was packed into the column, 10cm in internal diameter. The volume of adsorbent was 2,000 cm³. This column was set into the experimental equipment which consisted of pumps and a fraction collector. The waste groundwater was pumped into the column at the flow rate of 20 BV/h. The boron concentration in groundwater was



Fig. 1 Elution equipment for boron adsorbent.

1.4 ppm. After the boron adsorption, the column packing boron adsorbent was carried to Takasaki site. Then, the adsorbed boron on the adsorbent was eluted with 1M HCl using less than 20 BV at a flow rate of 50 BV/h with the elution equipment in Figure 1. This equipment was assembled by column, pump and tanks. After elution of boron, the adsorbent can be used repeatedly for the treatment of waste groundwater.

3. Result and Discussion

Figure 2 shows the breakthrough curves of boron adsorption at a flow rate of 20 BV/h. The boron adsorption was carried out two times as preliminary experiments. The pH of groundwater were 9.8 and 10.4 for the first and the second adsorption, respectively. The adsorbent in the second run could treat more than 1,000 BV of groundwater until the boron concentration reached to 0.5 ppm, regulations in MIU. In contrast, the adsorption performance in the first run has breakthrough lower than 200 BV. This is because the boron capacity of the adsorbent dramatically decreases in the pH higher than 10. In addition, sludge was observed at the head of the column due to blasting excavations. These results indicate that the control the pH and the water quality by pre-treatment filtering are important for removal of boron from the groundwater with fibrous adsorption.

Reference

 H. Hoshina, *et.al*, "Synthesis of graft adsorbent with N-methyl-D-glucamine for boron adsorption", J.Ion Exchange, **18** (2007) 236.



Fig. 2 Breakthrough curves of boron adsorption at flow rate of 20 BV/h.

The first run (\bullet) and the second run (\bigcirc) .

2-03 Development of Water-Soluble Chelate Agent for Trapping-Heavy Metal (First Report)

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Water-soluble chelate agent for trapping heavy metal was synthesized by preirradiation graft polymerization of glycidyl methacrylate (GMA) on methylcellulose. The obtained water-soluble chelate agent having functional group of iminodiacetic acid had 2.65 mmol/g of lead-trapping capacity. However, its production cost is much higher than that of a conventional water-soluble chelate agent. Another water-soluble chelate agent was prepared by mutual grafting of acrylic acid onto starch of low price. In this grafting, a thickening agent was necessary to maintain the dispersion of starch powder in water. Lead-trapping capacity of starch based chelate agent developed was found to be 0.39 mmol/g. Whereas this value is still low compared with the GMA-based one, the cost-effectiveness has been improved drastically.

1. 研究の狙い

これまで、放射線グラフト重合技術により、ポリエ チレンや不織布を基材とした重金属捕集吸着材¹⁾とし て研究が行われてきた。一方、一般的に排水や飛灰中 の鉛などの有害重金属は、水溶性のジチオカルバミン 酸型キレート薬剤で不溶化され、凝集分離や固定化に より処理されている。そこで水溶性のキレート剤に置 き換え可能な、水に可溶で鉛が捕集可能な放射線グラ フト重合物の合成を検討した。

基材としては、生分解型かつ水溶性である素材を選定し、キレート剤使用時に有毒ガス発生の一因となる 硫黄成分を含まない官能基を、グラフト重合物に付与 することを条件とした。

2. 実験概要

(1)メチルセルロース型鉛捕集剤の調製

基材として選定したメチルセルロースに電子線を照 射した後、アルコールに溶かしたグリシジルメタクリ レート(GMA)をグラフト重合した後、イミノジ酢酸 を官能基として付与した。アルコール溶媒を用いた GMA 重合には、水分添加が不可欠と分かった。合成の 最適条件は、グラフト率が 40~60%の時で、イミノジ 酢酸基を付与後の鉛捕集容量は 2.65 mmol/g であった。

得られたキレート剤の捕集容量は、重金属処理剤として使用できるレベルではあるが、市販品に比べ費用 対効果の点から、実用化レベルではないと試算した。 (2)でんぷん型鉛捕集剤の調製

実用性を優先し、低原価素材による合成を検討する ため、でんぷんを基材として選定し、アクリル酸のグ ラフト条件を検討した。この組み合わせは吸水性ゲル の調製として知られているが、近年カルボキシル基付 与による重金属捕集能²⁾が注目されつつある。

でんぷんとアクリル酸の水懸濁液に、γ線(⁶⁰Co)を 5 kGy/h で1 h 照射する方法が、ゲル化および固形化せ ずに流動性を保つ好適な条件であった。でんぷん-アク リル酸水懸濁液の分散性を維持するためには、増粘剤 添加が必要であった。

でんぷん-アクリル酸型グラフト重合物のSEM 観察の結果、でんぷんの粒子同士がアクリル酸重合物で結合している様子が観察された(Fig.1)。

Fig. 2 に示すように、合成したキレート剤の鉛捕集能は 0.39 mmol/g と低いが、GMA を用いず 1 段階の反応 で直接官能基を付与することが可能であったことに加え、低原価素材を使用したことにより、費用対効果は メチルセルロース型の 3 倍に改善した。



Fig. 1 SEM micrographs of starch-graft-acrylic acid copolymer.



Fig. 2 Lead-trapping capacity of starch-acrylic acid graft copolymer.

3. 今後の方針

実用化可能な鉛捕集剤の調製方法を開発するため、 アクリル酸と同様なカルボキシル基を保有するモノマ ーの再選定を行って、鉛の捕集容量を向上させる。

- S. Shirako, et al., "Synthesis of the polymer to absorb heavy metals and its evaluation", 東京都立産業技術 研究所研究報告 第8号 (2005) 15-18.
- S. Keles, et al., "Competitive Removal of Heavy Metal Ions by Starch-Graft-Acrylic Acid Copolymers", Polym Plast. Technol Eng, USA (2006) 45,365-371.

2-04 Development of Millimeter-Wave Devices Using Grafted-PTFE Films

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As the importance of intelligent transport system (ITS) and imaging diagnostics increases, the fabrication of high performance millimeter-wave planar components becomes essential. This paper describes the development of high performance millimeter-wave planar components such as antennas using a low-loss fluorine material. The problems to be solved are the low degree of adhesion between copper foil and the fluorine material ¹⁻² and the shape of the antenna pattern. In order to solve the problems, surface treatment of fluorine films and a fabrication method using Electro Fine Forming (EF2) are utilized.

Polytetrafluoroethylene (PTFE) films were irradiated with electron beam, and different degrees of grafting were achieved by immersing the irradiated films for different grafting time in monomer solution. After the grafting, copper was sputtered on the surface of the films for measuring the peel strength between the metal and the grafted-films. As a result, a large increase in the peel adhesion strength was obtained by the graft polymerization using acrylic acid on the PTFE surface.

The processes of the EF2 fabrication technique include the lithography process and the electrotyping process. EF2 abilities are made possible by the integration of two unique technologies. One is a micro-lithography exposure technique used to enhance forming resolution and provide precise aperture patterns with micron level tolerances. Another is "Stay-Land Technology", which controls plating thickness and ensures an evenly distributed metal deposition so that parts are formed in a very controlled and uniform manner ³⁾. Figure 1 shows a millimeter-wave antenna prototype using EF2 fabrication technique.

We have designed an antenna pattern to improve antenna properties using CST Microwave Studio[®]. The fabricated antennas were based on commercially available conventional substrates and grafted-PTFE films for performance comparison. The millimeter-wave antenna array has 280 elements and the size of the antenna is about 88 mm x 64 mm. The near-field antenna radiation pattern was measured using a vector network analyzer (Anritsu / 37297C), while the far-field pattern is derived using conversion software. Figure 2 shows the calculated and measured characteristics of antenna patterns in the E-plane at a frequency of 76 GHz using 0.3 mm-thick grafted-PTFE films. The measurements are in good agreement with the calculation. In the measured antenna radiation patterns, the sidelobe level of the millimeter-wave antenna prototype was reduced in the E-plane by using grafted-PTFE films and

EF2 fabrication technique.

- 1) S. R. Kim: J. Appl. Polym. Sci. 77 (2000) 1913.
- S. Wu, E. T. Kang, K. G. Neoh, C. Q. Cui, and T. B. Lim: IEEE Trans. on Adv. Packag. 23, No.3 (2000) 538.
- L. Yang, N. Ito, C. W. Domier, N. C. Luhmann, Jr., and A. Mase: Proc. IEEE MTT-S Int., (2007) 1887.



Fig. 1 8-element beam steering antenna using EF2 fabrication technique.



Fig. 2 Concept of H-plane and E-plane (a) and calculated and measured characteristics of antenna radiation patterns in E-plane (b).

2-05 Radiation Polymerization of Low-Concentration Polyacrylamide Gel for Electrophoretic Separation of Proteins

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1. Introduction

Polyacrylamide gel is often used in electrophoresis as separation medium. In isoelectric focusing (IEF) separation, proteins were separated according to their isoelectric point (pI), and low-concentration gel is preferred because proteins can migrate fast through the relatively large pore of the gel. The authors developed low-concentration IEF gels supported by multifilament yarns¹⁾ and applied them to fast IEF separation of two-dimensional gel electrophoresis²⁾. However, lower concentration polyacrylamide than 2.5 %T (total monomer concentration; %T) is difficult to be obtained by the chemical polymerization process so far. In the present study, radiation polyacrylamide gel, and IEF separation performance was investigated.

2. Expermental

Monomer solutions containing acrylamide and N,N'-methylenebisacrylamide at 1.5, 2.0 and 2.5 %T and 0, 2 and 4 %C (cross-linker proportion; %C) were prepared. Multifilament yarns used as gel support were got through the glass capillaries (ID 1.15 mm). The monomer solutions degassed using ultrasonic and vacuum pressure were introduced into the capillaries. Co-60 gamma-ray was used as a radiation source and irradiated onto the monomer solutions through the capillaries at dose rate of 10 kGy/h. After polymerization, the gels were pulled out from the capillaries and provides as IEF media. Protein samples used for IEF were colored proteins: C-phycocyanin (MW 264 kDa, pI 4.3), hemoglobin (MW 67 kDa, pI 7.2) and cytochrome c (MW 13 kDa, pI 9.6).



Fig. 1 IEF separation of colored proteins using 2.0 %T, 0 %C polyacrylamide gel prepared by radiation polymerization process.

3. Results and Discussion

The 2.0 and 2.5 %T solutions were solidified at dose of 10 kGy (1.0 h irradiation) and the 0 %C solutions were also solidified and cross-linked by radiation polymerization, as well as 2 and 4%C solutions. The 1.5 %T solutions were solidified at dose of 15 kGy (1.5 h) and 20 kGy (2.0 h), but not at 10 kGy. The dose of 10 kGy was enough to polymerize the 2.0 and 2.5 %T solutions, but not for the 1.5%T solutions. IEF separation using the 2.0 %T, 0 %C polyacrylamide gel prepared by radiation polymerization process is shown in Fig. 1. The three colored proteins were separated along the gel in 20 min.

To evaluate IEF separation performance of the polymerized gels, migration distance of C-phycocyanin was monitored and plotted in Fig. 2. For comparison, the result using 2.5 %T, 5.4 %C polyacrylamide gel prepared by chemical polymerization process is shown in the solid line. The protein migrated fastest in the 2.0 %T, 0 %C gel polymerized at dose of 10 kGy. In the gels prepared at dose of 20 kGy, the protein moved almost at the same speed as the chemically polymerized 2.5 %T, 5.4 %C gel. This is considered because the excess dose of 20 kGy promoted polymerization and produced as small pores as the chemically polymerized 2.5 %T, 5.4 %C gel.

- J. Li, A. Ogasawara, T. Odake, T. Umemura and K. Tsunoda, *Anal. Sci.*, **20** (2004) 1673–1679.
- T. Odake, K. Kamezawa, K. Miyata H. Hotta, Y. Kojima and K. Tsunoda, *Proc. Total Analysis Systems* 2006 Symp., 1 (2006) 392–394.



Fig. 2 Migration distance of C-phycocyanin during IEF.

2-06 Absorption of Phosphate Ion in Swine Urine Using CMC Gel

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Rivers in Gunma Prefecture are sources of water supply to metropolitan areas. There are many swine households near the rivers and treated waste water from the swine households comes into the rivers. According to an inquiry by the Agricultural Production Bureau, Ministry of Agriculture, Forestry and Fishery, the biggest problem in the swine breeding is water pollution¹⁾. Reducing amount of phosphate ion and nitrogen coming into the rivers is expected for environmental conservation and high quality water supply. Meanwhile, shortage of phosphorus becomes a big concern in the world. Animal waste is a potential phosphorus source. Aiming to solve both problems, Gunma Prefecture started a project, "Prefecture Collaboration of Regional Entities for the Advancement of Technological Excellence" sponsored by Science and Technology Agency of Japan (JST). Industries, universities and governments join the project to realize clean Gunma and utilization of animal waste as resources. Each institution is in charge of different objective. Gunma Industry Support Organization and Japan Atomic Energy Agency collaborate to develop a gel to reduce the amount of phosphate ion contained in the effluent from the swine breeders using carboxymethyl cellulose (CMC).

CMC itself does not absorb phosphate ion, however, combination of CMC and metal ion can provide absorbent. Known as a preparation of CMC gel, multivalent metal ion forms ionic crosslinks among CMC molecules. Aluminum and iron ions are the most commonly used metal ions in the CMC gel preparation. A gel was formed instantaneously when CMC was mixed with ferric salt [hereafter expressed as Fe(III)] solution, however, it took time when ferrous salt [hereafter expressed as Fe(II)] aqueous solution was used.

Aqueous solutions of Fe(II) compounds are oxidized to



Fig. 1 Absorption of phosphate ion in artificial urine using CMC-Fe(II)-acid gel. ○:Gel prepared at 30 °C. ◇: Gel prepared at 50 °C.

those of Fe(III) compounds in the air. The oxidation proceeds gradually, however, we found CMC promoted the reaction. Immediately after the addition of CMC to aqueous solution of Fe (II) compound, the color of the solution changed from pale green to reddish brown.

We developed a novel method to prepare CMC gel using CMC and acid²⁾. Mixing CMC with acid resulted in a gel replacing sodium in CMC with hydrogen.

Combination of both reactions, oxidation and gelation, is expected to result in CMC-Fe gel which can absorb phosphate ion in swine urine effectively. Gels prepared from CMC, iron and water did not keep shape after absorbing phosphate ion, while, those prepared from CMC, iron and acid solution kept the shape even after phosphate ion absorption.

Gels prepared from CMC, Fe(II) and acid solution effectively absorbed phosphate ion in artificial urine and swine urine after primary treatment as shown in Figs. 1 and 2, respectively. The initial concentrations of phosphorus in the artificial and the swine urine were 457 and 78 mg/L, respectively. The concentrations decreased to less than 3 and 2 mg/L, respectively, after treatment by CMC-Fe(II) acid gel regardless of the species of acids. The values were less than regulation average value in a day eluted from swine breeders, 8 mg/L. Not only phosphate ion in the artificial urine but also phosphorus in the swine urine could be removed by the gels.

- 1) http://www.maff.go.jp/chikukan/1.issue.1.pdf
- M. Takigami, H. Amada, N. Nagasawa, T. Yagi, T. Kasahara, S. Takigami and M. Tamada, *Trans. Mater. Res. Soc. Jpn.*, **32** (2007) 713-716.



Fig. 2 Removal of phosphate ion in swine urine after primary treatment using CMC-Fe(II)-acid gels.

2-07 Preparation and Characterization of CMC-Konjac Mannan Mixture Gel

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Carboxymethyl cellulose (CMC) is a water soluble polysaccharide having various application fields such as civil engineering, oil drilling, fish feed, food additives, pharmaceuticals, and textile printing. CMC forms a gel when mixed with acid as a result of replacement of sodium in carboxymethyl group with hydrogen. If CMC can form mixture gel with other material in the presence of acid, the gel shows different properties with CMC gel.

Konjac mannan (KM) has high molar mass and high viscosity in low concentration aqueous solution in nature. The molar mass of KM can be easily decreased by γ -irradiation.

Molar mass of KM will have a big effect on CMC-KM mixture gel. The aims of this study are to examine mechanical properties of CMC-KM mixture gel and elucidate the effect of KM molar mass on characteristics of CMC-KM mixture gels.

CMC-KM mixture paste was prepared by mixing 8 g of CMC with 92 g of the KM aqueous solution containing 2 g KM using a mixer. Then the mixture was pressed to a sheet with the thickness of 1 mm. CMC-KM mixture gel was obtained by immersing the mixture paste in 0.5 M hydrochloric acid. Immersion time was 8 h, 16 h, 32 h and 64 h. Mechanical properties of the gel were measured using a compact table-top universal material tester and then Young's modulus and elongation were determined.

The Young's modulus of the CMC-KM mixture gel increased with increasing of the immersion time and showed a maximum at 16 h then decreased with the immersion time. The CMC-KM mixture gel is more elastic than the CMC gel. The strength of CMC-KM mixture gel is quite high as compared with that of the CMC gel. The reason of the maximum strength at 16 h immersion is estimated as follows: the CMC molecules shrink moderately when immersed for 16 h, however, CMC and KM molecules distribute uniformly. But CMC molecules shrink more after 32 h and 64 h immersion, CMC and KM



Fig. 1 The schematic model for gelation of CMC-KM mixture gel in acid.

molecules separate, and they become weaker than those immersed for 16 h.

Influence of molar mass of KM on gel property were investigated using CMC-KM mixture gels made from CMC and γ -irradiated KM with various molar mass by acid treatment. The CMC-KM mixture gel was more elastic than the CMC gel when molar mass of KM was higher than 1,000 kDa. But the Young's modulus of the CMC-KM mixture gel was almost the same with that of the CMC gel when molar mass of KM was lower than 500 kDa. Figure 1 shows the schematic model for gelation of the CMC-KM with high molar mass mixture gel. In the early stage, KM molecular chains might entangle with the loosely aggregated CMC molecules and the interaction such as hydrogen bonding could be occurred between the KM and CMC chains. The strength of the CMC-KM mixture gel should be increased by the synergistic effect.

When the immersion time increases, the CMC chains coagulate tightly by themselves, and the KM chains are excluded from the mixture. Then the system changes to the inhomogeneous structure and the strength of the mixture gel decreases with the immersion time.

In the case of the low molar mass KM (Fig. 2), the KM molecule might disperse more homogeneously in CMC-KM paste because of the short chain length. The degree of swelling of the CMC-KM paste in aqueous solution was higher than that of the high molar mass KM. This means that the interaction between CMC molecules and KM molecules with low molar mass is weak. When the paste is immersed in acid, the CMC molecules coagulate and make a network structure, while KM chains remain as the polymer with interpenetrating less interaction. Accordingly, the remarkable synergistic effect hardly appears, and the properties of the mixture gel show the similar properties of the CMC gel.



Fig. 2 The schematic model of CMC-KM mixture gel prepared by different molar mass KM.

2-08 Preparation of Blend Hydrogels Based on Cellulose- and Starch-Derivatives and Their Properties

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It is well known that hydrogels of carboxymethyl cellulose (CMC) and carboxymethyl starch (CMS) are produced by gamma or electron beam irradiation with their high concentrated aqueous solution¹⁾. The resulting biodegradable hydrogels have been applied in various fields such as medicine and agriculture. However, the hydrogels remain an issue concerning with water absorbability or mechanical strength in order to use as a super water absorbent in agriculture. In this work, blend hydrogels of CMC and CMS were synthesized by radiation crosslinking method to improve the properties such as a degree of swelling (Sw) and gel strength.

CMC and CMS were purchased from Daicel Co. Ltd, Japan and Gunei Chemical Industry Co. Ltd., Japan, respectively. The degree of substitution of carboxymethyl group of CMC and CMS were 1.34 and 0.12, respectively. The CMC/CMS aqueous solution of 40 wt.% (blend ratio of CMC/CMS: 100/0, 75/25, 50/50, 25/75, 0/100) were sealed in polyethylene-nylon bags after degassing. The irradiations were carried out using 60Co γ-ray source at the Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency. The samples of paste state were irradiated to total a dose of 1 to 40 kGy at room temperature to obtain blend hydrogels. Gel fraction of the obtained hydrogels was determined gravimetrically by measuring insoluble part after water extraction of sol. The Sw of the blend hydrogels was calculated from weight ratio, (Ws - Wd) / Wd, where Ws is the weight of the swollen gel and Wd is the weight of dried gel. The gel strength was measured by compressing CMC/CMS blend hydrogels cut into cylindrical form.





The gel fraction of CMC/CMS blend hydrogels increased sharply up to 10 kGy and reached a constant value at 30 kGy. Increase in the ratio of CMC increased the gel fraction of blend hydrogel. As a result of the measurement of swelling kinetics, these hydrogels reached the equilibrium swelling state from initial dried gel in about 2 hours at room temperature. Figure 1 shows the Sw of the CMC/CMS blend hydrogels swollen to the equilibrium state as a function of dose. The blend hydrogels with the maximum Sw of about 800 were obtained at only 2 kGy. The Sw decreased significantly with increasing the dose, and leveled off in the range of 20-40 kGy. At the dose range of 10 to 20 kGy, the CMC/CMS (25/75) blend hydrogels showed the highest Sw of 246 and 181 in the blend hydrogels, respectively.

The effect of the dose on the gel strength of the obtained CMC/CMS blend hydrogels is shown in Fig. 2. The gel strength of the blend hydrogels increased with increasing the dose, which was independent of the composition of CMC and CMS. As the ratio of the CMS increased from 0% to 50%, the gel strength at 10 kGy decreased to about 0.4 MPa. However, the further increase in the ratio of the CMS increased the gel strength, resulting in about 1.7 MPa.

From the above mentioned results, it was found that the CMC/CMS (25/75) blend hydrogel showed the both high Sw and high gel strength as compared with others. Therefore, that blend hydrogel at 10 kGy could be suitable to use as a super water absorbent for agriculture.

Reference

 F. Yoshii, et al., Nucl. Instr. and Meth. B, 208 (2003) 320-324.





2-09 Application of Ozone Decomposition Catalyst to Electron-beam Irradiated Toluene/Xylene/Air Mixtures

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Electron beam (EB) technology has an advantage for treating dilute environmental pollutants in off-gases due to high-density population of active species. When irradiated to air, various oxidizing species such as OH radicals and O atoms are produced from air components. In general, OH radicals play an important role of initiating the decomposition and removal of such pollutants. On the other hand, most of O atoms rapidly react with O₂ in air and produce inert O3. In our preliminary study using a few tens keV EBs, an O₃ decomposition catalyst was applied to an irradiated VOC/air mixture as a method for changing from inert O_3 into an active species¹⁾. This catalytic process enabled to oxidize the irradiation byproducts of VOCs into CO₂. In the present study, EB irradiation followed by the catalytic process (EB/catalytic process) was performed using 1-MeV EB under practical off-gas conditions such as lower initial VOC concentrations and multi-components VOCs/air mixtures to examine the effectiveness of this catalytic process to practical off-gases.

Air containing toluene and xylene at both initial concentrations of 10 ppmv was prepared at a flow rate of 5 L/min as sample gas for only EB irradiation and an EB/catalytic process. This sample gas was irradiated with 1-MeV EBs using a 1st accelerator and introduced into the layer of MnO_2 pellets, one of O_3 decomposition catalysts, heated to a temperature of 393 or 443 K. The volume of the catalyst was 21.5 cm³. The ratio of this volume to the gas flow rate was similar to that observed at practical usage of catalysts. The sample gas was analyzed by a FTIR system after 1 hr since the EB/catalytic process begun to examine the catalytic effect with keeping adsorption equilibrium.

The decomposition ratios of remaining toluene and xylene were obtained as a function of dose for only EB



Fig. 1 Decomposition ratios of toluene and xylene as a function of dose; ●: without catalyst, △: using catalyst at 393 K, and □: using catalyst at 443 K.

irradiation, EB irradiations with 393-K MnO_2 , and that with 443-K MnO_2 , as shown in Fig. 1. All decomposition ratio for EB irradiation using MnO_2 layers was 5–25% higher than that for EB irradiation. Another experiment demonstrated that toluene and xylene was not decomposed on such heated MnO_2 pellets. Furthermore, this decomposition ratio was obtained after adsorption equilibrium on the MnO_2 pellets was achieved. Therefore, the increase in the decomposition ratio with MnO_2 suggests that a portion of remaining toluene/xylene was decomposed by the catalytic process.

Product analysis results showed that EB irradiation produced CO₂ and CO through oxidation of toluene/xylene and their byproducts induced by active species. However, the catalytic process generated active oxygen from O₃ decomposition and subsequently produced only nontoxic CO₂ though oxidation of above organics and a portion of CO. Relative concentration of CO_2 and CO, η , in irradiated sample gases with and without using MnO₂ was obtained as a function of dose. (Fig. 2) The η under any conditions appeared to increase lineally with dose. The n for EB/catalytic process was 18-40% higher than that for EB irradiation due to the contribution of active oxygen produced on the MnO₂ pellet surface. These results indicate that the application of a catalytic process using O₃ decomposition catalysts enables the reduction in the dose of irradiation required for oxidation of VOCs and the byproducts into nontoxic CO2 under practical treatment conditions.

Reference

1) T. Hakoda et al., Radiat. Phys. Chem. 77 (2008) 585.



Fig. 2 Relative concentration of CO_2 and CO, η , in irradiated simple gases with and without using MnO_2 normalized to total carbon concentration of toluene and xylene before irradiation, as a function of dose.

2-10 Decomposition of Chloroethylenes in Air by Electron Beam

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Chloroethylenes are used for industrial solvents for dry cleaning and de-oil of metal, machine, and electric device¹⁾. They are released into the environment from the factory, and accumulated in the soil and groundwater. The accumulation has an influence on human and causes headache, nausea, dizziness and so on^{2} . Although electron beam (EB) is expected to be one of the useful methods for treating pollutants, it is difficult to treat chloroethyeles in the soil and water because of its very short range. Releasing chloroethylenes by aeration of the soil and water is required for EB treatment.

In general, kinds of chloroethylenes exist in the real soil and groundwater. Thus, it is important to do a basic research on the treatment of a mixture of chloroethylenes with electron beams. However, interaction among target chemicals after irradiation often makes it difficult to understand reaction mechanism in electron processing. In this study, trans-dichloroethylene (DCE), trichloroethylene (TCE), and tetrachloroethylene (PCE) in air were irradiated separately with electron beams. To compare the decomposition behaviors, the irradiation of a mixture of the chloroethylenes in air was also carried out.

Air containing 300 ppmv of chloroethylene (DCE, TCE, or tetrachloroethylene (PCE)) and 1 w% water was irradiated with EB at absorbed doses of 0.6-22 kGy. An accelerator was operated on 1 MV with a current of 0.22~8.7 mA. The irradiation was carried out at ambient temperature. The change in the concentration of each chloroethylene as a function of dose is shown in Fig. 1. The concentration decreased exponentially with dose. TCE and PCE were decomposed by about 90 %, but DCE is decomposed by about 50 %, at a dose of 2.8 kGy. From the



Fig. 1 Concentration of chloroethylene in air as a function of dose (irradiated separately).

initial slope of the curves in Fig. 1, the decrease in the chloroethylene concentration per 1 kGy was estimated to be 72, 285, 300 ppmv/kGy for DCE, TCE, PCE, respectively. These values are much larger than that of OH radical generated by EB, i.e., 10 ppmv/kGy. These results can be explained in terms that chloroethylenes were decomposed by a chain reaction by Cl radicals³⁾. Chloroethylenes were decomposed through reactions with OH radicals produced from the irradiation of air, and released the Cl radicals.

A mixture of 300 ppmv trans-DCE, TCE, and PCE was irradiated under the same condition as done separately. The change in the concentration of each chloroethyelene as a function of dose is shown in Fig. 2. The concentration decreased exponentially with dose. DCE and TCE were decomposed by about 60 %, but PCE was decomposed by about 40 % at a dose of 2.8 kGy.

Compared to the results of chloroethylenes irradiated separately, DCE in a mixture was readily decomposed. On the other hand, TCE and PCE in a mixture required much energy to obtain more than 90 % decomposition. It may be explained that DCE had a higher reactivity against the Cl radicals generated from chloroethylene decomposition than TCE and PCE. The amount of Cl radicals was abundant in the irradiation of a mixture of chloroethylenes.

- 1) http://www.eic.or.jp.
- 2) http://www.jaish.gr.jp/anzen/gmsds/0161.html.
- T. Hakoda, Z. Zhang, S. Hashimoto, *Radiat. Phys. Chem.*, Vol.54 (1999) 541.



Fig. 2 Concentration of chloroethylene in air as a function of dose (irradiated in a mixture).

2-11 Decomposition of Biologically Persistent Pharmaceuticals by Ionizing Radiation

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Introduction

There has been growing studies about the influence of pharmaceuticals on aquatic environment in the recent years. The pharmaceuticals are classified into emerging environmental contaminants that are extensively and increasingly being used in human and veterinary medicine. They have been detected in drinking water in the range of the concentration from ng to $\mu g / L^{-1}$. Some pharmaceuticals are persistence toward the activated sludge system. Long-term exposure of pharmaceuticals to the human body might have an influence on our physiological system. Treatment of pharmaceuticals should be investigated to minimize the long-term risk. Addition treatment methods such as advanced oxidation technologies (AOTs) were required to treat the secondary effluent of existing wastewater treatment plant with the activated sludge system. Ionizing radiation, one of AOTs, can treat a trace amount of persistent chemicals in water because it produces hydroxyl homogeneously^{2,3)}. radicals quantitatively and Decomposition of pharmaceuticals was investigated by ionizing radiation.

Experimental

Aspirin, ibuprofen, carbamazepine, and oseltamivir were selected as representative pharmaceuticals and dissolved in pure water. Activated sludge was obtained from wastewater treatment plant near our institute. The aqueous pharmaceutical solution was mixed with the activated sludge solution, which was pored into the glass bottle and stirred under air saturated condition. The γ -ray irradiations of the aqueous pharmaceutical solutions saturated with air were carried out using ⁶⁰Co γ -ray sources. The aqueous pharmaceutical solutions before and after biodegradation and γ -ray irradiation were analyzed with HPLC-MS.

Results and Discussion

Pharmaceuticals were treated with the activated sludge to confirm their biodegrability as shown in Fig. 1. Aspirin and oseltamivir in water were easily decomposed 2 hours, for which was half reaction time compared to that of ibprofen. Carbamazepine was not decomposed by the activated sludge system, which is consistent with the previous report¹⁾.

Ibprofen and syucarbamazepine at 1 mg/L in water were decomposed by γ -ray irradiation as shown in Fig. 2. Each concentration was exponentially decreased and they were eliminated at 120 Gy. The ionizing radiation could treat ibuprofen and carbamazepine which were hard to be decomposed by activated sludge system.

- 1) A. Fent, et al., Aquatic Toxicology, 76 (2006) 122-159.
- N. Getoff, Radiat. Phys. Chem., 65 437-446 (2002) 437-446.
- 3) A. Kimura, et al., *Radiat. Phys. Chem.*, 76 (2007) 699-706.



Fig. 1 Biodegradation of pharmaceuticals in water.



Fig. 2 Decomposition of ibprofen and carbamazepine in water by γ-ray irradiation.

2-12 Chemical and Biological Assays of Aqueous Chlorophenol Solutions Irradiated with γ-rays

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Introduction

Ionizing radiations can make reactive species in water, and are applicable to a waste water treatment. We have so far done the treatment research of water containing Endocrine Disrupting Chemicals (EDCs) by ionizing radiations^{1,2,3)}. 17 β -estradiol has the highest estrogen activity and is released from human and domestic animals. Nonylphenols are persistent and toxic artificial chemicals. They were decomposed by γ -ray irradiation. The biological activities of these aqueous solutions were evaluated by the enzyme-linked immunosorbent assay and the yeast two-hybrid assay before and after irradiations. These assays are based on the response of the endocrine system. In order to apply the ionizing radiations to the waste water treatment, a biological assay for the acute toxicity is also important because real waste water contains not only EDCs but also acute toxic chemicals. Organic chlorine compounds, for example, dioxin and PCB, have the toxicities but have been investigated mainly by the chemical analysis. 2- and 4-Chlorophenols were selected as first targets since they are model compounds of the organic chlorine compounds.

Experimental

2- and 4-Chlorophenols were dissolved in water to prepare samples at the concentration of 1 mmol L^{-1} . The aqueous chlorophenol solutions were poured in glass vials and bubbled with oxygen during irradiation. The γ -ray irradiations were carried out at 298 K using ⁶⁰Co sources in JAEA/Takasaki A HPLC (Agilent 1100 series) with a reverse phase column (Shodex RS pak DE-613) was used for the qualitative and quantitative analyses of the aqueous chlorophenol solutions before and after y-ray irradiation. The acute toxicity of the aqueous chlorophenol solutions before and after irradiation were evaluated by a biological assay using luminescence bacteria (photobacterium phosphoreum). The bacteria was exposed to chemicals and incubated in the culture medium. The light emitted from the bacteria measured with а luminometer was (Luminesencecer-JNR, AB-2100).

Results and discussion

2-Chlorophenol (2CP) was exponentially decomposed in water by γ -ray irradiation as shown in Fig. 1. Chlorohydroquinone and 3-chlorocatechol were formed as primary irradiation products by a low-dose irradiation (Scheme 1), and decomposed by a further irradiation. The



Scheme 1 Reaction scheme of 2-chlorophenol in water under oxygen saturated condition by γ-ray irradiations.

aqueous 2CP solution has a weak acute toxicity before irradiation. The toxicity of the solution, however, increased monotonously with dose. The solution irradiated at the dose of 8 kGy, at which 2CP and the primary irradiation products were almost decomposed, has a stronger acute toxicity than that before irradiation. The toxicity increased by irradiation was larger than the toxicity expected from the sum of the toxicities of 2CP and the primary products. The aqueous 4-chlorophenol solution has the toxicity before irradiation, and the toxicity increases with dose like 2CP. The increment of the acute toxicity can be considered to be caused by not only the primary irradiation products but maybe hydrogen peroxide and organic acids, and/or the synergistic effect of toxic irradiation products.

- A. Kimura, et al., Radiat. Phys. Chem., 69 (2004) 295-301.
- 2) A. Kimura, et al., Radiat. Phys. Chem., 75 (2006) 61-69.
- A. Kimura, et al., Radiat. Phys. Chem., 76 (2007) 699-706.



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3-64 3-65 3-66 3-67	 Asbestos Fibers <i>in vitro</i> 120 Y. Shimizu, K. Dobashi, T. Kusakabe, T. Nagamine, S. Matsuzaki, T. Ishizuka, M. Oikawa, T. Satoh, J. Haga, T. Ohkubo, Y. Ishii, T. Kamiya, K. Arakawa and M. Mori The Optimum Condition and Present Situation of the Analysis of Boron Micro-distribution in Tumor Cells Using PIGE
3-01 Discovery of a Novel Key Protein in the Radiation Response Mechanism of *Deinococcus radiodurans*

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Deinococcus radiodurans is characterized by an exceptional capacity to withstand the usually lethal effects of DNA-damaging agents including ionizing radiation, UV light and desiccation. Analysis of DNA-damage-sensitive strains of *D. radiodurans* has identified a novel regulatory protein, PprI¹). PprI is involved in regulating the induction of PprA, a DNA repair promoting protein²).

In a previous study, we identified the radiationresponsive minimal promoter region of the pprA gene and demonstrated that upregulation of pprA expression by PprI is triggered at the promoter level³⁾. PprI seems to be a membrane-bound protein, and this suggests that the effect of PprI on PprA induction is indirect, and that there may be yet to be identified components of the PprI-dependent response to radiation stress in D. radiodurans that are downregulated by PprI following irradiation. In an effort to explore this possibility, we compared the twodimensional protein profiles of wild-type and pprI disruptant strains that had been exposed to γ -rays followed by post-incubation. Over 300 protein spots were resolved on Coomassie Brilliant Blue-stained gels. Of those, some protein spots were demonstrably larger in the pprI disruptant strain, and were successfully identified by MALDI-MS spectra measurements and database searches. To determine whether the identified proteins are responsible for the radiation resistance of D. radiodurans, gene disruptant strains were generated and survival rate was As the result, dr0907 disruptant strain examined. exhibited markedly higher sensitivity to γ -rays than the wild type. This result suggests that DR0907 plays an important role in the *pprI*-dependent radiation response in D. radiodurans.

Furthermore, to determine whether the regulation of PprA induction amidst disruption of *dr0907* takes place at the transcriptional level, a luciferase reporter assay was employed using *D. radiodurans* strains carrying a reporter plasmid in which the radiation responsive promoter of *pprA* had controlled expression of a luciferase reporter gene (Fig. 1). The result suggested that DR0907 downregulates expression of the *pprA* gene at the promoter level under normal growth conditions and is involved in modulating the radiation response of *pprA*. Thus, DR0907 was designated PprM (for a modulator of PprA). This study strongly suggests that PprM plays an important role in the induction of PprA and is involved in the unique radiation response mechanism controlled by PprI (Fig. 2).



Fig. 1 Relative reporter activity of wild-type (WT), *dr0907* disruptant and *pprI* disruptant *D*. *radiodurans* strains. Following a post-irradiation incubation for 0 h (open boxes) or 2 h (filled boxes), luciferase reporter activity was measured.



Fig. 2 Hypothesis detailing the PprI-dependent signal transduction pathway in *D. radiodurans*.

- Y. Hua et al., Biochem. Biophys. Res. Commun. (2003) 306.
- 2) I. Narumi et al., Mol. Microbiol. (2004) 54.
- 3) H. Ohba et al., Gene (2005) 363.

3-02 Functional Analysis of the Low-fidelity DNA Polymerase AtREV1

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During the course of our study on UV-resistance mechanisms in higher plant, we have isolated a novel gene *AtREV1*, which encodes a member of Y-family DNA polymerases. The Y-family polymerases share common features, i.e., low fidelity and low processivity, and are thought to bypass various DNA damage. Our previous study showed that the *AtREV1*-deficient plant was more sensitive to UV-B or cross-linking reagents than the wild-type plant, but it was only slightly sensitive to γ -ray exposure than the wild type¹. Thus, it has been suggested that the *AtREV1* is involved in the bypass of UV- or other kinds of DNA damage.

To study the function of AtREV1 biochemically, here we expressed AtREV1 protein in *E. coli* and analyzed its enzymatic activity *in vitro*. The recombinant AtREV1 protein was purified by affinity and ion-exchange column



chromatographies to near homogeneity (Fig. 1). Then, the deoxynucleotidyl transferase activity was examined by using a 32 P-labelled oligonucleotide primer annealed with five different templates. As a result, AtREV1 performed quite low-fidelity replication and preferentially inserted a dCMP regardless of the template base (Fig. 2). Moreover, the AtREV1 inserted a nucleotide at the opposite of base-loss damage (AP), which is generated spontaneously or induced by DNA-damaging agents. In contrast, AtREV1 hardly inserted a nucleotide at the opposite of UV-induced damage ².

We next examined whether the low fidelity replication activity of AtREV1 causes mutations in vivo. A point-mutated uidA gene, as a reporter, was introduced into the wild type or AtREV1-deficient plants. The plants were treated with UV-C or γ -rays and reversion events, observed as blue GUS+ sectors, were counted. As a result, the UVγ-ray-induced reversion frequencies in or the AtREV1-deficient plants were significantly lower than that in the wild-type plants. These results suggest that AtREV1 is involved in the mutagenic replication of UV- or γ-ray-induced damage in vivo.

Although the recombinant AtREV1 failed to bypass the UV damage, the lower UV-induced mutation frequency in the *AtREV1*-deficient plants suggests the involvement of AtREV1 in the process of UV-damage. This inconsistency would be explained by the hypothesis that the AtREV1 co-operates with other protein(s), such as AtREV3 or AtPOLH³, to replicate UV-induced damage in Arabidopsis.

References

- 1) S. Takahashi et al., Plant Phys. 138 (2005) 870.
- 2) S. Takahashi et al., Plant Phys. 145 (2007) 1052.
- 3) H.J. Anderson et al., Plant J (2008) in press.



Fig. 1 Purification of recombinant AtREV1 by column





3-03 Analysis on Optimum Irradiation Condition for the Ion Beam Breeding

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Mutation breeding is one of the valuable breeding techniques that can produce various new phenotypes in plant and bacteria. Recently, ion beams have been used for mutation breeding. Ion beams have high liner energy transfer (LET) comparing with γ -rays. The mutagenic effects of ion beams and γ -rays have been studied in many organisms. In plant, Shikazono et al., who investigated the mutation events occurring on Arabidopsis chromosomal DNA with carbon ions, have reported the high incidence of chromosomal rearrangement^{1,2)}. However, analyses on ion-beam induced mutations in plants have been hardly proceeded because there was no efficient mutationdetection system as in bacteria and animal³⁾. It is essential to characterize the types of mutations induced by ion beams for the promotion of ion beam breeding. In this study, we analyzed mutation spectrum induced by ion beams and γ -rays by using *rpsL* transgenic *Arabidopsis* (Arabidopsis/rpsL) mutation detection system⁴⁾. In this system, a plasmid containing rpsL gene is integrated in Arabidopsis chromosomal DNA and this plasmid is collected by plasmid rescue after treatment of mutagen. Mutated rpsL clone is then screened in Escherichia coli. Since intact plasmid confers Sm^s and Km^r on Sm^r E. coli, plasmid-borne rpsL mutations can be easily detected from Sm^r and Km^r E. coli transformants. Our purpose of this study is to compare the mutation spectrum induced by ion beams and γ -rays and to estimate the efficient irradiation condition for ion beam breeding.

Arabidopsis/rpsL seeds were irradiated with 140 Gy of 220 MeV carbon ions and 740 Gy of γ -rays. Compared with background, mutant frequency was increased by 2.8 and 3.3 times with carbon ions and γ -rays, respectively (Table 1). Both radiations efficiently induced deletion mutations as reported in bacteria and animal^{5, 6)}. Frameshift mutation was the most frequent mutation occurred in γ -rays while this type of mutation was few in carbon ions (Fig. 1). The carbon ions induced G:C to A:T transition and complex type of mutation more frequently than γ -rays. These differences may result from different types of DNA damage formed by carbon ions and γ -rays.

Furthermore, we analyzed mutation induced by carbon ions near the Bragg peak in Arabidopsis/rpsL seed. The ion beams in the region near the Bragg peak comprise almost stopping ions and have higher LET than penetrating ion. From this experiment we will estimate the effect of LET strength on mutation induction in plant. Our preliminary data indicated that mutant frequency was seldom increased by exposure to 64 Gy of carbon ions near the Bragg peak (Table 1). We chose 64 Gy as the dose that gives the same killing effect as 140 Gy of penetrating carbon ions and 740 Gy of γ -rays, based on the dose response curve. The fluence of carbon ions near the Bragg peak would decrease by 1/4 because it has higher cell killing effect. In our irradiation condition, the fluence of carbon ions near the Bragg peak might be too low to induce mutation efficiently in the *rpsL* gene of 375 bp in length. To detect mutation induced by high LET ion beams, an alternative mutation detection system that can detect mutation occurred in longer region might be eligible. Further study is needed to elucidate the effect of LET strength on mutation induction.

Acknowledgement

We would like to thank Prof. Takimoto, Yamaguchi University, for providing Arabidopsis/rpsL seeds.

References

- 1) N. Shikazono et al., Genes Genet. Syst. 73 (1998) 173.
- 2) N. Shikazono et al., J. Exp. Bot. 56 (2005) 587.
- 3) T. Nohmi et al., Mutat. Res. 455 (2000) 191.
- 4) R. Yoshihara et al., J. Radiat. Res. 47 (2006) 223.
- 5) C. Miles et al., Mutat. Res. 227 (1989) 97.
- 6) Y. Kagawa et al., Mutagenesis 14 (1999) 199.

Table 1 Mutant frequency.

	Mutant Frequency
Background	3.3×10^{-5}
Carbon ion (220 MeV)	9.3×10^{-5}
γ-rays	1.1×10^{-4}
Carbon ion (Bragg peak)	4.3×10^{-5}

Mutant frequency is indicated as the ratio of mutants to total rescued clones.



Fig. 1 Mutant frequency of each type of mutation. Mutant frequency is indicated as the ratio of mutants to total rescued clones. Statistical analysis is performed by the method based on Poisson distribution.

3-04 Study on Molecular Mechanism of High-LET Carbon Ion Beam Induced Mutations in *S. cerevisiae*

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1. Introduction

The ion beams are widely used as the mutagen for developing new varieties of plants and also used for cancer therapy¹). However, precise effect of ion beams to DNA molecules has not been elucidated.

This study is intended to elucidate the molecular mechanism of the mutagenesis caused by ion beams. The budding yeast *Saccharomyces cerevisiae* was used as a model eukaryotic organism to comprehend the molecular mechanism of the mutagenesis caused by ion beams.

We reported that the main mutations induced by high-LET carbon ion were transversions last year²). In this report, we describe the in vivo mutagenecity of 8-oxodGs caused by ion beam irradiation in *ogg1* mutant strain, which is deficient in DNA glycosylase activity and thereby is not capable to remove 8-oxodG³).

2. Experimental procedures

S. cerevisiae strains used in this study were *S288c* (*RAD*⁺) and *ogg1*. The yeast cells were irradiated with carbon ions (${}^{12}C^{5+}$; 220 MeV) with the dose 100 Gy, and LET is 107 keV/µm. Carbon ion beams were generated from AVF cyclotron in JAEA. The mutation rates following irradiation were determined on the basis of colony-forming ability. And, selection of *ura3* cells was accomplished by plating the cells on the media containing 5-fluoroorotic acid.

The mutation sites of ura3 mutants were determined by

DNA sequencing.

3. Results and Discussion

The frequency of *ura3* mutation at 100 Gy in the *ogg1* was increased to 3.8×10^5 , 2-fold higher than the *wild* type.

Figure 1 shows the mutational spectrums and Figure 2 shows the summary of the sequence analyses. Our results indicate that the types of base changes in the carbon-ion induced mutants in wild type cells included GC to TA transversions (41%), the other type of base substitution (41%) and deletions/insertions (18%). In the case of ogg1, GC to TA transversions were largely observed (70%), but neither deletion mutation nor insertion mutation was observed. GC to TA transversions are known to be caused by misincorporation of 8-oxodG to DNA. Therefore, this result probably implies that the concentration of 8-oxodG in ogg1 mutant cells is higher than in wild type cells, since 8-oxodGs produced by ion beam irradiation are removed less efficiently in ogg1 mutant cells than in wild type cell. Our results suggest that the mutations by ion beams were mainly caused by the production of 8-oxodG in the nucleotide pool of the irradiated cells.

- 1) A. Tanaka et al., J. Exp. Bot. 53 (2002) 683.
- 2) Y. Matuo et al., Mutat. Res. 602 (2006) 7.
- 3) S. Boiteux et al., Biochimie 81 (1999) 59.





Fig. 1 Mutation spectrum induced carbon-ion beams irradiation of yeast.

Fig. 2 Mutation class of wild and ogg1.

3-05 Mutation Induction in Orchids Using Ion Beams

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Orchid is the main export for Malaysia's floriculture industry. Due to the increasing demand for orchid pot plants and cut flowers, new commercial varieties with new characteristics such as colour, shape and longer shelf life of flower are needed. The use of hybridization for orchid breeding is restricted by sexual incompatibility, sterility problems and long breeding time. Mutation induction by ion beam provides an alternative for improvement of orchids. In this study, we tried to produce mutants with new flower colour, shape and shelf life.

The protocorm-like bodies (PLBs) of two orchid species; *Dendrobium crumenatum* (Fig. 1) and *Dendrobium mirbellianum* (Fig. 2) were placed on 6-cm sterile petri dishes containing half strength MS media ($\frac{1}{2}$ MS)¹). The samples covered with a Kapton film were irradiated with 320 MeV ¹²C⁶⁺ ion beams at various doses ranging from 0 to 12.0 Gray (Gy) from the TIARA AVF cyclotron (JAEA, Takasaki). The irradiated PLBs were transferred onto fresh ¹/₂ MS and incubated at 25±2 °C under 12-hour photo period for proliferation. Subsequently, the cultures were transferred onto fresh media every four weeks for multiplication and regeneration. Plantlets were allowed to proliferate and multiply for several months before being hardened in glasshouse.



Fig. 1 Flowers of Dendrobium crumenatum.



Fig. 2 Flowers of Dendrobium mirbellianum.

In glasshouse, plantlets of *D.mirbellianum* were found to grow slower as compared to *D.crumentum*. Unlike irradiated *D.mirbellianum*, some *D.crumenatum* plants have been

already flowering. The length of blooming period was recorded. No extension of blooming period was observed in irradiated plants as compared to the controls, which bloom for only one day. Details on morphological changes observed in flowering mutant plants are given in Table 1, whilst variations on the flower sizes are shown in Fig. 3. One of the 6.0-Gy irradiated plantlets shows an increase in flower width. The flower measures 55 mm across as compared to that of the control, which is approximately 49 mm (data not shown).

A plant in the 0.2-Gy population exhibited a longer flower stalk which measures at 31.2 cm in length, approximately double the length of the flower stalk of the control which is averaged at 15 cm. More tissue culture plantlets were transferred from time to time into the glasshouse for screening. Plantlets of *D. mirbellianum* are growing very slow and have not reached flowering stage.

Table 1Morphological variations of flowering *D.crumenatum*plants irradiated by ion beams.

Dose	Number of	Number		/	Plant form		
(Gy)	plant	mutant	Flower size	Long stalk	Different shape	Different orientation	Dwarf
0	32	0					
0.2	7	2	1	1			
0.4	6	0					
4.0	19	1				1	
6.0	2	1			1		
8.0	23	1	1				
10.0	10	2	2				
20.0	7	7					7



Fig. 3 Large flowers of *D.crumenatum* variations irradiated at 0 (left; control), 0.2 (center) and 8.0 (right) Gy.

Reference

 T. Murashige and F. Skoog, Physiol. Plant. 15 (1962) 473.

3-06 Ion Beam Breeding of Flower Color Variations in Transgenic Plants with Multi-Disease Tolerance

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1. Introduction

Ion beams have different effect from that of electron beams on mutation generation of crops and have great impact on plant seed and seedling business. New carnation variety series with 10 flower color variations have been developed by ion beam breeding in the joint R&D between Kirin Agribio Co., LTD. and JAEA. They have been commercialized in Japan and Europe¹⁾, and their wholesale market has amounted to 450 million yen per year.

We are trying to develop an advanced application system of ion beams to produce color variations or to add improvements in transgenic plants that have acquired useful characteristics such as tolerance to multi-diseases by recombinant DNA techniques.

2. Materials and Methods

Agrobacterium-mediated transgenic chrysanthemum (Dendranthema plants grandiflora) expressing double-stranded RNA-specific ribonuclease gene (pac1) derived from Schizosaccharomyces pombe were used as materials for ion beam breeding. Leaf segments were placed in petri dishes containing Murashige and Skoog medium supplemented with 0.05 mg/L NAA and 0.1 mg/L BA, 30 g/L sucrose and 7 g/L agar. The samples were irradiated with 5 Gy to 12 Gy of 320 MeV carbon ion beams from the TIARA AVF cyclotron (JAEA, Takasaki). Irradiated plants have been cultivated in the biohazard glasshouse and their flower color and shape were investigated.

3. Results and Discussions

Transgenic chrysanthemum plants expressing pac1 gene showed tolerance against Chrysanthemum Stunt Viroid (CSVd) (Fig. 1) and Tomato Spotted Wilt Virus (TSWV)²). In total, 832 plants from ion beam irradiation have been cultivated and flowered. Flower color mutants such as pale pink, dark pink, salmon, white, yellow etc. have been obtained (Table 1, Fig. 2, 3).

Table 1Flower color mutants obtained from ion beambreeding of transgenic chrysanthemum plants.

5Gy; Light Pink (4), Dark Pink (2), Bronze (2)

- 10Gy; Dark Pink (2), Salmon (2), White (1)
- 12Gy; White (1), Yellow (2)

Ion beam breeding is effective to produce color variations, once the transgenic plants with virus- and viroid-resistance have been acquired.

From the results we confirmed that the advanced

application system of ion beams will contribute to the development of variety series of ornamentals with high quality at lower cost, as compared with the method to introduce useful characteristics into several varieties with different flower color by transformation one by one.



Fig. 1 Symptoms by CSVd infection. (from left to right) Non-transgenic with CSVd infection, non-transgenic without infection, transgenic with infection, transgenic without CSVd infection.



Fig. 2 Flowering test in biohazard glasshouse of transgenic chrysanthemum plants with multi-disease tolerance irradiated with 320 MeV carbon ion beams.



Fig. 3 Flower color mutants obtained from ion beam breeding of transgenic chrysanthemum plants with multi-disease tolerance. Upper left; parent.

- M. Okamura et al., Floriculture, Oranamental and Plant Biotechnology Vol.I, Global Science Books (2006) p.619.
- 2) T. Ogawa et al., Breeding Sci. 55 (2005) 49.

3-07 Evaluation of Resistance to Bacterial Wilt and Morphological Characteristics in Regenerants Derived from Ion beam Irradiated Microspores of Eggplant Rootstock

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We evaluated 'Yoshikinasudai2gou' and 'Yoshikinasudai3gou' regenerated from ion beam irradiated microspores of 'Hiranasu' (*Solanum interifolium*) for their resistance to bacterial wilt caused by *Ralstonia solanacearum* and observed their characteristics. Their susceptibility to five bacterial groups classified by differential reaction in *Solanum* species, were similar to 'Hiranasu', which is resistance to bacterial group I and II and susceptible to group III, IV and V. The percentage of diseased seedlings was lower than 'Hiranasu' on the pots test, but all of the seedlings were died within three weeks in infected field test. The 'Yoshikinasudai2gou' had longer internode lengths and higher plant height, and the 'Yoshikinasudai3gou' showed larger fruit size compared to the 'Hiranasu'.

ナスに深刻な被害をもたらす青枯病に対して強度 の抵抗性を持ち、福岡県の促成栽培に適するナス台木 品種はなく、新品種の育成が急務となっている。

これまで、青枯病抵抗性ナス台木品種を短期間に 育成する目的で、促成栽培の台木に適するが青枯病IV 群菌³⁾に罹病性のヒラナス(*Solanum integrifolium*)を供 試し、その小胞子にイオンビーム(220 MeV ¹²C⁵⁺、 320 MeV ¹²C⁶⁺、各0.5、1、2 Gy)を照射して突然変異 を誘発した半数体倍加系統261個体を作出した¹⁾。そ れらから、ポット栽培での青枯病接種試験で'よしき ナス台 2号'、'よしきナス台 3号'の 2系統を選抜した ²⁾。

本実験では、選抜した2系統について、青枯病の5 つの菌群³⁾と多発圃場での抵抗性、さらに土壌病害の 半枯病と半身萎凋病に対する抵抗性および形態の観 察を行って、実用性を評価したので報告する。

Material and method

・よしきナス台2号、、・よしきナス台3号、の自殖により採種したM2世代の種苗を供試した。青枯病の菌株は福岡県農業総合試験場で保存していたもの、半枯病および半身萎凋病抵抗性検定の菌株は独立行政法人農業生物資源研究所ジーンバンク事業により配付されたものを用いた。接種は、いずれも増殖した菌液に断根して浸漬と生育中に土にかん注した。青枯病菌で汚染した圃場での抵抗性検定は、独立行政法人野菜茶業研究所で行った。形態特性は、1/2000ワグネルポットで栽培して、ヒラナスと比較した観察結果を記した。

Result and Discussion

ヒラナスは、青枯病の I とⅡ 群菌に抵抗性で、Ⅲ~ V群菌に罹病性である³⁾。 'よしきナス台 2号'、'よ しきナス台 3号'は、ポットでの菌接種検定において、 ヒラナスと同様にⅢ~V群菌に罹病した。発病株率は ヒラナスより低い傾向であったが、圃場検定では定植 後 3週間以内にすべて枯死した(Table 1)。この結果 から選抜した 2系統の青枯病抵抗性は、ヒラナスより わずかな向上が示唆されるものの、現場に普及できる ほどの強度な抵抗性はないことが明らかになった。近 年、ナスの青枯病抵抗性に関与する遺伝子の数は、品 種によって異なるものの、複数存在することが報告さ れている⁴⁾。強度の青枯病抵抗性を突然変異で作出す るには、有用な変異が複数の青枯病に関与する遺伝子 領域で必要と考えられ、突然変異体をさらに多く育成 して選抜するか、繰り返しイオンビーム照射と抵抗性 系統の選抜を行って変異を集積していく必要がある。

なお、2系統とも半枯病菌には抵抗性、半身萎凋病 菌には罹病性であり、ヒラナスと同様であった(デー タ略)。形態の観察では、ヒラナスに比較して、'よ しきナス台 2号'は節間が長く伸長旺盛であり、'よし きナス台3号'は果実が大きく採種量が多かった。

今回、強度の青枯病抵抗性品種は得られなかったが、 節間が長く伸長が良好な'よしきナス台 2号'は、低温 での生育を評価し、促成栽培で冬場の暖房費節減のた めの育種素材としての活用が期待される。

l'able 1	Percent	tage of c	liseased	seed	lings
by	Ralstonia	solanac	earum.		

	Inocu	On				
Varieties		0	n pots			infected
		11		IV	V	fields
	%	%	%	%	%	%
Yoshikinasudai2gou	0	0	42	67	42	100
Yoshikinasudai3gou	0	0	33	50	17	100
Hiranasu	0	0	50	83	75	100
Senryo2go	58	50	58	42	58	100
Torubamu•biga	0	0	0	17	0	100
Tsunonasu	0	50	92	67	0	-

- K. Takata et al., TIARA Annual Report 2004 (2005) 48.
- Y. Uchimura et al., JAEA Takasaki Annual Report 2005 (2007) 80.
- 3) 尾崎克己. 植物防疫 44 (1990) 291.
- 谷本秀夫・中曽根渡. 園学研(Hort.Res.(Japan)) 1 (2002) 103.

3-08 Evaluation of Mutants from Ion Beam Irradiated Microspores and Seeds of Eggplant (Solanum melongena L.)

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To get thornless eggplant, the microspore of eggplant 'AE-P11' (*Solanum melongena L.*), and the seeds of 'AE-P03', 'AE-P08' and 'AE-P11' were irradiated with 320-MeV carbon ion beams from AVF cyclotron. No thornless eggplant was regenerated. Other mutants were observed, such as deletion for purple colour, spots of purple colour and leaf viridis, until the first flower stage. The overall mutant frequencies were 0.8% in irradiated microspore and 0.6% in irradiated seeds, respectively.

ナスの萼や葉柄にあるトゲは、作業時に不快感があ るので、トゲがない品種が望まれている。このため、 種子および小胞子へのイオンビーム照射を組み合わ せ、トゲのない突然変異体の作出を図った。

これまでに、ナス系統'AE-P11' (Solanum melongena L.)の小胞子に、0.5 ~ 20 Gyの¹²C⁶⁺・320 MeVイオン ビームを照射し、259個体を作出した¹⁾。また、'AE-P03, 08, 11'の合計1,043粒の種子に50 Gyの¹²C⁶⁺・ 320 MeVイオンビームを照射し、生育した607個体の うち、439個体の自殖後代(M2)を採種した。

ナスのトゲは、単因子劣性形質である²⁾。小胞子は 半数性細胞であるため、イオンビーム照射当代で劣性 の突然変異を観察できる。種子は2倍体であるため、 劣性突然変異の出現はM2世代である。

そこで、トゲの有無やその他の可視的な突然変異に ついては、小胞子からの再生植物体の照射当代で、種 子439個体のM2世代、1系統1~15個体ずつ、合計3,937 個体で、第1花蕾まで調査した。

小胞子からの再生植物体を調査した結果、トゲのな い形質を持つ突然変異系統は得られなかった。その他 の変異体として、葉脈、葉柄、茎の紫色素の欠失した 変異体が0.5 Gy照射区から1個体、本葉のビリディス (黄緑化)が1 Gy照射区から1個体得られた(Table 1, Fig. 1)。一方、種子でも、トゲのない形質を持つ突 然変異体は得られなかった。その他の変異体として、 子葉のアルビノ(白化)が3個体、子葉のキサンタ (黄化)が4個体、本葉のビリディスが1個体、葉脈、 葉柄、茎、果実の紫色素の欠失が1個体、葉や果実で

紫色素の斑が現れる突然変異体が1個体得られた (Table 1, Fig. 1)。

突然変異の誘発頻度は、生育途中で枯死したアルビ ノ、キサンタを除き、本葉のビリディス、紫色素の欠 失、斑からみて、小胞子では259個体のうち2個体(約 0.8%)、種子では439個体のうち3個体(約0.6%)で あった(Table 1)。小胞子と種子における可視的な突 然変異の違いは、アルビノとキサンタ、紫色素の斑が、 小胞子からの再生植物体では見られなかったことであ る。このことは、ナスの半数性細胞にとって、アルビ ノとキサンタが致死的な傷害であるためと考えられた。 また、紫色素の斑の突然変異体が現れなかったのは、 調査個体数が少なかったためと考えられた。 今後、トゲのない突然変異体を作出するためには、 小胞子から大量に植物体を再生する技術のさらなる効 率化や、カルスを経由せずに直接不定胚を形成させ、 短期間に植物体を再生させる培養技術の開発が必要と 考えられた。

Table 1	Evaluation	of mutants	from	ion-beam	irradiated
micro	spores and s	eeds of egg	plant.		

Mutations	Regenerants from ion beam irradiated microspores	M1 seeds ¹⁾
Number of plants investigated	259	439
Thornless	0	0
Albino (cotyledon)	0	3
Xantha (cotyledon)	0	4
Viridis (leaves)	1	1
Deletion of purple colour (vein, petiole and stem)	1	0
Deletion of purple colour (vein, petiole, stem, calyx and fruit)	0	1
Spots of purple colour (leaves and fruit)	0	1
P	0.0	0.6

Percentage of mutations 1) Progeny test

Mutations with viridis, deletion of purple colour and spots of purple colour



Fig. 1 Mutants obtained from ion-beam irradiated microspores and seeds of eggplant.

A: Deletion of purple colour on vein, petiole and stem (Regenerant from microspores), B: Deletion of purple colour on vein, petiole, stem, calyx and fruit (M2 Seeds), C: Spots of purple colour on leaves and fruit (M2 Seeds). Bar indicate 1 cm.

- Y. Saiki et al., JAEA Takasaki Annual Report 2006 (2008) 70.
- S. Kuno et al., Res.Bull.Aichi Agric.Res.Ctr. 37 (2005) 29.

3-09 Development of Commercial Variety of Osteospermum by a Stepwise Mutagenesis by Ion Beam Irradiation

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Introduction

Osteospermum is one of the composite perennials, and characterized by a long flowering time. Recently, the production of Osteospermum has increased as potted plants and flowerbed materials. Besides the ordinary mating, bud mutation has been used for the breeding of new varieties. However, the development of new and more efficient mutagenesis techniques with an artificial manner is expected at production fields. Previously, we irradiated Osteospermum 'Mother Symphony' with ion beams, and selected 37 flower-color and morphological variants out of approximately 3,000 individuals¹). Among them, an unprecedented pastel color variant was applied to the variety registration as 'Vient Flamingo (tentative name)' in March 2007²⁾. Besides, many petal color variants such as white and orange could be obtained from irradiated 'Mother Symphony' that has yellow petals. Usually, Osteospermum has different colors in each side of the petals. Interestingly, irradiated included orangethe variants and vellow-isochroous variants. However, we could not obtain a white-isochroous variant that has a potentially high market value. Therefore, in an effort to obtain a variant with white-isochroous petals, we irradiated again the white petal variant OM7 of 'Mother Symphony' with ion beams. The leaf disc sections of re-irradiated variant OM7 exhibited higher sensitivity to ion beams than those of 'Mother Symphony'. For this reason, the optimized irradiation dose for the selection of variants from OM7 was determined to be 0.1 to 5 Gy^{3} . Then, we irradiated the ion beam to a lot of individuals on this condition to obtain the white-isochroous mutant

Materials and methods

(1) Preparation of re-irradiation material

Leaf sections (quadrilateral pieces with 0.5 to 1 cm in size) of the white variant OM7 of Osteospermum 'Mother Symphony' were put on 1/2 MS medium supplemented with 0.1 mg/L NAA and 1.0 mg/L BA. After irradiation and post-cultivation, the re-differentiated individuals were obtained.

(2) Selection of variants using the optimized dose

Leaf disc sections were irradiated with carbon ions $({}^{12}C^{5+}$ and ${}^{12}C^{6+})$ at a range of 0.1 to 5 Gy. After 24 hr, the leaf disc sections were transferred to fresh 1/2 MS medium supplemented with 0.1 mg/L NAA and 1.0 mg/L BA to induce adventitious buds. After about 1 month, grown adventitious buds were transplanted on fresh 1/2 MS medium, and cultivated to obtain plantlets. The resultant plantlets were acclimatized and then grown in a greenhouse

to investigate flower color and morphological changes.

Results and Discussion

The white petal variant OM7 was derived from irradiated 'Mother Symphony' that originally has yellow petals. Second irradiation of OM7 resulted in acquisition of variants such as purple and multi-petals that did not appear in first irradiation experiment, suggesting that re-irradiation induced additional mutations in pigment synthesis or morphogenesis pathways. Total 1,582 plants were raised until now this year, and 22 individuals that have variation in petal color, petal shape and plant type were selected (Table 1). In the color variation, white purple and light bitter orange mutants, and light yellow in the back side of the petal were obtained (Fig. 1). Two individuals (OM705, OM706) that show discoloration in the back side of the petal were selected for further analysis.

Table 1 Mutants in the white variant OM7 by the irradiation of $0.1 \sim 5$ Gy $^{12}C^{5+}$ and $^{12}C^{6+}$ ion beams.

Radiation	No. of hardening	No. of mutants		
¹² C ⁵	754	color	$6(0.8)^{z}$	
C	/34	plant type	4(0.5)	
1206	010	color	7(0.8)	
C.	828	nlant type	5(0.6)	

^zThe percent is shown in the parenthesis.

Color variation : white purple 5, light bitter orange 2, back of the petal is light yellow 2, others 4

Plant type variation: type of petal 5, dwarf 2, others 2



Fig. 1 Mutants in color of the white variant OM7 by the irradiation of ion beams.

- A : OM7(derived from irradiated 'Mother Symphony')
- B : white purple
- C: light bitter orange
- D : back of the petal is light yellow.

- 1) M. Iizuka et al., TIARA Ann. Rep. 2004 (2005) 50.
- 2) M. Iizuka et al., JAEA Takasaki Ann.Rep.2005 (2006) 81.
- 3) M. Iizuka et al., JAEA Takasaki Ann.Rep.2006 (2007) 71.

3-10 Ion Beam Breeding of Sugarcane Cultivar "Ni17"

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Sugarcane cultivar "Ni17" has hard hair on its leaf sheath. We have intended to induce hairless or few hair mutants using ion beam irradiation to cultured tissue of leaf explants. As a result of nursery selection, individual selection and line selection, we have selected one line among the 2,223 regenerated plants. This line has a few hairs on its leaf sheath, and its growth as well as yield is higher than "Ni17". However, its stalk diameter is thinner than "Ni17". These results suggest that ion beam breeding is effective in improvement of sugarcane cultivar "Ni17".

サトウキビ品種「Ni17」は株出萌芽性,風折抵抗 性に優れる¹⁾が、葉鞘部に着生する粗剛な毛群が収穫 や採苗時に手に刺さる等の問題があるため、我々はイ オンビームを利用した変異誘発によって毛群の少ない 「Ni17」優良系統の作出に取り組んでいる。これまで 我々は、個体再生率並びに圃場生育からみたイオン ビームの適正照射線量を明らかにしてきた^{2),3)}。今回、 変異誘発を行った個体群の中から毛群が「Ni17」に比 べて少なく、収量性も高い系統が得られたので、その 選抜経過並びに現地圃場での生育特性について報告す る。

「Ni17」の茎頂付近の幼葉切片に、日本原子力研 究開発機構高崎量子応用研究所のAVFサイクロトロン により発生させたイオンビーム (320 MeV・¹²C⁶⁺) を0~15 Gy照射し、カルスから不定胚を経由し2.223 個体の再生植物体を育成し、ポリポットで育苗した後、 幼苗時の毛群の発生程度を調査した。毛群の発生程度 が無~極少数であった517個体について、圃場での個 体選抜試験により30個体を選抜し、次年度、南西諸島 の徳之島現地圃場の系統選抜試験に供試した。現地で の系統選抜試験では、ほとんどの系統において、収穫 時の茎径が細くなるなどの傾向が観察されたが、供試 系統のうち毛群の発生が少なく、生育量が優れる3系 統をさらに選抜した。選抜3系統については、引き続 き現地圃場において、さとうきび栽培における一般的 な作型である春植栽培、翌年の萌芽株を利用する株出 栽培に供試し、毛群の発生程度と併せて詳細な収量調 査を行った。

2年間の特性調査の結果、有望な系統「KB04-25」 を選抜した。「KB04-25」は春植栽培において、毛群 の発生が「Ni17」に比べて少なくなっており、収量面 においては、茎径が細くなり1茎重は18%減少したも のの、茎長が15 cm長くなり、茎数が52%増加したた め、1茎重×茎数である収量は19%増加した(Table 1)。また株出栽培においては、「Ni17」に比べ毛群 の発生は、春植栽培と同様に少なくなっていた。収量 面についても春植栽培と同様に茎径が細くなったため に1茎重は14%減少したが、茎長は22 cm長くなり、 茎数も41%増加したため収量は21%増加した(Table 2)。今回選抜した「KB04-25」は、両作型において、 茎径が細くなり1茎重は減少する傾向であったが、茎 長及び茎数の増加が大きく、その結果収量が増加した。

一方、今回の選抜過程において、イオンビームの 照射は行わず培養系を経由した無照射区の個体の中に も、茎長及び茎数が増加する変異体が見出されており、 培養変異も増収効果の一因と考えられた。しかしなが ら、無照射区では毛群が減少した変異体は見つかって おらず、毛群が減少する系統を作出するにはイオン ビーム照射による変異誘発が重要であると考えられた。 以上のことから、組織培養とイオンビーム照射によ る突然変異誘発を組み合わせた本育種法は、毛群の減 少,茎長,茎数の増加等といったサトウキビの特性改 良に有効であると考えられた。しかし、茎の細茎化に ついては風折抵抗性が低下することが想定されるため、 今後検討が必要である。

Table 1 Characteristics of the selected line by spring planting(2007)

							· · · · ·
Line	Dose	hair	millable stalk length	stalk diameter	weight per stalk	number of millable stalks	weight of millable stalks
	(Gy)		(cm)	(mm)	(g/number)	(number/a)) (kg/a)
Ni17	_	many	155	26.2	853	688	586
KB04-25	5	few∼ moderate	170	23.2	703	1,042	730

Table 2 Characteristics of the selected line by rationing(2008)

1 4010 2 0	Jiiuiu	eteristies of	the set	cotou mi	e og fatool	11115(2000	·)
Line	Dose hair		millable stalk stalk diameter length		weight per stalk	number of weight millable millabl stalks stalks	
	(Gy)		(cm)	(mm)	(g/number)	(number/a)	(kg/a)
Ni17	_	many	220	23.8	964	833	805
KB04-25	5	few∼ moderate	242	21.6	831	1,177	977

- M. Takemure et al., 鹿児島県農業試験場研究報告32 (2004) 81.
- F. Tohjima et al., JAEA Takasaki Annual Report 2005 (2007) 83.
- F. Tohjima et al., JAEA Takasaki Annual Report 2006 (2008) 73.

3-11 Induction of Mutations Affecting Pollen Formation by Ion Beam Irradiation to *Lilium × formolongi* hort (cv. White Aga)

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We have been working on producing marketable blue lilies. It has been impossible to produce blue lilies by crossbreeding or mutation, because they lack flavonoid 3',5'-hydroxylase (F3'5'H), which is necessary for synthesis of the blue pigment, delphinidin. Therefore, we developed an Agrobacterium-mediated transformation system for lilies to transfer the F3'5'H gene¹⁾. For the marketing of the transformed blue lilies, we have to regulate the spreading of the pollen to prevent them from crossing with other lilies, because they are genetically modified plants. The ion beam has been shown to be useful to improve one property such as flower color, flower shape, flowering time etc. without inducing undesirable mutations together with them^{2),3)}. Therefore, we adopted the ion-beam irradiation to obtain male-sterile mutants of lilies⁴⁾. In this study, we regenerated the plants from the irradiated calluses and investigated the pollen formation.

Filaments of a lily cultivar "White Aga" were cultured on a callus proliferation medium. The cultures were maintained at 25 °C in the dark. The calluses induced from the filaments were maintained by subculturing monthly onto the same fresh medium under the same condition. The penetration range of 100 MeV ${}^{4}\text{He}^{2+}$ from an AVF cyclotron (JAEA) was 6.2 mm. The diameters of the induced calluses were about 10.0 mm or more. Therefore, we crushed the calluses to about 1.0 mm diameter by squashing on stainless steel sieves of 1.29 mm mesh (the mesh size of 16) and filtration through them. The crushed calluses were placed on a modified callus proliferation medium and covered with sterilized Kapton film (7.5 µm in thickness, 45 mm square in size, Toray-DuPont, Japan), and exposed at total doses of 0-2 Gy of 100 MeV ${}^{4}\text{He}^{2+}$ beam from the AVF cyclotron.

The exposed calluses were transferred to a regeneration medium and cultivated for 4 months. The calluses were maintained by subculturing every month onto the same fresh medium. Regenerated shoots were transferred to bulb propagation medium and cultivated for 10 months and maintained by subculturing every 2 months onto the same fresh medium. The culture was performed under a 16 hr light/8 hr dark cycle at 25 °C. The propagated about 2500 bulblets were incubated at 4 °C for 2–3 months and planted in a field in April 2006. We investigated the aberration of anthers and selected 10 R0 plants, which showed less pollen. In October 2006, the selected 10 R0 plants were planted and examined for the aberration of anthers again. One of them showed twisted anthers and less pollen than original "White

Aga" (Fig. 1). This mutant had been exposed at total dose of 0.8 Gy and showed normal appearance excluding the anthers.

This mutant was planted in field in October 2007. This mutant will be examined again for pollen fertility by staining and culturing of the pollen grains.



Fig. 1 Selected lily mutant exposed at total dose of 0.8 Gy.A: Original "White Aga".B: Selected lily mutant, which showed twisted anthers and less pollen.

- 1) Y. Hoshi et al., Plant Cell Rep. 22 (2004) 359.
- M. Okamura et al., TIARA Annual Rep. 2003 (2004) 51.
- 3) K. Ueno et al., TIARA Annual Rep. 2003 (2004) 53.
- M. Kondo et al., JAEA Takasaki Annual Rep. 2005 (2007) 89.

3-12 Mutation Induction on *Delphinium* and *Limonium sinuatum* Irradiated with Ion Beams

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To obtain mutants of *Delphinium* and *Limonium sinuatum*, we investigated the influences of ion beam irradiation. The survival rate of leaf brades on *Delphinium* and the growth of shoot on *L. sinuatum* irradiated with 320 MeV carbon ions decreased at 0.5 and 1.0 Gy, respectively. The suitable doses for mutation induction were estimated to be around 0.5-1.0 Gy for *Delphinium* and 1.0 Gy for *L. sinuatum*. In the future, we will continue to test for characteristic of flower colour, etc.

イオンビームは従来の変異原と比較して変異効果、 変異スペクトルが大きく、キクやカーネーションでは 花色や花形の変化、腋芽数の減少などが報告されてい る¹⁾。本研究ではデルフィニウムおよびスターチスシ ヌアータの花色などにおいて特徴のある変異体獲得を 目的としてイオンビーム照射を行った。

カネコ種苗㈱育成のデルフィニウム'7P'の培養体の 葉身および、スターチスシヌアータ'P08'の花穂の節を 60 mmシャーレに置床し約1週間後に照射を行った。イ オンビーム照射は日本原子力研究開発機構、イオン照 射研究施設(TIARA)のAVFサイクロトロンを用いて、 炭素イオン(¹²C⁶⁺, 320 MeV)を照射した。照射後、 再分化培地に移植し1~2ヶ月後に生存率やシュートの 分化数について調査した。

デルフィニウムでは炭素イオンを0.5, 1.0, 2.0, 4.0, 8.0, 16.0および32.0 Gyで照射した。生存率は0, 0.5お よび1.0 Gyの線量までは90%以上であったが、2.0 Gy では60%, 4.0 Gyでは10%と著しく低下し、8 Gy以上 の線量では全て枯死した(Fig. 1)。

シュートの分化数は0 Gyと比較して照射した区で減 少した。2.0 Gy以上の線量では生存率を調査した2ヵ月 後に枯死していなくてもその後の培養中に枯死する個 体も多くみられたため、炭素イオン照射での適正線量 は0.5~1.0 Gyと推測された。

スターチスシヌアータでは炭素イオンを1.0, 2.0, 4.0, 8.0, および16.0 Gyで照射した。線量による生存 率の差は小さかったが、線量が強くなるにつれ水浸状 化およびシュートの分化がみられない節が多くみられ た(データ省略)。

炭素イオンビーム照射によるシュートの生育は0お よび 1.0 Gyで差はみられなかったが、2 Gy以上にな ると著しく抑制された(Fig. 2)。このことから炭素イ オン照射での適正線量は1.0 Gy付近であると推測され た。

デルフィニウムでは再分化した174個体をポットへ 移植し(Fig. 3)、また、スターチスシヌアータでは 441個体を順化し、花色等の特性について調査する予 定である。



Fig. 1 Influence of 320 MeV carbon ion beam irradiation to leaf blade on survival rate and number of shoots in *Delphinium*.



Fig. 2 Influence of 320 MeV carbon ion beam irradiation to in vitro shoots on growth in *L. Sinuatum*.



Fig. 3 Ion beam-irradiated plants of *Delphinium '7P'* that were planted and grown in pot.

Reference

 M. Okamura et al., JAEA Takasaki Annual Report 2005 (2007) 82.

3-13 Dose Response and Mutation Induction by Ion Beam Irradiation in Chrysanthemum

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Recently the irradiation with ion beams has become a new method for mutation breeding of Chrysanthemum. This study was conducted to evaluate the effect of ion beam irradiation on mutant induction of Chrysanthemum cultivar 'H13'. The irradiation by ion beams induced flower colour changing; vermilion, white/red, and several other colours were induced from the original 'H13' red. Furthermore, the variation of T type flower cluster was obtained from the conventional diamond flower cluster. These results suggest the valuable effects of beam irradiation on mutant induction in Chrysanthemum.

盆コギクは、仏花として出荷期が決まっているが、 天候による開花変動が大きく、産地では危険分散のた めに多くの品種が作付けされている。

こうした中、京都府では開花変動が少なく花色が赤 紫色の 'H13'を育成した。しかし、盆コギクとしては 赤・白・黄の3色セットが望まれている。そこで、イ オンビームを用いた突然変異育種法に取り組むことと した。イオンビームは、ガンマ線やX線などの放射線 と比較して幅広い変異を示すことが知られており¹⁰、 既にキクでは花色変異品種をはじめ、多くの品種が育 成された実績がある。

本研究では、'H13'から、イオンビーム照射と培養 により白色や黄色等に変化した品種を育成するために、 葉片にイオンビームを照射し、変異体の作出を試みた。

'H13'の葉片を3~5 mmに調製し、NAA 0.1 mg/LとBA 0.5 mg/Lを添加したMS培地を10 mL含む60 mmシャーレに置床した。次に、日本原子力研究開発機構(高崎市)の照射施設AVFサイクロトロンにおいて、これらサンプルに加速粒子¹²C⁵⁺、エネルギー220 MeVで0~32 Gyの範囲で照射し、培養によりシュートを再生させた。順次、順化・育苗した後、4号ポットに鉢上げしてガラス温室内で栽培し、開花調査を実施した。

また昨年度、ガラス温室内で、一次選抜を行った株 については、ほ場で育成し、花色・花型変異体の選抜 を行った。

栽培条件は、うね幅130 cm、株間を横10 cm、 縦15 cm、6条植え、無摘心とした。

その他の耕種的管理は京都府の慣行栽培に準じた。

'H13'からは花色が暗赤色、朱色、ピンク及び赤白 混合色の変異体を得ることができた。また花弁が肥大 した個体及び八重咲きの個体を選抜した(Fig.1)。

さらに'H13'の草型の改善点として指摘されていた、 ひし形咲きからT字咲きの個体を選抜した(Fig. 2)。

線量は、1 Gyよりも2 Gyの方が変異率が高く、変 異幅も広かった。現在、有望株について、慣行法によ りほ場で栽培している。今後、花色や草型等の特性を 調べ、二次選抜を行う予定である。

また、さらに大きな変異を誘発させる目的で、一次 選抜株に再度イオンビームを照射した。現在、育成し た株をガラス温室内で栽培中であり今後、開花調査を 実施する予定である。



Fig. 1 Selected mutations of cv. 'H13'.



Fig. 2 Selected mutation of cv. 'H13'. Left, conventional diamond flower cluster; right, T type flower cluster.

Reference

 S. Nagatomi et al., TIARA Annual Report 1997 (1998) 41.

3-14 Mutation Induction in Azalea Seedlings Using Ion Beam Irradiation

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1. Introduction

Creation of new flower color and shape is most important factor in ornamental plant breeding as well as in azalea breeding¹). For the purpose to obtain flower color and/or shape mutation and to analyze the key genes of these mutations, azalea seeds of several species and cultured leaf segments were irradiated by ion beams^{2), 3}). The germination rates of irradiated seeds and regeneration rate of shoot primordia and shoot from irradiated leaf segments were described in previous reports^{2),3}. The growth and survival rate of seedlings obtained from ion beam irradiated seeds are investigated.

2. Materials and Methods

Seeds of *Rhododendron ripense, R. japonicum, R. wadanum*, and *R. degronianum* var. *okiense* were irradiated with carbon ion beams (220 MeV $^{12}C^{5+}$) at 0 to 50 Gy. Irradiated seeds were sowed on sphagnum moss bed in pot. The germination rate was evaluated after 6 weeks. Seedlings were transplanted into pots and maintained with a conventional cultivation method. The growth and survival rate of seedlings were observed and compared with control plants grown under identical conditions at 10 months after sowing. At the same time, variations in leaf morphology were also investigated.

3. Results and Discussion

The optimal irradiation doses for seeds considering from median germination decrease were different depending on



Fig. 1 Dose response of survival rate in *Rhododendron* species at 10 months after sowing.

species^{2), 3)}. Growth and survival rate of seedlings were also similar to these results (Fig. 1). *R. ripense* and *R. japonicum* seedlings grew up to 20-Gy irradiation. *R. wadanum* and *R. degronianum* var. *okiense* seedlings grew up to 30-Gy irradiation. While germination rates were highest in unirradiated seeds of four azalea species, survivals rates were higher at low dose irradiation conditions. These results would be caused by stimulation effect of ion beam irradiation.

Variegated leaves were observed in some of seedlings (Fig. 2). Dwarf and poor growth seedlings were included in *R. japonicum, R. wadanum*, and *R. degronianum* var. *okiense.*

It takes 3 to 4 years to flower from seed sowing in azalea. It is expected to obtain flower color and shape mutant from these seedlings and their progenies, which contribute to study of azalea breeding.

- J. de Reik et al., The potential of molecular analysis and interspecific hybridization of azalea phylogenetic research. In: A.K. Sharma and A. Sharma (Eds) *Plant Genome: Biodiversity and Evolution*. Vol.1, E (2007) pp. 251.
- N. Kobayashi et al. JAEA Takasaki Annual Report 2005 (2007) 96.
- N. Kobayashi et al. JAEA Takasaki Annual Report 2006 (2008) 87.



Fig. 2 Varied growth seedlings of azalea species obtained from ion beam irradiated seeds.

3-15 Effects of Ion Beam Irradiation on Soybean Seeds Evaluated by Plant Growth and Seed-Setting in a Field

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Soybean (Glycine max) is an important crop in terms of production of food, oil, and forage. However, existing mutant lines of soybean is very limited, which is a constraint on performing a genetic study and breeding of this plant. This plant is considered to have derived from ancestral plant(s) that have a tetraploid genome, and as a consequence, more than 90% of nucleotide sequence in the soybean genome is duplicated¹⁾. It is conceivable that such a duplicated nature of the genome brought about a low frequency of mutant production by conventional methods for mutagenesis such as γ-ray or X-ray irradiation as well as chemical treatments. In these circumstances, we have started to examine whether ion beam irradiation is effective in producing a mutant in soybean because ion beam irradiation is expected to cause genomic changes that are more drastic than those induced by conventional mutagenesis.

We have previously analyzed the effects of ion beam irradiation on plant growth and morphology in soybean by exposing dried seeds to the 320 MeV carbon ions with the range of $0.2-25 \text{ Gy}^{2)}$. The irradiated seeds were sown on soil and plants were grown for three weeks in a greenhouse. A prominent decrease in plant height was observed depending on the dose in the range of 5-25 Gy, but not in the dose range lower than 5 Gy (0.2, 0.5, 1.0, and 2.0 Gy), indicating that irradiation doses higher than approximately 5 Gy affect plant growth rate. In addition to the inhibition of plant growth, morphological changes such as changes in the shape of primary leaves and unscheduled generation of stem(s) from the node where cotyledons were formed at the doses of 5 Gy and 2 Gy.

In order to establish a plant population that can be available for screening mutants, we further examined the effects of irradiation by growing plants in a field (Fig. 1). Plant height was measured 1.5 months after sowing irradiated seeds in the field (Fig. 2). A decrease in plant height depending on the doses of irradiation was observed in the field as in the greenhouse. Plants were grown further in the field, and seeds were harvested. The ratio of the number of plants that survived until seed-setting per the number of seeds sown in the field also depended on the doses of irradiation (Table 1).

We also grew plants from irradiated seeds in a large scale to obtain a population of M2 seeds: 3,200 seeds and 3,320 seeds irradiated at 5 Gy and 2.5 Gy, respectively, were sown in the field and plants were grown for harvesting seeds. The survival ratio until seed-setting was higher in plants irradiated with 2.5 Gy than 5 Gy mainly because the latter was more vulnerable to an environmental damage such as strong wind during the growth in the field. Seeds were harvested from more than 1,400 individual plants irradiated at 2.5 Gy, which was approximately two times higher than the number of 5-Gy irradiated plants that survived until seed-setting. We are currently producing the progeny of the irradiated plants to screen mutants.

- 1) R. C. Shoemaker et al., Genetics 144 (1996) 329.
- A. Kanazawa et al., JAEA Takasaki Annual Report 2006 (2008) 88.



Fig. 1 Young soybean plants grown from irradiated seeds in the field of Hokkaido University.



Fig. 2 Growth of plants after irradiation at the doses of 1-20 Gy. Average \pm SE are shown.

Table 1 Effects of irradiation on survival of plants until seed-setting in the field.

Dose (Gy)	0	1	2.5	5	10	20
No. of plants with seed- setting / no. of seeds sown in the field	39/80	24/40	31/80	19/80	12/80	2/80

3-16 Comparative Effect of ¹²C⁶⁺ Beam and Gamma-ray Irradiation on Callus Growth and Shoot Formation from Lavandin Isolated Cells

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In order to develop a new lavandin, *Lavandula* × *intermedia* Emeric., plant with different fragrance from original plant, effect of several doses of ${}^{12}C^{6+}$ beams and gamma-rays on callus growth and shoot formation from isolated cultured cells was analysed. Large differences for responses of callus growth and shoot formation were observed between two ionizing radiations. In ${}^{12}C^{6+}$ beam irradiation, callus proliferation was strongly restricted with increase of irradiation dose. On the other hand, the response of gamma-ray irradiated cells was different. Up to 20 Gy, callus proliferation was stimulated with increase of irradiation dose, although, over 20 Gy, callus growth was inversely restricted with increase of irradiation dose. Similar results were observed in shoot formation. These results suggested that a large difference of biological effect for lavandin cultured cells was existed between ${}^{12}C^{6+}$ beams and gamma-rays. Root differentiation and acclimatization are now conducted to obtain regenerated plants from shoots.

ラバンジンは真正ラベンダーとスパイクラベンダー との種間雑種であり、雑種強勢により精油成分の生産 性が両親種より高いことが知られている。一方、カン ファーあるいはボルネオール等の香質低下成分も増加 していることから、これら成分の生産が抑制された突 然変異体の作出が求められている。

前報(Tsuro et al. 2008)において、著者らは¹²C⁶⁺ ビームがラバンジン培養単離細胞の増殖に及ぼす影響 を調査し、2.0 Gy程度の線量が効率的な変異体獲得に 有効であることを明らかにした。本研究では、既報の 結果をもとに培養単離細胞へ種々の線量のガンマ線を 照射し、培養細胞の増殖に及ぼす影響を調査するとと もに、¹²C⁶⁺ビームおよびガンマ線照射細胞からの シュート再生を試みた。

植物材料としてラバンジン品種、スーパーセビリア ンブルー、を用いた。葉由来カルスから作成した液体 懸濁培養細胞を細胞密度が 2×10^5 個/mLとなるように 調整した。カルス培養用固形培地を入れた直径6 cmの プラスチックシャーレに細胞懸濁液を0.5 mL分注し、 320 MeV $^{12}C^{6+}$ ビームを0, 0.5, 1および2 Gyの線量で、 他方、 60 Coを線源としたガンマ線を0, 10, 20, 40, 80, 120, 160および200 Gyの線量で照射した。照射後、 25 °C, 暗条件で培養を行い、4週間後に増殖したカル スの大きさを0から5の6段階で評価した。評価後、カ ルスを1 mg/L BAを添加したMS固形培地に移植して 25 °C, 明条件で培養し不定芽を誘導した。実験は2回 反復で行った。

ガンマ線を照射後、1ヶ月間培養したときのカルス の増殖程度を評価した。0 Gy区での増殖指数(GI)は2.8 であったのに対し、10~40 Gyでは4.2~4.8と高い値を 示した。しかし、40 Gyを超える線量区ではGIが著し く低下し、200 Gy区で殆ど増殖が見られなくなった。 Fig. 1に前報(Tsuro et al. 2008)で示した¹²C⁶⁺ビーム および本研究で明らかとなったガンマ線における線量 反応曲線を示した。¹²C⁶⁺ビームを照射したとき、ガ ンマ線と比較して低い線量で増殖が抑制されており、 細胞の増殖に及ぼす線量効果が両者で著しく異なるこ とが明らかとなった。

増殖したカルスをシュート誘導培地に移植して培養 したとき、一部のカルスからシュートの形成が認めら れた。Table 1および2に培養2ヵ月後のシュート形成率 を示した。0~2 Gyの¹²C⁶⁺ビーム照射区ではいずれの 線量区でもシュートの形成率が線量の増加に従って減 少したが、11.1~14.9%と大差なく、前報(Tsuro et al. 2008)と同様な値となった。一方、ガンマ線照射区で は0~20 Gyまで形成率が増加したが、40 Gy区で低下 に転じ、80 Gy以上でシュートの形成が認められなく なった。これらの結果から、カルスの増殖誘導と同様 にシュート形成においても¹²C⁶⁺ビームとガンマ線照 射区において反応が異なることが明らかとなった。

現在,両照射区で得られたシュートの発根・馴化を 行っている。

Table 2 Effect of different doses of gamma-ray on shoot formation

5				trom	solated cell-derived callu	s in lavandin
	Table 1 Effect of different doses of ¹² C ⁶⁺ beam on shoot formation from isolated cell-derived callus in lavandin			Dose (Gy)	No. of Cultured Calli	No. of Shoot Formed Calli (%)
	Dose (Gy)	No. of Calli	No. of Shoot Formed Calli (%)	0	50 68	11 (31.4)
2	0	110	35 (46.7)	20	60	17 (45.9)
	0.5	189	12 (12.5)	40	60	7 (25.0)
0	1.0	180	13 (14.9)	80	60	0 (0)
0 25 50 75 100 125 150 175 20 Dose (Gv)	0 2.0	188	10 (11.1)	120	50	0 (0)
Fig.1 Callus proliferation response for different doses of ¹² C ⁶⁺ beam	and			160	15	0 (0)
gamma-ray in lavandin isolated cultured cells.				200	4	_

3-17 Screening of Higher Astaxanthin Producing Mutants of a Green Unicellular Alga *Haematococcus pluvialis* at Elevated Acetate Concentrations by Ion Beam Irradiation

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A highly anti-oxidative ketocarotenoid, astaxathin has been produced by culturing a unique green unicellular alga *Haematococcus pluvialis* in large-scale photobioreactors at rich solar energy areas such as Hawaii islands. Since it seems disadvantageous to carry out the algal culture phototrophically in a commercial basis in Japan, we have applied heterotrophic cultivation for the alga. In the dark condition, the alga was found to grow 2.5 fold more in cell number (reaching 1.12 million cells/mL) after subculturing three times. The acclimatization effect was also verified in several separate cultures at different acetate concentration. Upon ion-beam irradiation at various intensities, acetate-tolerant mutants and higher astaxanthin producing mutants were extensively subcultured in either liquid cultures or agar plate cultures containing several acetate concentrations from 30 to 60 mM. At over 45 mM plate cultures, only red colonies were formed unlike green colonies at the lower concentrations of acetate. The former was identified as mostly enlarged cysts cells, whereas the latter was a regular vegetative cell. The result indicated that the plate culture condition was likely to mimic the oxidative stress condition under phototrophic culture possibly as drought stress condition.

高度に抗酸化力を有するアスタキサンチン(以下 ASX)は、ほとんどの種類の甲殻類,サーモンの肉色, タイの体表皮など海洋生物に広く分布するケトカロテ ノイドである。近年、体内で必然的に発生する活性酸 素が種々の疾病,老化,発ガン作用に関与するとの報 告があるので、食品やサプリメントとして抗酸化作用 が注目され、特にASXについては、疲れ目予防,ロー ション,健康飲料など様々な用途に利用されるように なった。

微細緑藻 H. pluvialisは分裂増殖する栄養細胞 (Fig. 1左)においてカロテノイド色素は少量である が、栄養条件の悪化とともにシスト細胞と呼ばれる胞 子嚢細胞(同,右図)へ形態変化し、ASXを細胞重量 比で2-3%の著量に生成する¹⁾。



Fig. 1 Morphological change from vegetative cells (left) to cyst cells (right) of *H. pluvialis*, and massive astaxanthin formation in cyst cell.

そこで、われわれは本緑藻が、太陽光に依らずに暗 所で酢酸を炭素源に従属栄養的に増殖し、ASX生成を 行いうることに着目し、その大規模生産の可能性を検 討すべく、形態変化を伴わせたASX生産条件、および 基質種と濃度効果を調べた。H18年度の結果では、 ASX含量は光培養系に匹敵するものの、細胞濃度が上 がらず、酢酸は30 mMを越えると増殖阻害が起きたが、 H19年度において、暗所培養を3回継続して継代した 結果、栄養細胞が従来の2.5倍の高濃度化 (1.12×10⁶ cells/mL)が可能となった(Table 1)。ま たこの暗所条件に馴養させた結果、酢酸に対しての増 殖阻害が弱まり、45 mMでも従来の1.5倍程度の濃度 にまで増殖することが示された。

Table 1 Effect of dark culture acclimatization on the algal growth at 4 d.

U							
	[Cell] (×10 ⁵ cells/mL Acetate						
Acclimatization	15	30	45 mM				
Before	3.8	3.2	1.6				
After*	5.6	11.2	2.3				

The alga was sub-cultured three times in dark.

そこで、イオンビーム変異導入法を用いて、さら なる酢酸耐性変異株等を広範囲にスクリーニング・単 離することを目的に、イオンビーム強度を種々に変化 させて変異細胞調製を行い、変異株を種々の酢酸濃度 からなる液体培地または寒天培地に塗布し、案諸条件 で継代培養を検討してきている。寒天培地では、 45 mM酢酸濃度を含む場合には、従来の緑色コロニー となる栄養細胞としてではなく、シスト細胞からなる 強い赤色コロニーを形成することが明らかとなった。 顕微鏡観察によると、およそ栄養細胞の20 μm程度の 長径に対して、100 μm以上に肥大したシスト細胞が 多いことがわかった。これはASX生産条件に類似する ことから、寒天培地の乾燥条件がカロテノイド生産に 適していることが示唆された。

Reference

 M. Kobayashi et al., Appl. Environ. Microbiol. 59 (1993) 867.

3-18 Induction of Thornless Yuzu Mutant by Heavy Ion Beam Irradiation

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Yuzu (*Citrus junos*) tree bears long thorns that deteriorate fruit quality. We are trying to produce thornless mutant of Yuzu by carbon-ion irradiation. Cut surface of lower hypocotyls were exposed to carbon ions with a total energy of 320 MeV. Based on the shoot regeneration rate, we determined less than 8 Gy is appropriate for our culture system. We successfully obtained completely thornless mutant together with several mutant plants with weak thorns or lesser number of thorns. We top-grafted these mutant plants for aging into Satsuma mandarin, and will examine characteristics of fruit.

ユズは樹体に長いトゲが発生するため、栽培管理 上障害となっている。さらに、トゲによる刺し傷はキ ズ果や腐敗果の発生など果実の商品性を低下させる。

そこで、ユズ実生の胚軸断面からシュートを再生 させる培養系に重イオンビームを照射し、トゲ消失変 異体の作出を試みた。今回は前回の試験結果を受け、 2線量区で再生シュートを育成し、トゲ消失変異候補 個体を作出した^{1),2)}。さらに、前回育成した候補個体 の果実品質を確認するため、12年生温州ミカン(大津 4号)に高接ぎを実施したので報告する。

ユズ (Citrus junos Sieb.ex Tanaka) 種子を1/2MS培 地に播種後、3週間目に胚軸を2 cm程度残し切断した。 さらに、1週間後に切断面より発生したカルスに重イ オンビームを照射した。照射線種はCイオン (¹²C⁶⁺ 総エネルギー 320 MeV)を用い、照射線量は2 Gy、 4 Gyの2線量区とした。照射施設は原子力機構高崎量 子応用研究所のAVFサイクロトロンを用いた。 照射後、再分化個体数及びトゲ消失対数を調査した。 さらに前回照射した個体について、樹勢が弱いものに ついては、強勢台木のラフレモンに寄せ接ぎを図り、 生育促進を行った。寄せ接ぎ後、再度トゲ消失を確認 できた個体については、温州ミカンに高接ぎを実施し、 結実促進を図った。

2週間後の再分化シュートの発生率は、2 Gy以下で は100%以上であり、また8 Gyでも60%は再分化した。 なお、1胚軸より複数発生することもあるためシュー トの発生率が100%を越えるものもあった(データ 略)。各線量区とも1,000個体に照射を実施した結果、 2 Gy区、4 Gy区各々878個体、915個体が胚軸カルスよ り再分化した。さらに、トゲの発生がほとんど確認で きない候補個体は2 Gy区では21個体、4 Gy区では12個 体獲得することができた(Table 1)。また、前回の照 射試験により13個体のトゲ消失候補個体を獲得した (Table 2)。再度トゲの消失状況を確認し、高接ぎ可 能な程度に伸長した8個体については、果実品質特性 を把握するため12年生温州ミカン苗木(大津4号)を 中間台木として高接ぎを実施した(Fig. 1)。さらに、 県内カンキツ生産の主力品目である温州ミカンの有望 系統を作出するため、照射条件や線量の検討に着手し た。

今後はカンキツ由来の機能性成分として注目されて いるポリメトキシフラボン等の機能性成分を高含有し た品種を育成するために、照射個体の成分分析を実施 していく。

Table 1. Shoot regeneration rate from hypocotyls irradiated with carbon ions and Appearance rate Thornless Yuzu mutant.

		4 month after irradiation				
Dose	No. of irradiation	No. of shoots	Regeneration rate	No. of Thornless Yuzu mutant	Appearance rate	
(Gy)			(%)		(%)	
2	1,000	878	87.8	21	2.39	
4	1,000	915	91.5	12	1.31	

Table 2. The number o	f Thornless Yuzu n	nutant and Top-grafting.
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_	${}^{12}C^{6+}$: D ose (Gy)				
	1	2	4	8	Total
No. of Thornless Yuzu mutant	5	5	2	1	13
No. of Top-grafting	2	4	1	1	8



Fig. 1 Execution condition of top-grafting for aging (rootstock;Satsuma mandarin).

- Y. Matsuo et al., Hort. Res.(JAPAN) 6 Suppl.1 (2007) 40.
- Y. Matsuo et al., JAEA Takasaki Ann. Rep. 2006 (2007) 79.

3-19 Mutation Breeding on the Ornamental Plants of *Gypsophila* and *Gentiana* Species (II)

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Gypsophila paniculata and *Gentiana trifida* var. *japonica* are important ornamental crops for cut flower use in Japan. In an attempt to induce mutants for breeding use, plant shoots on the LS medium were irradiated with carbon ion beams from the JAEA AVF cyclotron. From the preliminary irradiation experiments, the doses suitable for mutation induction are estimated to be 2 to 4 Gy for *Gypsophila* and 0.5 to 1 Gy for *Gentiana*, respectively. The additional irradiation experiments were conducted, and 833 and 255 regenarated plants were obtained for *Gypsophila* and *Gentiana*, respectively. So far, 38 mutants have been selected in *Gypsophila*. Propagation of the mutants is under way for further analysis.

宿根かすみそう・りんどうは切花用として栽培・ 出荷されている重要な花き作物である。本研究は、こ れらの花き作物について、イオンビーム照射による新 たな有用変異の誘発の可能性についての知見を得るこ とを目的とする。

宿根かすみそう(品種「ブリストルフェアリー」) のLS培地での茎頂培養植物に、炭素イオンビーム (¹²C⁶⁺, 320 MeV)の照射後、発根培地への植え継ぎ、 順化およびポットへの鉢上げを実施した。そのさいに、 茎葉の奇形・生育不良などの異常率および生存率を調 べた結果、2~4 Gyの線量が変異誘発に適正であると 考えられ¹⁾、32 Gy以上の照射では、ポットへの鉢上 げまでに生存する個体はなかった。これは、すでに報 告されている他の研究と一致するものであった²⁾。

上記の結果を基に、「ブリストルフェアリー」およ び同じ類縁関係を持つ5品種(「ブランシー」、FG361、 FG365、FG382、FG411)について、4 Gyの炭素イオ ンビーム照射を実施した。宿根かすみそうの全照射試 験で833の再生個体が得られ、2007年11月からハウス 隔離栽培により変異の有無の調査を実施してきている。 現在(2008年5月)までに、Fig. 1 に示すような、葉 が幅広で肉厚である個体を含む38の変異個体を一次選 抜した。変異の内訳としては、早晩性5%、茎葉の形 態63%、花器の形態31%である(変異の種類には重複 もあるため便宜的な分類である)。今後、これらの変 異個体から挿し芽増殖をおこない、突然変異の確認お よび有用性検定のための二次選抜を行う予定である。

りんどう(在来系統)については、同様の照射試 験から、0.5~1 Gy照射が変異誘発に適正であると結 論した¹⁾。8 Gy以上になると順化時までに全て枯死し た。この結果から、1 Gy線量で照射した培養物から再 生した照射当代(255個体)では、花色などの有用な 変異体を選抜することはできなかった。2008年2月ま でに、95個体から自殖種子を得た。現在、次世代種子 が得られた系統について播種・育苗を行っており、今 秋、変異体の選抜を行う予定である。



Fig. 1 Morphological mutant of *Gypsophila*. Right: nonirradiated control plant, left: mutant plant obtained from 16-Gy carbon-ion irradiation.

- S. Tsuji et al., JAEA Takasaki Ann. Rep. 2006 (2008) 84.
- M. Okamura et al., 2006 Annul Report of the Research Project with Heavy Ions at NIRS-HIMAC (2007) 75.

3-20 Producing New Gene Resources in Fig by Using Ion-beam Irradiation

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The goals of our study are to produce new gene resources in fig by mutation induction with ion-beam irradiation, because the breading by varietal crossing in fig is very difficult. As the second paper of our study, we report the influences of carbon ion beam and soft X-ray irradiation to *in vitro* lateral buds on mutation induction in fig 'Banane'. *In vitro* buds were irradiated with carbon ion beams (320 MeV ${}^{12}C^{6+}$) at TIARA, and with soft X-rays (100 kVp, 5 mA, 120 Gy/h) at Aichi Agric. Res. Ctr. The efficiency of survival rate of buds, the fresh weight and the length of lateral shoots, and the rate of shoot that grows over 1 cm were investigated. The optimum doses for carbon ion beams and soft X-rays were 7.5 Gy of 320 MeV ${}^{12}C^{6+}$ and 35 Gy of soft X-rays, respectively. Radiation sensitivity of 'Banane' was lower than 'Masui- Dauphine'.

日本のイチジク産地で栽培されている品種は、ほ とんどが"桝井ドーフィン"である。また、日本での 栽培に適したフィッグ型イチジク品種はほとんど雌花 しか着生しないこと¹⁾から他品種との交雑育種は困難 であり、これまで遺伝子資源の拡大はされてこなかっ た。そこで本課題では、イオンビームを利用して高品 質で特徴ある新品種開発につながる遺伝子資源を創成 する。本年度は昨年度とは異なるイチジク品種の培養 苗腋芽に炭素イオンビーム及び軟X線を照射し、放射 線感受性の品種間差の有無を調査する。

無菌培養したイチジク品種"バナーネ"の葉を切 除した茎部を節ごとに切断し、6 cm径シャーレ内にの MS液体培地(ショ糖3%)を含ませた育苗用オアシ ス®(発泡フェノール樹脂)を入れ、腋芽の向きを上 に揃えて挿した。同試料に加速した炭素イオン粒子 (320 MeV・¹²C⁶⁺、LET:86 keV/µm) を種々の線 量で照射した。また、比較のため愛知農総試保有の軟 X線照射装置(ソフテックス株式会社製M-100WE 型:100 KVp、5 mA、120 Gy/h)を用い同様の方法 で照射した。照射試料は液体培地を含ませたオアシス ®に挿し、プラントボックス内で育てた。培養は 25 °C、照度3,500 lux、16 時間日長で行い、培養60日 後の腋芽の生存率、10 mm以上伸長した腋芽の割合、 腋芽の茎長、重量及び葉数を調査した。炭素イオン ビーム照射では各試験区ごとに腋芽40~50節を、軟X 線では腋芽30節を供試した。

炭素イオンビームを2.5 Gyから20 Gyまで段階照射 したところ、10 Gyを越えると生存率が低下した。ま た、10 mm以上伸長する腋芽の割合、伸長した腋芽の 茎長及び重量は5 Gy~7.5 Gy照射で非照射区(0 Gy) の平均値に対して50%程度にまで低下した。腋芽が生 存しても伸長しなければ選抜に用いることができない ことから、10 mm以上伸長する腋芽がわずかに残る 10 Gyが上限と考えられた(Fig. 1)。2回目の試験結 果では、非照射区の生育の50%程度に低下する線量は 7.5 Gyであった(データ未掲載)。

軟X線を12 Gyから72 Gyまで段階照射したところ、 48 Gy以上になると腋芽生存率が低下し、非照射区の 生育の50%程度に低下する線量は約35 Gyであった (Fig. 2) 。

以上の結果、腋芽の生育に同程度の影響が現れる両 線種の線量を比較すると、炭素イオンビームは軟X線 の6分の1以下であった。また、昨年度に調査したイ チジク品種"桝井ドーフィン"では、非照射区の生育 の50%程度に低下する線量が約5 Gyだったこと²⁾から、 "バナーネ"は"桝井ドーフィン"に比べ、放射線感 受性がやや低かった。



Fig. 1 Influence of 320 MeV carbon ion beam irradiation on growth of *in vitro* lateral buds in fig "Banane".



Fig. 2 Influence of soft X-ray irradiation on growth of *in vitro* lateral buds in fig "Banane".

- 平田尚美.農業技術体系-果樹編5,東京,農文協.イ チジク,基29 (1993).
- I. Asami et al., JAEA Takasaki Ann. Rep. 2006 (2008) 80.

3-21 Characterization of Enhancers and Suppressors of Circadian Clock Mutant *elf3* in Arabidopsis

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Developmental transitions of plants are strongly affected by light quality, intensity and duration. *Arabidopsis* is a facultative long-day (LD) plant that flowers earlier under LD than SD photoperiods. Exposure to blue or far-red lights promotes flowering of *Arabidopsis*. Many genes have been reported to be required for the photoperiodic regulation of flowering in *Arabidopsis*.

Photoperiodic flowering is affected by circadian clock¹⁾. Genetic approach has identified more than a dozen of key genes for clock functions in *Arabidopsis*²⁾. Although phenotypic characterization of the clock mutants and identification of the corresponding genes have been done, biochemical functions of the clock proteins have been largely unknown¹⁾. For example, EARLY FLOWERING 3 (ELF3), GIGANTEA (GI) and EARLY FLOWERING 4 (ELF4) are proposed to play key roles in the clock function of *Arabidopsis* and these clock genes were identified from the end of 1990's to the beginning of 2000's³⁾. However, *ELF3*, *GI* and *ELF4* genes encode novel proteins with no significant sequence similarity to characterized proteins in the existing public databases. There has been almost no clue to investigate biochemical roles of these clock proteins.

EARLY FLOWERING 3 (ELF3) gene of *Arabidopsis* regulates a wide variety of processes such as plant morphology, flowering time and circadian rhythms⁴⁾. The *elf3-1* mutation was the 1st loss-of-function allele of *elf3* and identified in a screen for early flowering under SD. Mutations in *ELF3* result in the loss of both photoperiod sensitivity and circadian regulation, making ELF3 a candidate for linking circadian clock function with the photoperiodic induction of flowering^{1,4)}. The *elf3* mutant plants flower earlier than the wild type under both short and long photoperiods. The mutant plants show phenotypes associated with defect in reception or transduction of light signals, namely, pale-green leaves, elongated hypocotyl and

petioles, suggesting that ELF3 is a multi-functional protein. However, it has not been elucidated for a long time how the ELF3 protein accomplishes the multi-tasks.

Several ways of approaches have been taken to understand how the mysterious protein "ELF3" regulates several important biological processes in Arabidopsis. Recently, we have identified new alleles of elf3 as suppressors of *lhy;cca1* under $LL^{2,3,5)}$. We have found that ELF3 physically interacts with both a clock protein CCA1 and a floral repressor SVP, suggesting that ELF3 may mediate between the oscillator and one of the outputs of circadian clock in Arabidopsis. To understand roles of ELF3 in more details, we have performed genetic screening of enhancers and suppressors of the elf3-1 by mutagenesis using irradiation of heavy ion beam. Seeds of elf3-1 mutant were exposed to 220-MeV carbon ions with a dose of 120 Gy. Mutants with altered hypocotyl and petiole length were screened in the M2 generation. In this genetic screening, we have identified 7 suppressors and 1 enhancer that have shorter and longer hypocotyls and petioles, respectively (Figs. 1 & 2). Genetic mapping is underway. Identification of novel factors that control stability or activity of ELF3 will much advance our knowledge on roles of circadian rhythms to regulate proper development of plants.

- 1) T. Mizoguchi et al., Int. Rev. Cytol. 250 (2006) 47.
- 2) T. Mizoguchi et al., Dev. Cell 2 (2002) 629.
- 3) T. Mizoguchi et al., Plant Cell 17 (2005) 2255.
- 4) T. Tajima et al., Plant Biotechnol. 24 (2007) 237.
- 5) S. Fujiwara et al., unpublished data.



Fig. 1 Suppressor of the *Arabidopsis* clock mutant, *elf3-1*. Wild type (left), *elf3-1* (middle) and suppressor line (right) are shown.



Fig. 2 Enhancer of the *Arabidopsis* clock mutant, *elf3-1*. Wild type (left), *elf3-1* (middle) and enhancer line (right) are shown.

3-22 Mutation Induction of Strawberry (*Fragaria* × *ananassa*) Using Ion Beam Irradiation

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We examined the irradiation effect of helium and carbon ions on regeneration rate of strawberry (*Fragaria* × *ananassa*), and investigated the effect of carbon ion on flower budding. Twenty M_1 plants showed earlier date of flower budding than untreated plants in tested 286 M_1 plants. Carbon ion irradiation seems to be an effective technique to induce strawberry mutants.

1. はじめに

イチゴ(Fragaria × ananassa)の栽培は、促成品種の 導入と施設栽培技術の発達により収穫期間が前進化し、 且つ長期化している。本試験ではイチゴ品種の変異拡 大を目的として、イチゴに対するイオンビームの適正 線量を測定するとともに、イオンビーム照射が花房の 発生時期に及ぼす影響を調査した。

2. 実験方法

イチゴ品種は「もういっこ」を用いた。無菌培養で 増殖した腋芽を含む切片に対して、ヘリウム⁴ He²⁺ (100 MeV)または炭素イオン¹²C⁶⁺(320 MeV)を照射し、 その後にベンジルアデニン0.1 mg/Lを含むMS培地で切 片から再生した植物体を育成しM₁世代(突然変異第1 世代)とした。2007年3月5日に照射処理して育成した M₁世代を、9月11日にセルトレイへ植え順化し、10月 18日に本葉が5から6枚展開した苗を無加温ハウスへ 定植した。無処理区には、イオンビーム無照射処理の 切片から再生した14株を供試した。着蕾日は、肉眼で 株から頂花房の蕾が確認できた日とした。順化後の日 長管理は自然日長条件で行った。

3. 結果及び考察

イオンビームに対する再生率の線量反応を調査した 結果、ヘリウムでは16 Gy,炭素では32 Gyより高い線 量で再生率が低下した(Table 1)。イオンビームをイ チゴの種子に照射した報告^{1,2)}においても、40 Gy以上の ヘリウムおよび炭素イオンで発芽率が低下することが 示されている。これらの知見から、イチゴの再生率に 影響が無い炭素イオンの8 Gyと16 Gyで照射処理を行 い、合計286株のM₁世代を得た。

無処理区で最も早い着蕾日は1月4日であった。この 日より早く着蕾した8 Gy処理12株と16 Gy処理8株の合 計20株を選抜株とした(Table 2)。一方、照射により 着蕾が遅延した株も観察された。多くの作物について 人為突然変異による早生化が報告されているが³⁾、本結 果からイオンビームによるイチゴの変異誘発が、早生 化への品種改良に対して有効な手法であると期待でき る。今後、収穫期間における花房発生の連続性や収量・ 品質等について継続的に調査を行う。

Table 1 Effect of carbon and helium ion beams on regeneration rate.

Dose	Regeneration Rate		
(Gy)	(% of Untreated)		
	He	С	
2	97		
4	93	100	
8	90	100	
16	100	100	
32	57	91	
64	54	81	
128	43		
256	11		

Table 2 Number of tested and selected M₁ plants.

	Dose of	f Carbo	n (Gy)	
Date of Flower Budding	8Gy	(%)	16Gy	(%)
December 6, 2007	2 (1.0)		
December 10, 2007	3 (1.6)	1	(1.1)
December 12, 2007	1 (0.5)		
December 17, 2007	3 (1.6)	1	(1.1)
December 27, 2007	3 (1.6)	6	(6.4)
January 4, 2008 \sim	180 (93.8)	86	(91.5)
January 4, 2008	U	ntreate	d	
Total	192 (100)	94 ((100)

- 1) M. Iizuka et al., TIARA Ann. Rep. 1999 (2000) 33.
- 2) M. Iizuka et al., TIARA Ann. Rep. 1998 (1999) 30.
- (1) 渡辺好郎,山口彦之 監修 "突然変異育種" 養賢 堂 (1983).

3-23 Mutation Induction by Ion-Beam Irradiation in Fragrance Cyclamen (*Cyclamen persicum* × *C. purpurascens*)

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Fragrance cyclamen of amphidiploid (2n=4x=82) was produced by chromosome doubling of the hybrid (2n=41) that was hybridized between cultivar (*Cyclamen persicum*, 2n=2x=48) and fragrant wild species (*C. purpurascens*, 2n=2x=34). However, flower of fragrance cyclamen lacked the diversity in the color, the shape and the size compared with those of cultivar. Mutation breeding that combined ion-beams with tissue culture technique was attempted for overcoming this issue. Petals were collected from immature flower buds of the amphidiploid. Two types of etiolated petiole, one was induced from the amphidiploid in the dark, and the other was induced from the haploid plantlets derived by anther culture of the amphidiploid in the dark. These petals and etiolated petioles were irradiated with 220 and 320 MeV carbon ion-beams at doses of 0-50 Gy respectively, and then they were cultured in the regeneration medium. The survival rate of petals and etiolated petioles decreased rapidly at doses higher than 2 Gy, and mutants containing useful traits were obtained at doses of 0.1, 0.2, 0.5, 1 and 2 Gy.

シクラメンの栽培種(Cyclamen persicum, 2n=2x=48)と 芳香性野生種(C. purpurascens, 2n=2x=34)の種間交雑に より雑種(2n=41)を育成し、その染色体倍加により複二 倍体(2n=4x=82)の芳香シクラメンを育成した。しかし、 芳香シクラメンの花は色,形,大きさの変化が乏しい ため、形質の多様化が求められている。そこで、イオ ンビーム照射と組織培養を利用した突然変異育種によ る変異の拡大を検討した。

複二倍体芳香シクラメンの系統の中から0015 (紫の 花色),99pi7 (ピンクの花色)および99pu1 (濃い紫の 花色)を選定した。99pu1の種子を無菌的に播種し、暗 黒下で培養することにより発生させた黄化葉柄、既報 の方法¹⁾により99pi7の葯培養から作出した半数体を暗 黒下で培養することにより発生させた黄化葉柄,およ び99pi7と0015の開花個体から着色する前に採取した未 熟な花弁を供試した。花弁には220 MeV炭素イオンを、 黄化葉柄には320 MeV炭素イオンをそれぞれ0~50 Gy の範囲で照射し、その後、既報の方法^{2),3}により培養を 行った。照射後の花弁および黄化葉柄の生存率を調査 した。次に生存率のデータを基にイオンビーム照射を 行い、再分化個体を開花個体に成長させ、形態観察に より変異体を選抜した。

黄化葉柄および花弁へのイオンビーム照射では、2 Gyを超える線量区でともに生存率が急激に低下した (Fig. 1)。99pi7の花弁と黄化葉柄における0.1, 0.2, 0.5, 1, 2 Gyの線量区の開花個体から花の色,形に関する有望 な変異体が得られた。99pi7の黄化葉柄における5, 10 Gy



Fig. 1 Effect of ion beam irradiation on survival rate in fragrance cyclamen (*Cyclamen persicum xC. purpurascens*).
A: Survival rate of etiolated petioles of 99pu1.
B: Survival rate of petals of 99pi7.

の線量区の開花個体の中にも変異が見られたが、花が 著しく奇形になり、実用的ではなかった。99pulの黄化 葉柄における0.2, 0.5, 1 Gyの線量区で変異体が得られ たが、花弁へのイオンビーム照射による変異体の誘導 についてさらに検討する必要がある(Table 1)。

以上の結果を基に、99pi7および0015の花弁に2 Gyの 照射を行い、それぞれ734個と1504個の開花個体を育成 し、その中からそれぞれ、47個と12個の変異体を選抜 した(データ省略)。

イオンビーム照射により花弁に現れた変異は、花色 の濃淡,花弁の矮小化,花弁周囲の鋸歯、斑入りなど であったが、花弁以外にも葉の斑入りなどの変異が現 れた。今後、花の色素,香気成分に関する変異の解析 および世代更新による有望な変異形質の固定をとおし て品種育成を進める予定である。

- 1) E. Kondo et al., J. Japan Soc. Hort. Sci. 72 Suppl. 2 (2003) 550.
- E. Kondo et al., J. Japan Soc. Hort. Sci. 72 Suppl. 2 (2003) 551.
- 3) E. Kondo et al., J. Japan Soc. Hort. Sci. 73 Suppl. 2 (2004) 582.

Table 1 Effect of ion beam irradiation on mutation induction in fragrance cyclamen (*Cyclamen persicum* × *C. purpurascens*).

		<i></i>	
	99pi7	99pi7	99pu1
	Petal	Etiolated petiole	Etiolated petiole
Dose (Gy)	(Amphidiploid)	(Haploid)	(Amphidiploid)
(Uy)	No. of	No. of	No. of
	flowering plant	flowering plant	flowering plant
	(No. of mutant)	(No. of mutant)	(No. of mutant)
0	150(0)	108(0)	58(0)
0.1	58(0)	114(2)	18(0)
0.2	32(5)	180(2)	28(1)
0.5	40(1)	176(3)	53(5)
1	46(0)	184(1)	75(16)
2	43(2)	147(3)	30(0)
4	-	-	3(0)
5	2(0)	72(4)	-
8	-	-	1(0)
10	0	71(4)	-
16	-	-	2(0)

3-24 Development of Rice Mutant with Low Cadmium Trait -Mutation Induction by Ion Beam Irradiation-

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In order to obtain rice mutants with low cadmium trait, rice seeds (cv. Koshihikari) were irradiated with ${}^{12}C^{6+}$ at 320 MeV of ranging from 40 to 160 Gy using AVF cyclotron. The germination ratio was hardly affected by 40 Gy. However, a severe inhibitory germination was observed by the doses of 80-160 Gy. The shoot growth was steeply suppressed with increasing the dose of irradiation. Even 40 Gy decreased the shoot growth by 60 % of the non-irradiated control. Although we need to investigate the effect of ion beam irradiation on seed ripening, the optimum dose for developing Koshihikari mutant was estimated to be 40 Gy or less.

食の安心・安全に対する社会的関心が非常に高まっ ている中、農作物のカドミウム汚染を低減させる画期 的な技術の開発が求められている。特に我が国の基幹 作物である「コメ」のカドミウム汚染の低減は国家的 な急務である。カドミウム吸収の少ないイネ品種の開 発は、従来行われてきた客土や資材、湛水管理等によ る対策技術に比べ、環境負荷が少なく、経済的かつ恒 久的なカドミウム低減技術として期待できる。しかし ながら、これまでカドミウム吸収抑制を目的に、新た なイネ品種が作出された例はない。そこで、本研究で は、栽培面積が最も広く良食味米であるコシヒカリに 重イオンビームを照射し、得られた照射個体の中から、 カドミウム吸収の低い株をスクリーニングするととも に、現地カドミウム汚染土壌による実証試験を行い、 低カドミウムイネ系統を育成することを目的としてい る。今回はコシヒカリ変異体誘発のための最適なイオ ンビーム照射線量を検討した。

コシヒカリ乾燥種子を用いて、胚に損傷を与えな い程度の強さで籾摺りを行い、玄米種子を得た。胚に 対して効率的にイオンビームを照射するため、6 cm× 6 cmシャーレ中に胚を上向きにして玄米を縦に一粒づ つ陳列した。1シャーレあたり約400粒を並べた。重イ オンビーム照射 (320 MeV・¹²C⁶⁺) を日本原子力研究 開発機構イオンビーム照射研究施設(TIARA)のAVF サイクロトロンで行った。照射線量は40,80,100,120, 160 Gyの5段階とした。照射した種子は、催芽処理後、 イネ用培土に各100粒づつ播種し、25 ℃に設定した温 室内で栽培した。また、対照区として照射していない 玄米種子(0 Gy) 100粒も同時に栽培した。播種1週 間後に発芽率を測定した。発芽率は0 Gyで発芽した苗 に対する割合として計算した。さらに、播種3週間後 の幼苗を収穫し、各線量区での地上部新鮮重を調査し た。

40 Gyで照射した場合、コシヒカリ種子の発芽率に は何ら影響がなかったが、80 Gyで約50%に低下し、 それ以上ではほとんど発芽しなかった(Fig. 1)。生 育に対する影響を地上部新鮮重で検討したところ、 40 Gyの照射は0 Gyの約60%の生育量であった。また、 80 Gyでは約10%の生育量であり、それ以上ではほとんど生育しなかった(Fig. 2)。

以上、発芽率と地上部生育量に対する影響からコシ ヒカリ変異体誘発の適正線量は40 Gyかそれよりも低 い線量と考えられた。今後、稔実率を調査する必要が あるが、今のところ40 Gyで約1万粒の種子に照射を行 い、圃場による栽培を通してM2種子を採種後、低カ ドミウム吸収変異株を選抜する予定にしている。



Fig. 1 Effect of ion beam irradiation on germination ratio in rice (cv. Koshihikari).



Fig. 2 Effect of ion beam irradiation on shoot fresh weight in rice (cv. Koshihikari).

本研究は、生物系特定産業技術研究支援センターの 競争的資金である新技術・新分野創出のための基礎研 究推進事業「食の安全を目指した作物のカドミウム低 減の分子機構解明」で実施している。

3-25 Mutation Breeding of a New Chrysanthemum Variety by Irradiation of Ion Beams to 'Jinba'

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In order to raising deep yellow "Jinba", light yellow mutant line that was spontaneously obtained in Oita prefecture was irradiated with 1, 2, 3, 4, 5 Gy of carbon ions. Seven candidate plants that shows delayed color degradation were obtained.

秋輪ギク「神馬」(白)は、大分県南部の佐伯市を中心 に多く栽培されている。しかし、「神馬」の栽培特性に 類似した高品質の黄色品種がなく、黄色品種を計画的 に生産できないことが、生産・販売戦略上大きな課題 となっている。一方、花き研究所では、平成14年度に 佐伯市蒲江町で発見された「神馬」淡黄色系統につい て、系統選抜を行い、濃黄色系統を選抜してきたが、 開花時に退色する欠点を改善されていない。

そこで、「神馬」淡黄色系統に対して花色変異に有効 なイオンビーム照射を行い、濃黄色系統への誘発を試 みた。

供試材料は、「神馬」淡黄色系統(以下元株)と平成 18年度に選抜した濃黄化系統(「No.1」~「No.11」)の 葉片及び花弁片で、これらを 2×5 mm に切断し、1 週間 程度培養したものに照射した。照射は、TIARA 内の AVF サイクロトロンを利用し、 $^{12}C^{6+}: 320$ MeV、 $^{12}C^{5+}: 220$ MeV、1、2、3、4、5 Gy で、それぞれの照射試料から 再生個体を作出した。再生個体は、順次鉢上げし、花色 の確認を行ったが、今回は元株を材料とした再生個体 のみを調査した。色彩値(Hunter L.a.b 値)は、色彩色 差計(CR-321、MINOLTA)で測定した。

2007年4月~11月に作出した再生個体は Table1 のと おりで、2006年度に未調査であった個体を含めて、11 月までに順次約4,000個体を調査した。この中から、黄 色の濃い個体を選抜し、蕾、開花時(外弁、内弁)の 色彩値を測定した(Table 2)。

濃黄化個体数は、3 Gy 照射したものが4 個体で最も 多く、2 Gy 照射したものは2 個体、1 Gy 照射したもの は1 個体であった。この内、5 個体(No.14、15、16、 17、18)が葉片から、2 個体(No.19、20)が花弁から の再生個体、また、5 個体(No.14、15、16、17、19) は¹²C⁶⁺を、2 個体(No.18、20)は¹²C⁵⁺を照射して得ら れた再生個体であった。

選抜した 7 個体の蕾の花色は、元株と比較して黄色 が濃くなり、市販 3 品種と同程度の濃さであったが、 開花時では、内弁、外弁とも退色した。 現在も再生個体の順化及び花色調査を随時実施中であり、濃黄化個体(「No.1」~「No.11」)への再照射により得られた個体は、2008年2月から開花調査を行っている。また、選抜個体(「No.1」~「No.20」)の栽培特性調査についても実施中である。

今回選抜した 7 個体は、元株より黄色が濃くなった が、開花時に退色する欠点を改善できなかった。しか し、「No.20」は、開花時の退色程度が軽いため、栽培 特性に問題がなければ品種化が可能だと考えられた。 今後は、選抜個体の栽培特性調査を行い、品種化につ いて検討するとともに、今回選抜した個体の再照射も 実施していきたい。

Table 1 The number of regenerated.

Tissue	Source	Energy(MeV)	number
Leaf	${}^{12}C^{5+}$	220	1378
Petal	${}^{12}C^{5+}$	220	776
Leaf	${}^{12}C^{6+}$	320	657
Petal	${}^{12}C^{6+}$	320	1461

Table 2 Changes of color indices b.

Individual	Dose	Bud	Full I	Bloom
	(Gy)		Inside	Outside
No.14	2	40.28	18.92	7.96
No.15	3	40.74	34.45	24.64
No.16	3	35.77	26.16	8.46
No.17	3	36.04	35.90	10.69
No.18	2	35.46	32.21	7.79
No.19	1	-	-	-
No.20	3	44.59	31.93	33.77
'Hama no hikari'	-	44.47	-	-
'Kishuuhou no tikara'	-	37.46	-	-
'Seikou kougyoku'	-	41.09	-	-
'Jinba'	-	11.16	2.02	2.41
Light yellow'Jinba'	-	30.84	23.67	1.57

3-26 DNA Damage Yields in Hydrated DNA Exposed to Neon Ion Particles

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Various types of DNA lesions, such as strand breaks, base lesions are generated after exposure of cells to ionizing radiation. One of the unique feature of the radiation-induced lesions is that they contain clustered damage sites, which consist of two or more elemental DNA lesions within one or two helical turns of DNA¹⁾. Clustered DNA damage sites are considered to be less readily repaired than isolated lesions and therefore may induce deleterious and serious genetic alterations in cells. The complexity of clustered damage is proposed to increase with increasing ionizing density of the radiation. It has been demonstrated that the direct effect of low LET (linear energy transfer) radiation produces 30-40% of the lesions in cells²⁾. This value increases up to 70% when cells are irradiated with α -particles²⁾. However, few investigations have been systematically undertaken to elucidate the nature of lesions produced by the direct effects of radiation with varying ionizing density.

To gain insights on the nature of lesions induced by the direct effect of ionizing radiation, yields of single- and double-strand breaks (ssb's and dsb's), and base lesions in hydrated plasmid DNA were measured after exposure to neon ions (²⁰Ne⁸⁺, 17.5 MeV/n, AVF cyclotron (TIARA, JAEA)) with different LETs. The LETs were calculated using the E-LOSSM code. In hydrated DNA, lesions would be generated from direct effect of the radiation, and indirect effect (attacking of DNA by radicals arising from bulk water) could be neglected. Base lesions were detected as additional strand breaks after treatment of irradiated DNA with two glycosylases, Nth and Fpg proteins, which convert the oxidative base lesions into ssbs.

The yield of prompt ssb after exposing hydrated plasmid DNA to neon ion particles with a LET of 361 keV/ μ m was



Fig. 1 Number of prompt and enzymatically induced single strand breaks (ssbs) as a function of LET (keV/µm). Prompt (○), Fpg+Nth treatment (●).

 5.8×10^{-11} ssb/Gy/Da (Fig. 1). This value of around 6×10^{-11} ssb/Gy/Da is comparable to that obtained after γ -irradiation³⁾. The yields were slightly smaller for neon ion particles with LETs of 491 and 842 keV/µm, which gave 4.8×10^{-11} and 4.7×10^{-11} ssb/Gy/Da, respectively. The yields of prompt dsb, they appear to be fairly constant in the LET range (Fig. 2). Consequently, the ratio of prompt ssb to dsb decreases as the LET increases, indicating that clustered strand breaks, some of which would produce dsb's, are more readily induced at higher LETs.

Results of enzymatically induced ssb and dsb revealed that the amount of base lesions susceptible to cleavage by Nth and Fpg become less as the LET gets higher (Fig. 1 and Fig. 2). We cannot fully disregard the possibility that the number of radiation-induced damaged bases is actually reduced at higher LET. However, we favor the interpretation that base lesions are clustered and thus become less readily cleaved by the glycosylases, as strand breaks shows a trend to be more clustered at higher LETs. A higher value of enzymatically induced ssb and dsb with neon ions at LET of 491 keV/ μ m compared to that with carbon ions at similar LET (507 keV/ μ m) also supports the conclusion that the amount of cleavable base lesions becomes less with increasing ionizing density⁴).

In summary, our results indicate that DNA lesions in hydrated plasmids become more readily clustered on increasing ionizing density of the radiation.

- 1) D. T. Goodhead, Int. J. Radiat. Biol. 65 (1994) 7.
- 2) C. M. de Lara, et al. Radiat. Res. 144 (1995) 43.
- 3) A. Yokoya, et al. J. Am. Chem. Soc. 31 (2002) 8859.
- 4) T. Ushigome, et al. JAEA-Review 2007-060 (2007) 97.



Fig. 2 Number of prompt and enzymatically induced double strand breaks (dsbs) as a function of LET (keV/μm). Prompt (°), Fpg+Nth treatment (•).

3-27 DNA Damages Analysis Induced by ⁴He²⁺ Ion Beam Compared to Those of ⁶⁰Co γ-rays in the Solid State

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1. Introduction

Information in relation to chemical structures and their distribution on DNA is quite important to clarify following biological repair procedures. In case of ionizing radiations, especially, it is expected that three-dimensional distribution of DNA damages is diversified as well as their chemical structures, and the diversity makes investigations of physicochemical processes in radiation effects difficult. In fact, double-strand breaks produced by high linear energy transfer (LET) alpha particles are more difficult to rejoin in living cells than those produced by low LET 137 Cs γ -rays¹). To investigate difference of DNA damage spectrum between radiations, we have developed new methodology to analyze strand break-termini and nucleobase lesions using snake venom phosphodiesterase (SVPD), calf intestinal alkaline phosphatase (CIAP) and piperidine²⁾. In this report, the results of the DNA damage spectra induced by ⁴He²⁺ ion beam and $^{60}\text{Co}\ \gamma\text{-rays}$ are demonstrated. The $^4\text{He}^{2+}$ ion beam used was obtained from TC1 port of 3MV tandem accelerator in TIARA at Takasaki Advanced Radiation Research Institute. The 60 Co γ -rays, as a standard radiation source, were irradiated in 7-cell in the 2nd building of γ -irradiation facility at the same institute.

2. Renewal of the irradiation system

The depth-tunable cell irradiation equipment³⁾ at TC1 port we have used and its current measurement system dysfunctioned owing to aging at an experiment in September. Then, we have restructured the system with higher userbility and better reproducibility in quantitative data. As a result, a value of beam current at the exit of the beamline became more stable around 10 pA ($\sim \pm 1$) than before. Moreover, the theoretical fluence (5.0×10^7 particles/nA/cm²/pass) was comparable with the fluence determined experimentally at 10 pA by pit counting on CR-39[®] dosimeter (4.4×10^7 particles/nA/cm²/pass) at a vertical velocity with 70 mm/s of the irradiation stage. Although there is a problem on difficulty of beam adjustment, we are sure that the TC1 port has been revived to use at least at low dose-rate.

3. Experiments

Linear formed pUC19 plasmid DNA digested by Sma I (pUC19/Sma I) was used. pUC19/Sma I aqueous solution was mounted on a silicon plate and dried thoroughly in vacuum under P_2O_5 . For γ -irradiation, the dried DNA sample on a plate was put into a glass tube sealed in vacuum to achieve secondary electron equilibrium. Each sample was irradiated at a dose (dose rate) of 50 (0.694), 100 (1.389), 200 (2.778), 400 (5.556), and 900 kGy (12.5 kGy/h) at r.t., respectively. For ⁴He ion beam, the DNA- mounted plate

was fixed in a cylindrical chamber (39 mm ϕ , hight: 22 mm) with a Kapton[®] window (8 µm) under argon gas. Each sample was irradiated with the beam with LET of ~ 85 keV/µm, which was controlled using the *renewal* depth-tunable cell irradiation equipment, was irradiated at a dose (dose rate) of 102 (1.068), 406 (1.068) and 913 kGy (507 Gy/pass) at r.t. (beam current: 90 nA). The doses were determined based on the number of pits on CR-39 dosimeter at 10 pA. The irradiated DNA samples were recovered by water at 0°C to be 0.32 µg/µL. The new method for DNA damage analysis is introduced in previous paper in datail²). Based on the series of the experiments, the yields of total 3'termini, those of 3'termini with or without phosphate, and piperidine-labile lesions were calculated.

4. Results and Discussion

Table 1 shows the yields of DNA lesions calculated. Additionally, the yields of unaltered nucleobase release are also demonstrated as a reference. At a glance, All DNA damage yields estimated for the ⁴He ion beam were three times larger than those for the γ -rays. This result implies either (1) the energy transfer efficiency of the ion beam to DNA is higher than that of the γ -rays, (2) it is difficult to extrapolate the real fluence at 90 nA from the experimentally-obtained fluence at 10 pA, or (3) the Faraday cup is not in position to receive whole beam. Anyhow, more accurate beamline adjustment should be needed to obtain trustful data to be discussed particularly at a high dose-rate.

Table 1 Yields of DNA lesions for ⁴He ion beam and 60 Co γ -rays

	Yield (µmol/J, x 10 ⁻⁹ Gy ⁻¹ Da ⁻¹)			
Category of lesions	⁶⁰ Co γ-rays (06405 irrad.)	⁶⁰ Co γ-rays (080422 irrad.)	⁴ He (~ 85 keV/μm) (080327 irrad.)	
Total 3' termini	0.102	0.080	0.238	
3'termini without phosphate	0.024	0.028	0.064	
3'termini with phosphate	0.078	0.052	0.174	
Piperidine-labile lesions	0.084	0.080	0.217	
Unaltered nucleobase release (ref.)	0.018	0.018	0.055	

Acknowledgments

We would like to gratefully thank several researchers in Takasaki, JAEA, Drs. Y. Kobayashi and M. Kikuchi for the γ -ray experiment, Drs. Y. Hase and Y. Sugo for maintaining and operating the TC beamline, Drs. S. Uno. M. Taguchi and Y. Saito for optimizing dosimetric system of TC1, Dr. Y. Yokota for supporting experimental dosimetry using CR-39.

- 1) C. M. deLala, et al., Radiat. Res. 144 (1995) 43.
- 2) K. Akamatsu, Anal. Biochem. 362 (2007) 229.
- A. Tanaka, et al., Nucl. Instr. Met. Phys. Res. B 129 (1997) 42.

3-28 Analysis of Mutagenic Effect Induced by Ion Beams for Breeding of Aspergillus oryzae

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We investigated the mutagenic effect induced by ion beams for breeding of *Aspergillus oryzae*, which is used in the manufacture of traditional Japanese fermented foods. We irradiated conidia of *A. oryzae* with 220 MeV carbon ions by AVF cyclotron at JAEA at doses of 300-500 Gy. Genomic southern analysis of *sB* gene indicated that large size deletions occurred in variants. Furthermore, pulsed-field gel electrophoresis revealed large structural alteration in chromosomal DNA pattern. These results suggest that ion beam irradiation is effective as a new mutation breeding method for *A. oryzae*.

【緒言】

麹菌(Aspergillus oryzae)は醤油、清酒、味噌など の醸造産業において利用される重要な微生物である。 そのため、麹菌の育種が古くから精力的に行われてき た。近年、植物などの育種においてイオンビームが注 目されており、その変異効果が従来の変異原に比べて 高く、変異スペクトルも様々であることが報告されて いる。そこで我々は、麹菌育種の新しい手段としてイ オンビームの利用を検討することとした。

前報より、凍結乾燥処理を行った麹菌分生子へのイ オンビーム照射では 400 Gy で変異効果が高く、遺伝 子変異が高い割合で生じていることが示唆された¹⁾。 そこで、このような遺伝子変異株について、ゲノミッ クサザン解析、パルスフィールド電気泳動などにより、 遺伝子レベル、ゲノム構造レベルにおいてイオンビー ム照射が麹菌へどのような影響を及ぼすのか検討した。

【実験方法】

A. oryzae (niaD300) の分生子を凍結乾燥し、照射サ ンプルとした。照射はTIARAのAVFサイクロトロンを 用いて加速した¹²C⁵⁺ (220 MeV, 107 keV/ μ m) で、300 ~500 Gy照射した。スクリーニング方法は前報に準じ た¹⁾。ゲノミックサザン解析には*sB* 遺伝子領域をプ ローブに用いた。パルスフィールド電気泳動のための 染色体DNAの調製は、既報を参考に行った²⁾。

【結果】

得られた全 51 株のセレン酸耐性株において、*sB* 領域のPCR解析を行ったところ、増幅の認められな かった株は 11 株であった。これらの株についてゲノ ミックサザン解析を行ったところ、Fig.1 に示すよう に、親株に比べてバンドパターンが大きく異なってい た。このことから、イオンビーム照射により、*sB* 遺 伝子領域に大規模な欠損や転位、挿入などの構造変化 が起きていることが予想された。また、No.11 に関し ては、数回のサザン解析の結果、約 2.5 kbp の遺伝子 欠損が生じていることが確認された。

そこで次に、この株の染色体構造について検討する ため、パルスフィールド電気泳動を行った。その結果、 泳動パターンが親株に比べて異なり、特に 8 s番染色 体が大規模に欠失していることが推察された (Fig. 2)。 以上のことから、イオンビーム照射は、麹菌遺伝子の 大規模な変異を生じさせ、かつ、染色体構造を大きく 変化させることが可能であり、麹菌の新しい変異育種 方法として有効であることが考えられた。



Fig. 1 Genomic southern analysis of the *sB* gene of *A*. *oryzae*. M, DNA size marker; Lane 1 to 11, variants; WT, wild type.



Fig. 2 Separation of *A. oryzae* chromosomal DNAs. M, *Schizosaccharomyces pombe* chromosomal DNAs as size markers; WT, niaD300 ; No.11, variants.

- Y. Toyoshima et al., JAEA Takasaki Annual Report 2006 (2007) 97.
- K. Kitamoto et al., Biosi. Biotech. Biochem. 58 (1994) 1467.

3-29 Deletion of Minor Enzyme Activities of *Rhizomucor miehei* by Heavy Ion Irradiation

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Heavy ion beam of 220 MeV ¹²C⁵⁺ and 350 MeV ²⁰Ne⁸⁺ was irradiated respectively at a dose of 200 Gy on *Rhizomucor miehei* sporagiospore to obtain low lipase activity mutants. Coagulation enzyme activity was used to evaluate the phenotype of irradiated samples. No mutant with low lipase activity was isolated but a few mutants with decreased coagulation activity were isolated. Mutant D-111, one of them, produced only 10% coagulation activity in comparison with its parent strain. Analyses of the cultivated broth by SDS-PAGE and anion-exchange chromatography revealed that the protein profile and productivity of the mutant were almost equal to those of parent strain. Several point mutations were found in a protease gene of the mutant, one mutation was located in a glycosylation site. No mutation was observed at the catalytic site of the protease. Deletion mutation was not found in the protease gene including its upstream and downstream regions.

Rhizomucor miehei(以下R. miehei)が菌体外に分泌 する凝乳酵素はチーズ製造に広く使用されている。遺 伝子組換え技術を応用して製造される組換えキモシン もチーズ製造に使用されているが、食品製造への遺伝 子組換え製品の使用は消費者の抵抗があり、非遺伝子 組換えの微生物凝乳酵素に対する需要が根強い。しか し、R. mieheiは凝乳酵素(乳蛋白質の特定部分を分解 するプロテアーゼの一種)を大量に生産する一方で異 なる部位を分解するプロテアーゼ、油脂分解酵素のリ パーゼ、及びアミラーゼなどチーズ製造には好ましく ない少量の酵素を生産する。遺伝子組換えには該当し ないイオンビーム照射による突然変異は、遺伝子の欠 失等が起きやすいとされており、このような夾雑酵素 活性の除去を目的とする突然変異原として好適と考え られる。

これまでに、He (50 MeV)、C (220 MeV)、Ne (350 MeV) 及びAr (460 MeV) の4種類のイオン ビームを胞子に照射し、照射線量と生存率を比較した 結果から、Cイオン及びNeイオンの照射に生物学的効 果が高いことを確認した。そこで、2種のイオンビー ムを200 Gy照射することとした。照射菌株は、2種類 の生産株(B及びD系統)を用いた²⁾。胞子は少量の界 面活性剤を添加した緩衝液に懸濁してシャーレに塗布 後、シリカゲルを用いて乾燥させて照射サンプルとし た。イオンビーム照射後、胞子を再懸濁液して寒天培 地上に生育したコロニー数より生存率を算出した。ま た、同時にアデニン要求性突然変異により赤色に発色 するコロニーの出現数から突然変異率を算出した1)。 除去する夾雑活性としてリパーゼ活性を対象とした。 寒天培地上に生育したコロニーを液体培養後、培養液 に分泌されたリパーゼ活性と凝乳活性を測定した。ま た、凝乳活性の生産が大きく低下した変異株について、 凝乳酵素の遺伝子解析を行った。

B系統生産株からは、オリーブ油を加えた寒天培地 上に形成されるハロウを指標に、2株のリパーゼ低生 産変異株を選抜した²⁾。得られた2株について、単胞 子分離を行い安定した低生産性変異株の確立を試みた が、不安定な生産性を示すのみであった。また、リ パーゼの生産性評価と併せて遺伝子解析を今後予定し ている。

炭素イオンビーム照射により、D系統生産株から凝 乳活性低下変異株を分離した。単胞子分離を行った後 に、液体培養して活性生産が約15%に低下しているこ とを確認した。凝乳活性低下の原因を明らかにするた め、液体培養後の菌体内と菌体外の酵素活性を比較し たところ、生産株及び凝乳活性低下変異株ともに、菌 体内1に対し菌体外5の比であり、菌体外への酵素の分 泌には異常がないことが分った。培養液のSDS-PAGE と陰イオン交換クロマトグラフィー解析においては、 凝乳活性低下変異株は全体にタンパク質分泌量が低下 していたが、生産されたタンパク質の種類に違いは殆 ど違いが見られなかった。これらのことから、タンパ ク質の合成・分泌系には異常がないことが示唆された。 遺伝子解析から、凝乳酵素に3ヶ所ある糖鎖結合 部位³⁾の1箇所に変異が起こり(Asp410Ser)、糖鎖が付 加されない酵素タンパク質を生産していると推定され た。糖鎖は酵素の耐熱性に影響するとされるが、 72 °C 10分の熱処理を加えたところ、野生型酵素はほ ぼ完全に失活するのに対して、変異型酵素は約60%の 残存活性が得られた。ただし、糖鎖結合部位以外にも 数箇所変異が起きているので、その影響も考慮する必 要がある。

- F. Jin et al., Biosci. Biotechnol. Biochem. 68 (2004) 656.
- K. Sakai et al., JAEA Takasaki Annual Report 2006 (2007) 95.
- 3) K. Murakami et al., J. Bacteriol. 176 (1994) 2635.

3-30 Mutation Breeding of Koji Mold Induced by Ion Beam

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Introduction

A defective mutant of the *pyrG* gene, which codes the orotidine 5'-phosphate decarboxylase, is used as a host cell for transformation system of *Aspergillus* species. The *pyrG* deficient mutant results in uracil auxotrophy and resists to 5-fluoroorotic acid (5FOA) that is a substrate analogue. It is difficult to obtain a *pyrG* deficient mutant of *Aspergillus* oryzae RIB40 by conventional mutagens without genetic manipulation.

In this report, we tried to obtain a pyrG deficient mutant of *A. oryzae* RIB40 by ion beam irradiation to improve the occurrence frequency and pattern of mutation in *Aspergillus* species.

Materials and Methods

(1) Isolation of 5FOA resistant mutants

The suspension containing 10^6 conidia (*A. oryzae* RIB40) were spread on Malts agar plate containing 10 mM uracil and incubated at 30 °C for 6 hours for germination. Germinated conidia were irradiated with 220 MeV $^{12}C^{5+}$ (121.8 keV/µm) ion beam accelerated by AVF cyclotron at TIARA, with the doses of 150 and 200 Gy. Irradiated conidia were cultivated at 30 °C for 3 days and resuspended in 0.01% Tween80 solution. The suspension containing 1×10⁶-irradiated conidia was spread out on Czapex-Dox medium containing 10 mM uracil and 3.0 mg/ml 5FOA. 5FOA resistant mutants were purified three times by single conidium isolation. The uracil auxotrophy of mutants was confirmed by the nonviability on Czapex-Dox medium. Sole mutant was picked up from each irradiated plate.

(2) DNA analysis of 5FOA resistant mutants.

The 5FOA resistant mutants with uracil auxotrophy result from not only the *pyrG* deficiency but also the *pyrF* deficiency that encodes an orotate phosphoribosyl-transferase¹). The 5FOA resistant mutants were transformed by PCR fragment of the *pyrG* gene and/or *pyrF* gene amplified from wild type for the genetic complementation analysis. The *pyrG* or *pyrF* gene of each mutant was cloned and sequenced to detect the mutation pattern.

Results and Discussion

The thirty-three mutants resistant to 5FOA with uracil auxotrophy were obtained from 5×10^8 of irradiated conidia (50 irradiated plates). Reversion was observed in the eight mutants. Among the remaining twenty-five mutants, the thirteen mutants were complemented by *pyrG* and the twelve mutants were complemented by *pyrF* gene. To investigate the type of mutations, the *pyrG* gene or *pyrF* gene of each mutant was cloned and sequenced respectively. Among thirteen *pyrG* deficient mutants, two mutants had single base deletion mutations in the coding region and resulted in frameshift mutations. The others had single base substitutions in the coding region that causes nonsense mutations. All the sites of base substitution were different from each other and the number of transversion was approximately equal to that of transition (Table 1).

On the other hand, among twelve pyrF deficient mutants, six mutants had deletions and the others had single base substitutions. Among six deletion mutants, two had regional deletions (25 base- or 1,409 base-deletion), while remaining had single base deletions in the coding region. Among six single base substitution mutants, mutations occurred at the one of the initiation codon in four mutants. The initiation codon of pyrF gene might be a hot spot for ion beam irradiation.

In conclusion, it was easy to obtain *pyrG* deficient mutants but also *pyrF* deficient mutants of *A. oryzae* RIB40 without genetic manipulation by means of ion beam irradiation. Mutagenesis by ion beam is a powerful tool to obtain a novel mutant in *Aspergillus* species especially for the gene that are tough to obtain by conventional mutagens.

Table 1. Mutation patterns of 5FOA resistant mutats

Mutation nottoms	pyrG mutants	pyrF mutants		
Nutation patterns -	Number of mutants			
Base substitution	<u>11</u>	<u>6</u>		
transition	5	3		
GC→AT	2	1		
CG→TA	3			
TA→CG		2		
transversion	6	3		
GC→TA	3			
CG→CG		2		
CG→TA	2			
AT→TA	- 1			
TA→GC		1		
Deletion	2	<u>6</u>		
Total	13	<u>12</u>		

Reference

1) Skory et al., Appl. Environ. Microbiol. 56 (1990) 3315.

3-31 ESR Detection Method of Irradiated Black Pepper

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ESR techniques for studying the detection of radicals induced in gamma irradiated pepper were presented. In particular, the saturation behaviors of radicals, as well as threshold value of ESR peak intensity, were illustrated in the case of radicals observed in irradiated pepper. With a standard addition method for solid sampling technique we revealed a novel detection method for irradiated pepper.

照射食品の検知法として近年,熱ルミネセンス (Thermoluminescence, TL)法が導入された。しかし 輸入胡椒の実用的検査法としては十分ではないことが 報告されている¹⁾。EUではTL法は照射食品の公定法 として利用されている²⁾。同様に電子スピン共鳴 (Electron Spin Resonance, ESR)法もEU公定法に導入さ れている²⁾。著者らは、照射誘導ラジカルの逐次飽和 挙動を詳細に検討しESRを用いた照射食品厳密分析法 を確立し、EU公定法の改良法として提唱している³⁾。 海外では、胡椒のガンマ線照射による殺菌処理が実用 化されている²⁾。我が国では胡椒を輸入しているので、 照射殺菌胡椒が入ってくる可能性がある。そこで、本 研究では、ESRによる照射食品検知法の実用化を目的 とし、照射胡椒検査法について検討した。

試料はインドより輸入した黒胡椒である。収穫後乾 燥処理したものであり殺菌処理はしていない。照射処 理はガンマ線を用い、原子力機構高崎量子応用研究所 にて行った。照射量は7 kGyである。ESR試料管に未 処理胡椒、照射胡椒、混合試料(照射黒胡椒含有率:2%, 5%, 10%, 20%)を入れ、酸素の影響をなくすためにAr 置換をして封じ入れ⁴⁾、ESR計測に用いた。

黒胡椒のESRスペクトルはFig.1に示すように3種 であり、最も強く鋭い一本線はg=2.0に観測され、有 機フリーラジカルと推定される。六本線はMn²⁺の超 微細構造線である。Fe³⁺由来の信号はg=4.0近くに観 測された。Fig.2に、有機フリーラジカルの信号強度 の変化を示した。マイクロ波強度が増すに従い、いず れの試料においてもESR信号強度は増大し飽和点に達 した。しかし、一定のマイクロ波強度のときに観測さ れたことから、照射処理により飽和点が変動しないこ とがわかった。



Fig. 1 ESR spectrum of pepper.



Fig. 2 Threshold value of black pepper before and after irradiation.

このことは、照射胡椒混合比が未知の試料でもESR 信号の飽和挙動には本質的な差異はない事を示唆して いる。混合試料のESR信号強度を混合比に対してプ ロットすると(Fig. 3)、直線で近似することが可能 であったことから、標準添加法として有用であるので、 実用的検知法¹⁾としてESR法が導入できると結論した。



Fig. 3 ESR peak intensity of radiation induced radical at various irradiation dose.

- 1) 後藤典子 et al., RADIOISOTOPES 56 (2007) 103.
- 2) 等々力節子, 食品照射, 40, (2005) 49.
- M. Ukai, et al., Spectrochimica Acta A63 (2006) 879.
- 4) 亀谷宏美 et al., RADIOISOTOPES 56 (2007) 437.

3-32 Practical Application of Chemiluminescence ELISA for the Detection of Oxidative DNA Base Damage in Irradiated Meats Using Anti-8-OHdG Antibody

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Food irradiation is carried out for benefits of public health, the reduction of post-harvest losses and quarantine treatment of certain foods¹⁾. Consumers can choose irradiated or un-irradiated foods following its label²⁾. Detection methods for irradiated foods are required to endorse the reliability of labels. To date, physical, chemical and biological detection methods have been developed³⁾. However, there are various kinds of difficulties on practical use of those methods. Therefore, an easy versatile detection method for irradiated foods is necessary for appropriate management. For the purpose, we focus to detect oxidative DNA damages as a common target on living things or foods using ELISA technique. Preliminary data for the affinity of anti-8-OHdG antibody against 8-oxoG in DNA were reported⁴). Toward the practical use of our novel ELISA method, precise detection for various meats will be described here.

Ground meats purchased from a market were irradiated to 60 Co γ rays with a dose rate of 3 kGy/h at TARRI/JAEA. Genomic DNAs of the meat were isolated using DNeasy Tissue kit and eluted in sterilized water. DNA concentration was measured at 260 nm. The meat DNAs were heat-denatured, mixed with anti-8-OHdG antibody, and put into wells of protamine-coated microtiter plate. The plate was incubated for 3 h at room temperature. After several washing, those were incubated with peroxidase-labeled secondary antibody for 1 h. The plate was washed five times, and then LumiGLO Reserve substrate was added into the wells. Chemiluminescence was measured using Lumi-Imager.

To confirm the distinguishable dose of 8-oxoG using ELISA method, DNAs isolated from irradiated meats with various doses were investigated. As shown in Fig. 1, the chemiluminescence intensity increased with increasing doses although non-irradiated sample didn't give the value of zero since spontaneous 8-oxoG lesions occur on DNA due to OH radicals that come from endogenous metabolism in its alive



Fig. 1 Detection of 8-oxoG for irradiated meats using ELISA. Each point represents the mean value and standard error from three independent experiments.

period. A linear dose-intensity response for irradiated beef was observed up to 9 kGy, while that for chicken meat seemed to be within 6 kGy, and that for pork might be up to 3 kGy.

To examine the effect of long period storage, we detect 8-oxoG in irradiated meats after 1-year storage at -20 °C. Our detection method was capable of showing the dose dependency of the intensity related to 8-oxoG content in the meat samples (Fig. 2). We think that the release of 8-oxoGs was seldom occurred in cells of meat, especially under normal storage condition. Therefore, this method is applicable to detect 8-oxoG even after long storage.

Although most food irradiation is carried out below 10 kGy^{5} , our experiments dealt with meat samples subjected to irradiation up to 12 kGy. In the case of meat, 20 mg sample was sufficient for the 8-oxoG detection, and 3 kGy- or higher-irradiated meats were expected to be distinguished from non-irradiated samples⁶. This method has a potential that can be applied to most foods that contain DNA, perhaps including grains and fruits to be examined in future.

References

- 1) Anon., STI/PUB/788, IAEA (1989) 135.
- 2) Anon., CODEX STAN 106-1983, REV.1-2003, (2003).
- 3) S. Todoriki, Food Irradiation, Japan (2005) 49.
- M. Kikuchi et al, JAEA Takasaki Annual Report 2005 (2007) 72.
- 5) Anon., Clearance of Irradiated Food Database, IAEA.
- 6) M. Kikuchi et al., RADIOISOTOPES, 56 (2007) 509.

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Fig. 2 Detection of 8-oxoG for irradiated meats after 1-year storage in freezer using ELISA.

3-33 Cell-killing Effect of Heavy Ions Having Different LETs in Immortalized Human Cells

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Introduction: High-LET ion beams have greater biological effects (e.g. reproductive death, chromosome aberration and mutation) than low-LET radiations¹⁻³⁾. As a part of its mechanism, it is expected that DNA damage induced by high-LET ions are complex and difficult to repair precisely. However, they are not understood well how complex DNA damage is induced and how do the DNA damage causes great biological effects. In this study, we investigated the cell-killing effect of ion beams having different LETs in immortalized normal human cells.

Materials & Methods: Telomerase-immortalized human diploid fibroblast BJ-hTERT cells were seeded at 5×10^3 cells/cm². Culture medium was refed on day 4, and confluent cell monolayer was irradiated with γ -rays (LET=0.2 keV/µm), helium (17 keV/µm), carbon (70, 110, 150 and 212 keV/µm), neon (310 and 430 keV/µm) and argon ions (1320 and 1530 keV/µm) on day 7. Cell survival was determined using a clonogenic survival assay ²). Surviving fraction was obtained by dividing the colony formation rate of irradiated cells by that of sham-irradiated cells. Survival data were fitted to the equations: $y = 1 - [1 - exp (-ax)]^b$ for γ -rays and helium ions or y = exp (-ax) for carbon and heavier ions where *a* and *b* are the coefficients, *x* and *y* are the dose and surviving fraction, respectively. The dose needed to reduce the surviving fraction to 0.1 (D₁₀)



Fig. 1 Survival curves. Carbon ion irradiation showed the steepest slope among the radiations used in this study. Each point represents mean ± standard deviation of 2-4 independent experiments. Only the results of some radiations are shown here to avoid congestion.

was calculated from the equations. Relative biological effectiveness (RBE) was calculated by dividing the D_{10} of γ -rays by that of ion beams.

Results and Discussion: Slopes of the survival curves were dependent on LET and became steepest at carbon ions (Fig. 1). RBE based on D_{10} arrived at the maximum of 4 at 100-200 keV/µm of carbon ions (Fig. 2), indicating that the cell-killing effect of carbon ions per dose was almost fourfold greater than that of γ -rays. This LET-RBE relationship is similar to the reported result in normal human fibroblasts having a finite lifetime²), raising the possibility that BJ-hTERT cells are well suited for radiobiological studies because of their extended lifetime without transformed phenotype^{4,5)}. In future plan, we will quantitatively analyze the initial induction and rejoining of DNA double-strand breaks that are induced at intervals of one hundred bp to several mega bp for better understanding of high-LET ion-induced DNA damage.

- 1) Y. Yokota et al., Int. J. Radiat. Biol. 79 (2003) 681.
- 2) N. Hamada et al., Radiat. Res. 166 (2006) 24.
- 3) Y. Yokota et al., Radiat. Res. 167 (2007) 94.
- 4) A. G. Bodnar et al., Science 279 (1998) 349.
- 5) X. Jiang et al., Nat. Genet. 21 (1999) 111.



Fig. 2 LET-RBE relationship on clonogenic survival. RBE peaked at 100-200 keV/μm of carbon ions.

3-34 Effects of Ionizing Radiation on Learning Behavior of *Caenorhabditis elegans*

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Ionizing radiation (IR) induced learning impairment and abnormalities in the nervous system are important potential risks in interplanetary space missions and radiation therapy. Using *Caenorhabditis elegans* (*C. elegans*) as a neuronal model organism, we have studied the effects of IR on the learning behavior, and found the followings; (1) the modulatory effect of IR on salt chemotaxis learning via gamma subunit of G protein¹, (2) the difference of initial effects of gamma-ray irradiation on olfactory adaptation to benzaldehyde², and (3) the effects of carbon ion irradiation on locomotion. Here, we report the new findings in 2007.

(1) Modulatory effect of gamma irradiation on salt chemotaxis learning. We reported that gamma-ray exposure influences chemotaxis to NaCl only during the transition stage of salt chemotaxis learning³⁾. To investigate the mechanism by which this occurs, we used the gpc-1 mutant, which is defective in GPC-1 (one of the two gamma subunits of the heterotrimeric G-protein) and responsible for the chemosensory avoidance of NaCl. This mutants learn by combined stimulation with starvation and exposure to NaCl as well as wild type animals. Wild type and gpc-1 mutant animals were exposed to gamma-rays 30-60 min after the start of salt chemotaxis learning conditioning. Immediately after irradiation, we measured the chemotaxis to NaCl. We found that IR-induced decreases in chemotaxis were significantly suppressed in the gpc-1 mutant (Fig. 1). The result suggested that the effect of IR on salt chemotaxis learning in C. elegans was mediated by GPC-1.



Fig. 1 Decrease of chemotaxis in wild type and *gpc-1* mutant animals. The error bars represent SE of 4 or more experiments. * indicates the significant difference (p < 0.05).

(2) Effects of gamma irradiation on olfactory adaptation. To investigate the effect of IR on learning behavior of *C. elegans* ohter than salt chamotaxis learning, we employed the olfactory adaptation to benzaldehyde, and examined the effect of IR on the transition stage of the adapation to benzaldehyde. Animals were exposed to gamma-rays 15-30 min after the start of benzaldehyde adaptation. Immediately after irradiation, we measured the chemotaxis to benzaldehyde (Fig. 2). There was no significant decrease in chemotaxis to benzaldehyde of *C.elegans*, indicating the difference in the IR-induced response between the adaptation to benzaldehyde and the salt chemotaxis learning.



Fig. 2 No significant difference in the chemotaxis to benzaldehyde between sham-irradiated and irradiated wild type animals. The error bars represent SE of 4 or more experiments.

(3) Effects of carbon ion irradiation on locomotion. To test whether IR induces any decrease in locmotion, we examined the effect of gamma and carbon irradiation. After gamma irradiation at the dose of 500 Gy, exposed animals showed the decrease of 30% in body-bends of the non-irradiated ones. On the other hand, carbon exposed animals showed the normal locomotion. The results contradict to the fact that high linear energy transfer (LET) radiation induces more severe biological effects than low-LET radiations.

- 1) T. Sakashita et al., FASEB J. 22 (2008) 713.
- 2) T. Sakashita et al., Biol. Sci. Space 21 (2007) 117.
- T. Sakashita et al., JAEA Takasaki Annual Report 2006 (2007) 110.

3-35 Utilization of New Ion Species for Analysis of Heavy-ion Radiation-induced Bystander Effect

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Using the collimated heavy-ion microbeam system, we have been carried out the analysis of bystander effect for assessing the effect of heavy-ion radiation on biological system ¹⁻²⁾. The only ion species that were utilized in heavy-ion microbeam system and has more than 1,000 keV/ μ m of LET has been ${}^{40}Ar^{13+}$ 460 MeV (LET=1,260 keV/µm). However this ion has only 260 µm projectile range in water. Although the medium of target sample is removed before irradiation to make the ion penetrate whole sample and the ion to detect and count by scintillator-PMT assembly installed in opposite side of beam exit, it sometime became impossible to detect ions according to the media-removal condition. Therefore, for solving this issue, we adopted ⁴⁰Ar¹⁴⁺ 520 MeV ions (LET=1,130 keV/µm), which become available recently and have a projectile range in water of 320 µm, and verified whether this ion species can be utilized in the target irradiation of individual cells by heavy-ion microbeam system. In addition, generation of cocktail microbeam of 40 Ar¹⁴⁺ 520 MeV and 20 Ne⁷⁺ 260 MeV were also tested.

To confirm the feasibility of collimated microbeam of ${}^{40}\text{Ar}{}^{14+}$ ion, we first carried out the irradiation of CR-39 film. The energy profile of ions extracted into the air via microaperture of 5 or 20 μ m in diameter showed clear energy distribution peak without low-energy ion mainly arisen by scattering at the edge of the collimator, indicating successful generation of collimated microbeam of ${}^{40}\text{Ar}{}^{14+}$ ion. The special distribution of ion track visualized by etching of irradiated CR-39 film indicated that the ion hit was localized within the size of collimator used for microbeam generation (Fig. 2a).



Fig. 1 Energy distribution of ions irradiated on CR-39. The energy of ions penetrated CR-39 film was detected by PMT and measured by MCA.

We next verified that the cocktail beam of ⁴⁰Ar¹⁴⁺ and ²⁰Ne⁷⁺ is able to utilize in our collimated heavy-ion microbeam system. By changing the acceleration frequency of AVF cyclotron, we were able to obtain two different energy profiles of extracted ions within 5 min without re-tuning of beam line and microbeam end station. The etched pit visualized on CR-39 film indicated that (1) there were no significant differences in special distribution of ion, and (2) the size of etched pit showed clear difference between different acceleration frequencies, i.e. different ion species (Fig. 2). These result indicated that, by changing the acceleration frequency of AVF cyclotron, we can rapidly switch the ion species each other at any time and irradiate on individual cell target in our collimated heavy-ion microbeam system.

From these results, we concluded that ${}^{40}\text{Ar}^{14+}$ ion and cocktail beam of ${}^{40}\text{Ar}^{14+}$ and ${}^{20}\text{Ne}^{7+}$ ion is able to utilize in our collimated heavy-ion microbeam system. The ion of ${}^{40}\text{Ar}^{14+}$ will solve the issue of unstable count of hit ions on cellular targets. Moreover, the rapid switching of ion species using cocktail microbeam makes enable to irradiate multiple ion species within single beam time, so that it will be useful for comparing the ion hit effect of different ion species on biological system. Taken all together, this utilization of new ion species will accelerate the analysis of radiation induced bystander effect, and helps understanding the effect of heavy-ion radiation on biological system.

References

- 1) T. Funayama et al., J. Radiat. Res. 49 (2008) 71.
- 2) T. Funayama et al., IEEE Trans. Plasma. Sci. 36 (2008) *in press.*



Fig. 2 Irradiation of CR-39 with collimated microbeam of ${}^{40}\text{Ar}^{14+}$ or ${}^{20}\text{Ne}^{7+}$ ions. A film of CR-39 was irradiated with collimated microbeam of 20 μ m in diameter (10 particles). The ion tracks were visualized as etched pit by etching with alkaline solution (13.4 N KOH, 75 °C, 5 min). (a) Ion track of ${}^{40}\text{Ar}^{14+}$ 520 MeV microbeam. (b) Ion track of ${}^{20}\text{Ne}^{7+}$ 260 MeV microbeam.

3-36 Distinct Response of Irradiated Normal Human Fibroblasts and Their Bystander Cells to Heavy Ions

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Convincing experimental evidence has accumulated demonstrating that ionizing radiation induces biological effects in nonirradiated bystander cells having received signals from directly irradiated cells¹). However, little information is available hitherto as to the potential impact of energetic heavy ions on the bystander effect. Taking into consideration that fewer irradiated cells should coexist with more nonirradiated counterparts in a population exposed to a lower dose of higher-LET heavy ions, a clarification of the effects arising not merely in irradiated cells but in their bystander cells would be crucial to comprehend the mechanism of biological action of heavy ions.

Herein, we set out to examine heavy ion-induced bystander effects in confluent density-inhibited fibroblast cultures. AG01522D primary normal human diploid fibroblasts at passages 7-9 were inoculated at 1×10^4 cells/cm², replenished on days 4, 7 and 9, and used for irradiation experiments on day $11^{2,3}$. To see the effects in bystander cells, we employed precise microbeams of carbon ions (18.3 MeV/u, 103 keV/µm) and neon ions (13.0 MeV/u, 375 keV/µm) for targeting only a very small fraction of cells in confluent cultures⁴. Convertional broadfield irradiation with broadbeams of carbon ions (18.3 MeV/u, 108 keV/µm) and neon ions (13.0 MeV/u, 437 keV/µm) was conducted in parallel to see the effects in irradiated cells^{5,6}.

First, cell survival was determined by the clonogenic survival assay. Exposure of 0.00026% of cells led to nearly 10% reductions in the survival regardless of ion species⁷). Second, apoptosis was detected by the terminal deoxynucleotidyl transferase-mediated dUTP-biotin nick-end labeling method. Irradiation of 0.00026% of cells caused twofold rises in the apoptotic incidence irrespective of ion species⁷⁾. Whilst apoptotic frequency increased with time up to 72 h postirradiation in irradiated cells, its frequency escalated up to 24 h postirradiation but declined at 48 h postirradiation in bystander cells, indicating that bystander cells exhibit transient commitment to apoptosis⁷). Third, p53 phosphorylation was analyzed with western blotting. Carbon- and neon-ion microbeam irradiation similarly caused almost twofold increments in the levels of serine 15-phosphorylated p53 proteins, irrespective of whether 0.00026, 0.0013 or 0.0066% of cells were targeted. Whereas the levels of phosphorylated p53 protein were

elevated and remained unchanged at 2 h and 6 h postirradiation in irradiated cells, its levels rose at 6 h postirradiation but not at 2 h postirradiation in bystander cells, suggesting that bystander cells manifest delayed p53 phosphorylation⁷⁾. Finally, gene expression changes in carbon ion-irradiated cells and their bystander cells were evaluated with the microarray analysis. Irrespective of the target numbers (0.00026, 0.0013 or 0.0066% of cells) and the time (2 or 6 h postirradiation), similar expression changes were observed in bystander cells⁸⁾. Nearly three quarters of the genes whose expression changed in bystander cells were downregulated, and most of the genes upregulated in irradiated cells were downregulated in bystander cells⁸⁾. Pathway analysis revealed serial activation of G protein/PI-3 kinase pathways in bystander cells, but that of p21^{Wafl} and NFKB pathways in irradiated cells⁸⁾. Taken together, interleukin genes were upregulated in irradiated cells while its receptor gene was upregulated in bystander cells, indicative of the signal transmission from irradiated to bystander cells⁸⁾.

In conclusion, we have demonstrated that exposure of less than 0.01% of cells to heavy ions results in bystander responses, which can be manifested as inactivated clonogenic potential, a transient apoptotic response, delayed p53 phosphorylation, and gene expression changes at a genome-wide level. The data highlight distinct response of irradiated cells and their bystander cells. These induced bystander responses could be a defensive mechanism that would avert or minimize further expansion of aberrant cells.

- 1) N. Hamada et al., J. Radiat. Res. 48 (2007) 87.
- 2) N. Hamada et al., Radiat. Res. 166 (2006) 24.
- 3) N. Hamada et al., Mutat. Res. 637 (2008) 190.
- T. Funayama, N. Hamada et al. IEEE Trans. Plasma Sci. 36 (2008) in press.
- N. Hamada et al., Radiother. Oncol. (2008) in press, doi: 10.1016/j.radonc.2008.02.013.
- N. Hamada et al., Cancer Lett. (2008) in press, doi: 10.1016/j.canlet.2008.03.029.
- 7) N. Hamada et al., Mutat. Res. 639 (2008) 35.
- M. Iwakawa, N. Hamada et al., Mutat. Res. (2008) in press, doi: 10.1016/j.mrfmmm.2008.04.007.
3-37 Heavy Ion-induced Bystander Killing of Human Lung Cancer Cells: the Role of Gap Junctional Intercellular Communication

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To clarify the mechanism of cell death induced by heavy-ion irradiation, human lung cancer cells were exposed to X-rays or carbon ions and cell survival was tested by clonogenic assays. Targeted exposure of 0.0004-0.002% of cells within the confluent culture resulted in 8-14% reduction of surviving fraction. Furthermore, this marked reduction was diminished when cells were treated with the inhibitor of gap junction. These results suggest that gap junctional intercellular communication may play an important role in the mechanism of heavy charged particle irradiation-induced bystander effect.

1. はじめに

これまでの我々の実験から, 肺癌細胞において, カーボンイオン照射のD₁₀(10%生存線量)とX線照射 のD₁₀はそれぞれ4.16 Gy、2.46 Gyであった。よって D₁₀における生物学的効果比は1.60であった。このよ うにX線よりも高い殺細胞効果を有する重イオンビー ムであるが,その細胞死の機序にバイスタンダー効果 が関与していることが知られている。本研究ではバイ スタンダー効果における細胞間のギャップ結合の関与 に注目し、重イオンビーム照射後の細胞生残率につい て検討を行った。

2. 実験方法

細胞はヒト肺癌由来の細胞株A549を用いた(p53 statusは野生型)。重イオン照射は原子力機構TIARA のマイクロビームにて行った(220 MeV¹²C⁵⁺, 108 keV/µm)。照射後の細胞生存率はコロニー形成法で 検討した。細胞を高密度(コンフルエント)と低密度 に培養した状態で照射するため、照射2日前に底面に 固体飛跡検出材(CR-39)を貼付した35 mmのグラス ベースディッシュ上に細胞を準備した(高密度群では 5×10⁵個、低密度群では5×10⁴個)。標的細胞はディッ シュ中の1、5または25個の細胞をランダムに選択し、 照射するイオンの数は一つの細胞あたり1,5または10 個とした。照射後は60 mmのディッシュに至適個数の 細胞をまきなおし、14日後に細胞を固定・染色した後、 コロニー数をカウントし、非照射細胞の生存率を対照 とし細胞生存率を検討した。また、高密度に培養した 細胞ではギャップ結合の促進剤(8-Br-cAMP:シグマ 社製:照射の48時間前に添加し1 mMに調節)、およ び阻害剤(Lindane:シグマ社製:照射の2時間前に添 加し0.1 mMに調節)を加えて照射し細胞生残率を求 め、ギャップ結合の関与について検討した。

3. 結果および考察

コンフルエントな状態の細胞のうち選択的に1つの 細胞にのみイオンを照射した際の細胞生存率は非照射 細胞とほぼ同様であった。照射細胞数をディッシュ内 の5個または25個、また各細胞への照射イオン数を5個 または10個としたとき,照射していない細胞の生存率 に比較し8-14%低下した(P<0.05:データの提示な し)。一方、低密度に細胞を培養し、細胞間の接触を なくした状態で同様に照射したときは、細胞生残率は 非照射の細胞と同様であった(データの提示なし)。 コンフルエントに培養した細胞でギャップ結合の促進 剤(cAMP)を併用して照射すると、照射単独に比べ 細胞生残率の低下が大きくなる傾向があった(Fig. 1の ①, p=0.07)。逆にギャップ結合の阻害剤(lindane) を併用して照射したときは、照射単独に比べ細胞生残 率の低下が小さくなった(Fig. 1の②, P<0.05)。

本実験ではディッシュ全体の細胞のうち0.0004-0.002%という極少数の細胞を重イオンビームで照射 した時、ディッシュ全体の細胞生存率が有意に低下す る結果が得られたが、細胞間の接触をなくした状態で 同様の実験を行った場合にはこの現象は認められな かった。またギャップ結合の阻害剤を細胞に接触させ ることにより照射後の細胞生残率低下は抑制された。 これらより重イオン照射による細胞死に照射細胞から 非照射細胞へのバイスタンダー効果が関わっており、 また、そのシグナル伝達経路にギャップ結合が深く関 与していることが示唆された。





3-38 Radiation Induces Upregulation of Myogenic Genes in Drosophila Schneider Cells

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The cells exposed to genotoxic stress, such as ionizing-radiation and DNA damaging agents, either arrest the cell cycle to repair the genome, or undergo apoptosis, depending on the extent of the DNA damage. DNA damage has been implicated in various differentiation processes. It has been reported that gamma-ray exposure or treatment with DNA damaging agent such as neocarzinostatin and etoposide could induce myogenic differentiation in Schneider cells although the mechanism underlying this process was poorly understood.

Exposure of *Drosophila* Schneider cells to 20 Gy of C^{5+} heavy ion beam or X-rays caused both increase of TUNEL-positive cells and conversion of round-shape cells to elongated cells (Fig. 1) confirming the previous report. Quantitative RT-PCR analysis showed upregulation of genes related to myogenesis including *Mef2* which is a MADS-box family of transcription factor, *nautilus* which is *MyoD* ortholog, and *blown fuse* which is required for proper myoblast fusion. Immunostaining with anti-myosin heavy chain antibody also confirmed that the morphological

changes were accompanied with myogenic differentiation.

It has been known that extracellular stresses increased intracellular ceramide concentration, and exogenous ceramide was able to induce apoptosis in mammalian cells. We observed that intracellular ceramide level was increased in Schneider cells after exposure to radiation such as UV, and X-rays. We examined whether exogenous ceramide could mimic radiation induced myogenic differentiation. Addition of membrane permeable C2-ceramide, which has shortened fatty acyl chain, to Schneider cells also increased TUNEL-positive cells and cells with elongated morphology. Induction of myogenesis related-genes was also confirmed by real-time RT-PCR analysis and immunostaining with anti-myosin antibody. It has been reported that no differentiation was observed when cells were treated with non-genotoxic inhibitors of cell cycle, indicating that cell cycle arrest is not the trigger of the differentiation process.

These results suggested that ceramide plays important roles in both apoptosis and the radiation-induced myogenic differentiation process.



Fig. 1 Heavy ion irradiation induced apoptosis and morphological changes in *Drosophila* Schneider cells. The Schneider line 2 cells were irradiated with 20 Gy of heavy ion beam (A). Untreated control cells are shown in (B). Cells were fixed and subjected to TUNEL assay 48 hours after irradiation. TUNEL positive cells are stained with purple dye. Arrows indicates cells with elongated morphology. Notice that both TUNEL positive cells and cells with extended morphology is increased in irradiated cells compared to unirradiated control cells.

3-39 Proliferation and Cell Death of Human Glioblastoma Cells After Carbon-ion beam Exposure: Morphologic and Morphometric Analyses

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Purpose

Histological analyses of glioblastoma cells after carbon-ion exposure are still limited and ultrastructural characteristics have not been investigated in detail. Here we report the results of morphological and morphometric analyses of a human glioblastoma cell line, CGNH-89¹⁾, after ionizing radiation to characterize the effect of a carbon-beam on glioblastoma cells.

Materials and Methods

CGNH-89 cells with a G to A transition in codon 280 in exon 8 of the TP53 gene had nuclei with pleomorphism, marked nuclear atypia and brisk mitotic activity.

For X-ray and heavy-ion beam exposure, CGNH89 cells were divided into 3 groups; control, 5 Gy and 10 Gy exposure. For each group, cells were fixed at the same time-points. Hematoxylin-Eosin (HE) and immunohistochemical staining were done in formalin-fixed cells. For electron microscopy, cells were fixed in fixative (1% glutaraldehyde, 10% formaldehyde). These samples were postfixed with osmium and embedded in Quetol 812. Ultrathin sections were examined under a transmission electron microscope.

Results and Discussion

After carbon-ion and X-ray exposure, living cells showed decreased cell number (Fig. 1), nuclear condensation, increased atypical mitotic figures, and a tendency of cytoplasmic enlargement at the level of light microscopy. The deviation of the nuclear area size increased during 48 hours after irradiation (Fig. 2), while the small cell fraction increased in 336 hours. In glioblastoma cells of the control, 5 Gy carbon-beam, and 10 Gy carbon-beam, and MIB-1 labeling index decreased in 24 hours (12%, 11%, 7%, respectively) but increased in 48 hours (10%, 20%, 21%, respectively). Ultrastructurally, cellular enlargement seemed to depend on vacuolation, swelling of mitochondria, and increase of cellular organelles, such as the cytoskeleton and secondary lysosome. We could not observe apoptotic bodies in the CGNH-89 cells under any conditions. We conclude that carbon-ion irradiation induced cell death and senescence in a glioblastoma cell line with mutant TP53. Our results indicated that the increase of large cells with enlarged and bizarre nuclei, swollen mitochondria, and

secondary lysosome occurred in glioblastoma cells after carbon-beam exposure.



Fig. 1 Cell counts per 1mm² of CGNH-89 cells before and after irradiation. In non-irradiated cells, cell numbers increase over time. In irradiated cells, cell numbers show a tendency to decrease dependently dose over time. Control: non-irradiated CGNH-89 cells. 1 Gy, 5 Gy, and 10 Gy: CGNH-89 cells irradiated by carbon-ion of 1 Gy, 5 Gy and 10 Gy.



Fig. 2 In CGNH-89 cells irradiated by carbon-ion at 48 hours, with dose response, the deviation of the nuclear area size show a tendency to increase dose dependently.

Reference

1) S. Ishiuchi et al, Nature medicine 8 (2002) 971.

3-40 Irradiation with Carbon Ion Beam Induces Cellular Senescence in a Human Glioma-Derived Cell Line

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A morphological phenotype consistent with cellular senescence, that is, an enlarged and flattened appearance, was observed in a human glioma-derived cell line, NP-2, after irradiation with carbon ion beam (18.3 MeV/amu ¹²C). The senescent nature of these cells was further indicated by positive staining for SA- β -gal activity, accumulation of lipofuscin, and marked increase in lysosomal mass. Most of SA- β -gal-positive NP-2 cells did not incorporate BrdU, indicating that these senescent cells did not synthesize DNA. We also detected phosphorylated p53 (Ser15) and induction of p21 in irradiated NP-2 cells, suggesting that p53-p21 pathway might play an important role in induction of cellular senescence in NP-2 cells after irradiation with carbon ion beam.

老化細胞は、非可逆的な細胞増殖の停止と特徴的な 表現型 [細胞質の扁平化と巨大化、細胞内顆粒の増加、 Senescence-associated β-galactocidase (SA-β-gal)活性の 増加、自家蛍光の増大など]を示す細胞と定義されて いる。われわれは、重粒子線 (炭素線 18.3 MeV/amu ¹²C)を照射したヒトグリオーマ由来 NP-2 細胞に、こ れらすべての老化細胞の特徴を示すマーカーを検出 した。6 Gy 照射では照射後 10 日ぐらいでは大部分の 細胞 (99%以上)が細胞死となり浮遊しているが、残 りのディッシュに付着した 1%以下のほぼ全ての細胞 が、SA-β-gal 陽性の老化細胞であった。5-ブロモデオ キシウリジンの取り込み試験により、SA-β-gal 陽性細 胞は細胞増殖能を大きく喪失していることを確認し た。

老化細胞では、酸化ストレスが高まっていることが 知られている。そこで、細胞内 Reactive Oxygen Species (ROS)に特異的な蛍光プローブ DCFDA で染色する と、炭素線によって誘導された老化細胞においても細 胞内 ROS の増大が観察された。細胞内で ROS の高い 部位は核とその周囲に認められた。興味深いことに、 核周囲の ROS の高い領域とライソゾームの部位が一 致し、老化細胞における高い ROS 産生にライソゾーム が関与していることが示唆された。 炭素線照射だけでなく、X線照射によっても NP-2 細胞に老化誘導が起きるかどうかについては、 SA-β-galをはじめとする特徴的な老化形質の誘導が検 出できた。

がん抑制遺伝子、p53 の応答を次に検討した。NP-2 細胞では、未照射においても高いp53 タンパク質の発 現量を示し、照射後もその量に変化は認められなかっ た。しかしながら、p53 の 15 番目のセリン残基のリン 酸化は照射後特異的に上昇が検出できたことから、 NP-2細胞においても炭素線照射によるp53のリン酸化 とういう質的変化による活性化が生じていることが 推測された。実際、p53 によって誘導されることが知 られている Cyclin-dependent kinase (CDK) 阻害因子 p21 タンパク質の発現誘導を確認できた。これら、p53 活性化から p21 誘導に至る一連のシグナル伝達が重粒 子線を照射した NP-2 細胞における老化誘導、そして 細胞死誘導に重要な働きをしていることが示唆され た。

3-41 MicroRNA Expression in Response to Heavy Ion Beam Irradiation

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The influence of the heavy ion beam irradiation on the expression of microRNAs was analyzed. C2C12 myoblasts were irradiated with broadbeam (20 Ne⁷⁺: 260 MeV, 2 Gy) and the level of microRNA expression was quantified at 1 hour after irradiation by microRNA array. Compared to the non-irradiated control, some microRNAs were increased, and some were decreased in the expression level after irradiation (Fig. 1). Using real-time PCR technique, the time course of expression levels of No. 9 and 11 of Figure 1 after irradiation were analysed. The expression levels of both No. 9 and 11 reached maximum at 1hour after irradiation and then gradually decreased.

我々はこれまでに細胞の重粒子線照射によるストレ ス応答を電子顕微鏡レベルの構造変化¹⁾、オートファ ジーの亢進、および遺伝子発現レベルの変化の観点か ら解析を行ってきた。

microRNA は、近年注目を集めている非翻訳型 RNA の一種で、成熟型はおよそ 20 塩基ほどの短鎖の一本鎖 RNA であることから、この様に呼ばれている。ヒトで はゲノム上に約 1,000 個の配列が存在し、現在約 500 個が成熟型として確認されている。microRNA はメッ センジャーRNA に相補的に結合することでメッセン ジャーRNA の翻訳過程を阻害し、タンパク質発現を負 に制御していると考えられている。microRNA の機能 については細胞増殖、分化、発生、などの生理的現象 や、さらには癌をはじめとする種々の疾患に関与する ことが明らかにされてきたが、各々の microRNA がど のメッセンジャーRNA をターゲットとしているかな ど詳細なメカニズムについては未解明の部分も多い。

今回我々は、重粒子線照射時の遺伝子発現応答を microRNA に着目し解析を行った。マウス由来の培養 筋芽細胞 C2C12 を用い、重粒子線照射 (²⁰Ne⁷⁺: 260 MeV, 2 Gy) 後 1 時間 静置 した 細 胞 と 非 照 射 の 細 胞 の microRNA 発現量をプローブ数約 600 個のアレイ解析で 網羅的に解析した。

重粒子線照射の前後で発現が誘導されるもの、変化が 見られたもの、発現が抑制されたものが見られた (Fig. 1)。これらの microRNA のうち図中で9,11と記 した重粒子線照射により顕著に発現が誘導されるもの についてさらに検討をおこなった。

リアルタイム PCR を用いた解析の結果、9,11の発現 量は照射後1時間でコントロールの約2倍に達し、その 後緩やかに発現量が減少することがわかった。

今後、9,11 に対するアンチセンスを導入した細胞を 用い照射後の生存率を調べることで9,11 の放射線応答 時の役割を明らかにしたい。

Reference

1) M. Hino et al., Cell Struct. Funct. 32 (2007) 51.





3-42 Biological Effects of Carbon Ions and X-ray on the Postnatal Cerebellum in Organotypic Slice Cultures

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Though the usefulness of carbon ions (12 C) therapy has been reported recently, little has been known about effects of 12 C on the normal brain tissue, especially at the developing stage. Hence, in this study, we investigated the effects of 12 C and X-ray on the early postnatal cerebellum using a modified organotypic slice culture system. In the 12 C-irradiated slices, the hematoxylin and eosin (HE) staining showed that the granule cells (GC) of the external granule layer (EGL) spread over the molecular layer and the zone of the EGL was extended. The GC in the irradiated slices at 5 days after 12 C-irradiation still existed in the EGL, whereas almost all of the GC in the non-irradiated slices migrated toward internal granule layer. TUNEL methods showed that most of the GC in the EGL was densely stained at 24 h after 12 C-irradiation. In addition, the processes of Bergmann glia (BG) retracted in the 12 C-irradiated slices. These changes were similar to those occurred by X-irradiation. The abnormality of EGL caused by irradiation was as a result of the loss of migration ability of the GC and the disorganization of BG processes. The relative biological effectiveness of 12 C-particles to X-ray was 1.4~1.6 in the EGL.

【目的】小児の脳腫瘍に対する放射線治療は、高次脳 機能に晩期有害事象を生じる可能性が成人に比べ高い ことが知られている。また、小児の脳腫瘍は小脳に発 生することが比較的多く、小児の小脳における放射線、 特に今後適応拡大が期待される重粒子線の影響を詳細 に検討することは重要な課題である。しかしながら、 重粒子線照射が小児の小脳に及ぼす影響については、 現在までにほとんど研究されていない。そこで、脳切 片培養標本を用いて未熟な小脳組織における重粒子線 およびX線の生物学的効果について検討した。

【方法】生後10日目の未熟なラットから小脳を取り出 し、約600 µmの切片培養標本を作製した。切片培養 標本を培養後1日目にTIARAにて炭素線(18.3 MeV/amu, 108 keV/µm, 0~20 Gy)、群馬大学にてX線 (140 kVp, 4.5 mA, 0~20 Gy)を照射した。照射後の切片 を経時的に固定し、照射前後での組織変化および細胞 変化について検討した。

【結果】炭素線照射後の標本をHE染色し、組織変化 について調べた結果、炭素線を照射したそれぞれの標 本において外顆粒細胞層の異常が認められた (Fig. 1)。 外顆粒細胞は通常、小脳皮質の内側へと遊走し、内顆 粒細胞層を形成することが知られていることから、 我々は、炭素線照射後の外顆粒細胞の遊走能について 検討した。その結果、小脳切片における外顆粒細胞は、 未照射切片では5日後に遊走がほぼ完了し外顆粒細胞 層が消失したにもかかわらず、照射切片については外 顆粒細胞層がほとんど残っていた (Fig. 2)。この結果 から、炭素線照射切片における外顆粒細胞の遊走は停 止もしくは遅延していると考えられた。

次に外顆粒細胞の細胞死についてTUNEL法を用い て検討した。炭素線照射3時間後の組織において、外 顆粒細胞層のもっとも外側の細胞のみがTUNEL陽性 像を示した。その後TUNEL陽性細胞数は増加し、照 射24時間後には外顆粒細胞層のほとんどの細胞が陽性 を示した。また、外顆粒細胞の遊走を支持しているベ ルクマングリア細胞の形態についてGFAP抗体を用い て調べた。その結果、未照射切片ではベルクマングリ ア細胞の突起が小脳皮質の先端まで真直ぐに伸びてい たのに対し、照射切片においては突起が萎縮し、小脳 皮質の先端まで形成されていなかった。

以上の結果は、X線照射実験においても同様の結果 が得られ、外顆粒細胞層の異常および外顆粒細胞の細 胞死を指標としたRBEは1.4~1.6であった。

【考察】照射による外顆粒細胞層の異常は、ベルクマ ングリア細胞の突起形態異常および外顆粒細胞の遊走 能の低下・停止の結果、誘発されると考えられた。



Fig. 1 Images of HE stain for tissue that cultivated 12 h after irradiation. These image shows slice following non-irradiation (a), ¹²C-irradiation of 10 Gy (b) and X-irradiation of 10 Gy (c).



Fig. 2 Images of HE stain for tissue that cultivated 5 days after irradiation. These image shows slice following non-irradiation (a) and ¹²C-irradiation of 10 Gy (b).

3-43 Bystander Mutagenic Effect in Normal Human Cells Induced by Carbon-ion Microbeams

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In the field of fundamental biological studies for high-LET radiations, there are many reports regarding bystander cellular effects after exposure to alpha particles derived from ²³⁸Pu or helium-ion microbeams. However, only limited sets of studies have examined bystander effects after exposure to different ion species heavier than helium, such as carbon ions¹⁾. In this study we have been investigating bystander cellular responses in both normal human cells and human tumor cell lines irradiated with different ion species heavier than helium, such as carbon, neon and argon ions. Last year, we have begun to study bystander cellular effects, such as cell-killing effect, in normal human skin fibroblasts irradiated with carbon-ion microbeams generated with the HZ1 port. This year, expanding to this work, we have examined other cellular effect, such as mutation induction, using the same experimental condition with the last year's study.

Carbon-ion microbeams (12C5+, 220 MeV) were generated with the HZ1 port. Approximately 6×10^5 exponentially growing cells were inoculated into each of microbeam dish, which was made of acrylic resin ring with 36 mm diameter and attached 7.5 um-thick polyimide film on the bottom of the ring, 2 days before irradiation. One day before microbeam irradiation, half of the sample dishes were treated with a specific inhibitor of gap-junction mediated cell-cell communication (40 µM of y-isomer of hexachloro-cyclohexane). At the irradiation period, cultures were confluent. Cell cycle distribution of the confluent cultures was analyzed using a flow cytometory and around 95% of the cells were G1- or G0-phase. Irradiation was carried out using a 256 (16×16) -cross-stripe method described in the last year's report. The value of linear energy transfer (LET) was estimated to be 103 keV/µm at the sample position. Microbeams of 20 µm in diameter were irradiated in each point with 8 delivered ions.

Figure 1 showed the results of mutation induction at *hprt* locus mapped on the X chromosome, which was detected with 6-thioguanine resistant clones, in microbeam-irradiated dishes (IR) and microbeam-irradiated dishes with a specific inhibitor of gap-junction mediated cell-cell communication (L+IR). The mutation frequency in cells irradiated with carbon-ion microbeams was 6 times higher than those of non-irradiated control cells. In our 256-cross-stripe irradiation method, we estimated that the percent of carbon-ion direct hit cells was around 0.2% of all cells in the dish. We evaluated that the mutation frequency in the microbeam-irradiated cell population detected with our

256-cross-stripe irradiation method was the same level with non-irradiated control population, based on the data using carbon-ion broad beams (LET=80 keV/ μ m, 0.4 Gy). The result showed a higher mutation induction in the microbeam-irradiated cell population than that of the expected frequency of mutation induction. There is clear evidence that carbon-ion microbeam irradiated cells have enabled to induce mutation by a bystander cellular response in neighboring cells, which are not directly hit by carbon ions in the same dish, as well as cell-killing effect.





Furthermore, the mutation frequency of carbon-ion microbeam irradiated cells with a specific inhibitor of gap-junction mediated cell-cell communication (L+IR) was almost the same level with non-irradiated cells only treated with a specific inhibitor of gap-junction mediated cell-cell communication (L+Control). Our studies provide clear evidence that a possible mechanism of bystander cellular effects observed in both cell-killing effect and mutation induction is gap-junction mediated cell-cell communication. Now we have begun to examine bystander cellular effects using human tumor cell lines, which are not active of gap-junction mediated cell-cell communication, comparing to the normal human skin fibroblasts.

Reference

1) N. Hamada et al., Mutat. Res., 639 (2008) 35.

3-44 Analysis of Molecular Mechanisms for Radiation-Induced Bystander Effects Using Heavy Ion Microbeams

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The objective of this project is to elucidate molecular mechanisms of the bystander response using heavy ion microbeams in JAEA. We found that the foci of γ H2A.X and pNBS1 were formed in the unirradiated cells in the target colony including the irradiated cell 6 h after irradiation with 460 MeV ⁴⁰Ar beams and that this formation of the foci was almost completely suppressed by the addition of DMSO or Lindane. Also we found that the foci of γ H2A.X and pNBS1 were formed in the unirradiated cells in the untargeted colonies 6 h after irradiation and that this formation of the foci was almost completely suppressed by the addition of aminoguanidine or c-PTIO. Our findings strongly suggested that ROS and NO are initiators/mediators for evoking heavy ion microbeam-induced bystander responses.

低線量/低線量率放射線に対して生物が示す特異的 な応答様式の一つに放射線誘発バイスタンダー効果が ある¹⁾。我々は、日本原子力研究開発機構におい て開発された重イオンマイクロビーム細胞照射シス テム(細胞局部照射装置)を用いて、この放射線誘発 バイスタンダー効果の分子メカニズムを明らか にすることを計画した。

1. 実験方法

- (1)細胞:ヒト正常線維芽細胞(AG1522 細胞)を用いた。
- (2)培養:開孔(径 12 mm)35 mm ディッシュの内面中央に20 mm四方のCR-39をパラフィンで固定したものを使用した。またCR-39の表面をcollagenコートして細胞を培養した。2×10⁵cells/mlの細胞懸濁液5µlをCR-39樹脂上に5箇所スポットし(1,000 cells/colony)、15~20時間培養したものを照射実験に供した。
- (3)照射: Funayama ら²⁾の方法に従って、中央に スポットしたコロニーの細胞 1 個に 5 粒子の 460 MeV⁴⁰Ar¹³⁺を HZ1 ポートにおいて照射した。 また対照実験として 460 MeV⁴⁰Ar¹³⁺をHY1 ポー トにおいても照射した。
- (4)細胞の蛍光免疫染色:照射後、細胞を 37°C で 培養し、0.5 および 6 時間後にメタノールで固 定し、リン酸化型 H2AX (γH2AX) 抗体および リン酸化型 NBS1 (pNBS1) 抗体を用いた蛍光 抗体染色法により染色し、蛍光顕微鏡下で観察 した³⁾。標的細胞を含む正方形枠(250×250 μm; C) 内、そこから右方向に設定した 3 個の正方 形枠(250×250 μm; X、Y および Z) の内部、 および遠隔に存在するサテライトコロニーの 中心部の正方形枠(250×250 μm; S) 内のフォ ーカス形成頻度を測定した。

2. 結果および考察

(1)標的細胞でのγH2AX および pNBS1 のフォーカス 形成:Ar線マイクロビームの照射により、照射され た細胞において γH2AX および pNBS1 のフォーカ ス形成が認められ、それらは同所局在していた。

- (2)バイスタンダー細胞における γH2AX および pNBS1 のフォーカス形成: Ar 線マイクロビーム 照射により、バイスタンダー細胞においても γH2AX および pNBS1 のフォーカス形成が認められ、それ らは同所局在していた。従って標的細胞から分泌さ れた可溶性低分子物質(バイスタンダー因子)によ りバイスタンダー細胞に DNA 二本鎖切断が誘発さ れていることが示唆された。またバイスタンダー細 胞におけるこれらのフォーカス形成は標的細胞から 離れるに従ってその頻度は減少した。
- (3)バイスタンダー因子の種類に依存したバイスタンダ 一効果の発現: Ar 線マイクロビーム照射により、バ イスタンダー細胞における γH2AX および pNBS1 のフォーカス形成が Dimethylsulfoxide (DMSO, 0.1%)、 Lindane (40 μM)、Aminoguanidine (AG, 10 μM) お よび carboxy-PTIO (c-PTIO, 10 μM) によって抑制さ れた。DMSO は ROS の消去剤、Lindane は細胞間の ギャップ結合の阻害剤、AG は nitric oxide (NO)ラジ カル合成酵素の阻害剤、c-PTIO は NO ラジカルの消 去剤である。DMSO による抑制は、C、X および S において顕著であり、それ以外の部位では抑制効果 は認められなかった。Lindane による抑制効果は C においてのみ顕著であった。AG および c-PTIO によ る抑制効果は、Z および S において顕著に認められ た。

以上の結果より、ギャップ結合を移行しているバイ スタンダー因子および標的細胞近隣の細胞に培地を介 して作用しているバイスタンダー因子は活性酸素種 (ROS)であることが示唆された。一方、遠隔的に作 用しているバイスタンダー因子は活性窒素種(RNS)、 特に NO ラジカルであることが示唆された。

- 1) H. Matsumoto et al., J. Radiat. Res. 48 (2007) 97.
- 2) T. Funayama et al., Radiat. Res. 163 (2005) 241.
- 3) A. Takahashi et al., Cancer Res. 64 (2004) 8839.

3-45 Ion Beam Irradiation Has Different Influences on Morphology of the Cultured Ciliary Body Exposed to L-dopa between ¹²C and ⁴He

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Deficiency of L-dopa causes degeneration of the substantia nigra in the brain and L-dopa is used in the treatment of Parkinson's disease. We proved that L-dopa produced NO and superoxide, and had the cytotoxic effects on the retinal pigment epithelial cells¹⁾. L-dopa injected into the vitreous of the rats dilated the vena in the ciliary body²⁾. The gamma ray (60 Co) causes ionization uniformly in the whole irradiated tissue. We found out that high dose of gamma irradiation prevented programmed cell death regulated by p53 and bcl-2 genes in the ciliary body dissected from living body, and was useful for preservation of organ after culture by the protective influence of inflammatory reaction³⁾.

The ion beam is an ionization ray that is induced by acceleration of the ionizing atom of 12 C and 4 He, 1 H so on, but has a characteristic to lose energy and stop at the constant depth of tissue. The reaching depth to the tissue depends on the ion species and the acceleration energy. The reaching depth in water is 1.08 mm in 220 MeV 12 C, and 1.64 mm in 50 MeV 4 He.

If ion beam can selectively irradiate NPE in the ciliary body that causes hyper-secretion of aqueous humor, we might be able to cure ocular hypertension and glaucoma instead of surgery. Gamma ray penetrates into the tissue, but an ion beam has the strongest energy and a maximum influence at a point inside the tissue. Moreover, the ion beam is supposed to have a bystander effect around the irradiated tissue. We investigate which part of the ciliary body the ion beam influences, and we compared the effects among the species of the ion. As for as we know, this is the first report concerned with morphological changes in ciliary body cultured after the ion beam irradiation.

The anterior segments from porcine eyes consisting of the ciliary body, iris, and cornea incubated with L-dopa (250 μ M) for 2 hrs were exposed to an ionization radiation that is induced by acceleration of the ionizing atom of 220 MeV ¹²C and 50 MeV ⁴He. The anterior segments were fixed in a combined fixative of 5% formalin and 2.5% glutaraldehyde after 4, and 8h of incubation, embedded in paraffin, and cut into 3 μ m thick sections using a microtome. After deparaffinizing the sections with xylene and ethanol, the specimens were stained with hematoxylin and eosin, dehydrated in a graded series of ethanol and xylene solutions, and mounted. For studying the bcl-2 and p53 expression, formalin -fixed and paraffin -embedded specimens were immunohistochemically stained with a mouse monoclonal anti-human bcl-2 or p53 antibody.

We reported that ion beams with longer projectile range such as ⁴H damaged NPE of the ciliary processes and folds strongly⁴). It might be possible to apply to glaucoma therapy as microsurgery technique using the ion beams with longer projectile range. The genes related to apoptosis in the NPE of the ciliary processes were easy to be influenced by the exposure to the ion beam with shorter projectile range such as ²⁰Ne. However, the ion beams (⁴He) whose projectile range is long, had the strong effects on the NPE of the ciliary folds. These ion beams increased the products by Bax gene the most among Bcl-2, Bax, and p53.

In these experiments, the incubation with L-dopa improved the damage of NPE in the organ cultured ciliary bodies, decreased the products by expression of p53. The morphology of the ciliary body was preserved by 220 MeV ¹²C and 50 MeV ⁴He, that inhibited the products by bax genes. The products by p53 genes was induced by 50 MeV ⁴He, and inhibited by 220 MeV ¹²C. In the ciliary bodies exposed to L-dopa, the products by p53 genes were increased by 220 MeV ¹²C, and localized in the NPE at the tips of the ciliary pcocesses. The effects on programmed cell death regulated by p53 and bcl-2 genes was related to the reaching depth in water as well as the different accumulation of the energy irradiated at a point by various ions.

- 1) K. Akeo et al., Pigment Cell Res. 13 (2000) 80.
- 2) S.A. Amaki et al., Pigment Cell Res. 14 (2001) 256.
- 3) K. Akeo et al., Exp. Animals. 55 (2006) 375.
- 4) K. Akeo et al., ARVO Annual Meeting 2005.

3-46 Analysis of Lethal Effect Induced by Ion Beams in Canine Spontaneous Tumor Cells

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The dose per fraction of heavy ion therapy may become lower than that of X ray therapy. In this study we investigated lethal effect induced by low dose that considered important on heavy ion radiotherapy. Cell lines used for this experiment were melanoma cell from spontaneously generated canine tumor. These cells were irradiated 220 MeV ${}^{12}C^{5+}$ ions (108 keV/µm) or γ rays. Low dose survival rates by carbon ion irradiation were less than extrapolated values from high dose irradiation. This result indicates that the lethal effect by low dose carbon ion irradiation is higher than estimated lethal effect. And it was considered that low dose higher lethal effect of carbon ion irradiation caused production of radicals after low dose carbon irradiation by method of radical staining and treatment of radical inhibitor. In this study, it was suggested that application of carbon ion radiotherapy to malignant melanoma therapy would lead high therapeutic gain.

1. はじめに

本研究では、今後期待される獣医領域における粒 子線治療の基礎研究として、いまだ報告の少ない粒子 線に対する腫瘍細胞の感受性を評価した。また、粒子 線は高い細胞致死効果が得られるため1回照射線量は 従来のX線治療に用いられる1回照射線量よりも低くな ることが推察される。さらに、これまでは低線量域で の細胞応答は高線量照射による細胞応答からの外挿で 見積もられてきたが、現在、低線量域での細胞応答は 高線量域照射による細胞応答の外挿では説明できない 現象が報告されている。そこで、粒子線治療の普及に つれて無視できなくなるであろう低線量照射による基 礎データを得ることも併せて目的とした。

2. 実験方法

悪性黒色腫を用い、 γ 線およびAVFサイクロトロ ンによって加速された220 MeV ¹²C⁵⁺(LET= 108 keV/µm)を照射した。細胞致死効果の評価には colony formation assayを用いた。照射後に産生される OHラジカルを測定するため、細胞内Reactive Oxygen Species (ROS) に特異的な蛍光プローブCM-H₂DCF-DA で染色し、蛍光強度をマイクロプレートリーダで測定 した。さらに、OHラジカルの細胞致死効果への関与 を検証するため、ラジカル捕捉剤のDMSO処理による 生存率の算出を行った。

3. 結果および考察

これまでにX線やy線の低線量照射における生存率 は高線量域の生存曲線からの外挿値とほぼ一致してい たが、炭素線の低線量域での生存率は、高線量域の生 存曲線からの外挿値よりも低い値を示され、炭素線で は低線量域でも高い致死効果が得られることを明らか にした。この低線量域での炭素線の細胞致死効果の増 強の要因としてOHラジカルの関与を明らかにするた め、まず、低線量炭素線照射後のラジカル産生を解析 した。y線および炭素線0.4 Gy照射後に産生されるラ ジカルをコントロールに対する比で表した(Fig. 1)。低 線量過剰致死効果の観察されないy線照射では細胞内 ラジカルはコントロールとほぼ同一であったが、過剰 致死効果が認められた炭素線照射では細胞内ラジカル はコントロールよりも高くなることが観察された。さ らに、この低線量炭素線照射によって産生されるラジ カルが細胞致死効果に関与するかを検証するため、 OHラジカルの捕捉剤であるDMSO処理を行った。炭 素線照射後にDMSO添加培地で培養し、生存率を算出 したとき(Fig. 2)、DMSO処理における低線量炭素線照 射の生存率はDMSO無処理時の生存率よりも有意に高 くなることが観察された。この結果は低線量炭素線照 射による過剰細胞致死効果には照射後に細胞内で産生 されるOHラジカルが関与することを示唆している。 これらのことから炭素線の悪性黒色腫治療への適用に より高い治療効果が期待されると考えられた。









3-47 DNA Repair after High LET Irradiation of the Anhydrobiotic Insect *Polypedilum vanderplanki*

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Polypedilum vanderplanki is the only insect possessing ability to resist almost complete dehydration in its larval stage and revive within an hour after placing back to water. In the previous studies, we showed that dried (dry) larvae show higher radiation tolerance compared with active (wet) ones^{1,2)}. Such improvement of radiation tolerance in anhydrobiotic larvae, to some extent, can be due to the presence of excess of sugar trehalose, which could provide structural and biochemical protection of the cells in anhydrobiotic state³⁾. Our recent results suggested that, however, there are more important factors other than protectant⁴⁾. Therefore, we focused on DNA-related aspects of irradiation-response of the larvae, and analyzed DNA breaks and expression of genes involved in general stress-response cascades and DNA reparation, after irradiation. We used the doses of ion beams which were previously found to be ID₅₀ (median inhibitory dose) for adult emergence (70 Gy) and pupation (160 Gy) of the chironomid ^{1,2)}. The larvae were exposed to 50 MeV ⁴He (LET_{∞}=16.2 keV/ μ m) ion beam delivered from the azimuthally-varying-field cyclotron at TIARA. DNA status and repair were analyzed with alkaline Comet Assay standard technique using the fat body cells as a model cell type.

Estimation of relative percentage of DNA in comet "tails", as a quantitative marker of DNA breaks, showed that the larvae experienced severe DNA damage after irradiation with doses both set of doses (70 Gy and 160 Gy) (Fig. 1, inset panel). However, we have not found any evidence of massive cell death and/or increasing level of apoptotic cells in irradiated larvae. The reparation of DNA to the level comparable to one of non-irradiated larvae took at least 96 h (Fig. 1). Moreover, non-irradiated dry larvae after revival also possessed cells with severely damaged DNA, which were also repaired within 96 h after rehydration (Fig. 1).

The damaged DNA after ⁴He irradiation and desiccation stress should be mended by repair enzymes and other stress-response associated proteins. Indeed, we cloned complete cDNA of a number of stress-related genes of *P. vanderplanki* (ROS-response, chaperons, apoptosis-related, DNA-repair involved) and analyze their expression after irradiation and desiccation stresses. Thus far, at least one gene (UV excision repair protein Rad23, family InterPro: IPR004806) was found to be increased expression level significantly in response to both radiation and desiccation stresses (Fig. 2 A, B). On the bases of the results, we assume that higher tolerance of dry larvae to different types of radiation could be a result of synthesis of stress-related proteins in the larvae prior to entering the anhydrobiosis. The upregulation of these genes could not prevent DNA damage completely upon desiccation, but might give certain advantages for the recovery of the revived larvae.

References

- 1) M. Watanabe et al., Int. J. Radiat. Biol. 82 (2006) 587.
- 2) M. Watanabe et al., Int. J. Radiat. Biol. 82 (2006) 835.
- M. Sakurai et al., Proc. Natl. Acad. Sci. USA. 105 (2008) 5093.
- 4) M. Watanabe et al., J. Insect Physiol. 53 (2007) 573.



Fig. 1 Dynamic of DNA reparation in the larvae of *P*. *vanderplanki* exposed to ⁴He ion beam (0-160 Gy) and naturally reviving after anhydrobiosis with no irradiation (Rehydration). Inset panel shows a comet of a cell from irradiated larva (70 Gy, 24 h after). a: intact DNA. b: broken DNA. Damaged DNA (%) = b/(a+b)x100.



Fig. 2 Relative expression of Rad23 in the larvae of *P*. *vanderplanki* during desiccating (Desi) and following rehydration (Rehy), and in non-desiccated larvae after irradiation with 70 Gy of ⁴He (Irrad).

3-48 Regeneration Mechanism of Heavy-ion Irradiated Hemopoietic Organ of the Silkworm, *Bombyx mori* –Expression Analysis of BeK after Irradiation–

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In *Bombyx mori*, hemopoietic organs are located near or attached to the imaginal wing discs at thoracic region, and produce and release hemocytes into hemolymph. However, the detailed mechanism of hemopoiesis in the organs has not been fully understood until now.

Our previous studies elucidated that the hemopoietic organ disrupted by selectively localized heavy-ion irradiation could regenerate at wandering stage¹⁻²⁾. Moreover, developmental conditions in larva, including hormonal condition, are considered to influence the timing for degradation and regeneration of the irradiated organ. These observations seem quite interesting to investigate the mechanism of hemopoiesis in *B. mori*.

Last year, we reported that the two protein markers would be involved in degradation process of the organs. Those proteins were estimated as soluble alkaline phosphatase and eIF2 alfa kinase, by peptide-mass fingerprinting. In this report, molecular cloning and expression analysis of *Bombyx* eIF2 alfa kinase (BeK) gene were performed.

First, a PCR primer pair was synthesized based on the DNA sequence of BeK gene. RT-PCR was performed using the primer pair and total RNA extracted from testis of 5th instar day 3 larvae. One specific PCR product was obtained (Fig. 1) and sequenced. The sequence of the product was identical to that of BeK gene.

Next, we examined the real-time RT-PCR analysis using different tissues (fat body, midgut, testis, silk gland, epidermal tissue) from 5th instar day 3 larvae. The results showed that the expression of BeK gene was detected ubiquitously, and expressed more prominently in testis and midgut (data was not shown).



Fig. 1 Cloning of BeK gene by RT-PCR using specific primer for BeK. Lane1: 250bp marker, Lane 2: Product of RT-PCR without RT reaction, Lane 3: Product of RT-PCR. Arrow indicates BeK gene.

Next, we investigated the effects of irradiation on the expression level of BeK gene. The hemopoietic organs only at left side were selectively irradiated with carbon ion beams (100 Gy). Then the irradiated organs were dissected every 24 hours. Right hemopoietic organs were used as the control. The expression level of BeK of each sample was examined by RT-PCR. As shown in Fig. 2, BeK mRNA of the irradiated organs from 5th instar day 1 larvae was clearly higher than that of the control organ. As the putative BeK protein increased at 5th instar day 2, this result seems reasonable. However, the expression of BeK was also observed at the other stages. Especially, BeK expressions were detected in all samples from the irradiated organs. Therefore, further studies are necessary to conclude the participation of BeK in the degradation process of the irradiated organs.

Now, we are searching for new molecular markers which would be involved in regeneration process of the irradiated organs.

References

- 1) Ling et al., J. Insect. Biothecnol. Sericol. 72 (2003) 95.
- 2) Ling et al., Develop. Comp. Immunol. 30 (2006) 531.



Fig. 2 Developmental changes of BeK expression on the product of RT-PCR using specific primer for BeK.

Lane 1:250bp marker. Lane 2, 4, 6 & 8 are samples from irradiated hemopoietic organs, and lane 3, 5, 7 & 9 from control organs. Lane 2&3 are samples from 5th instar day 0, lane 4&5 from day1, lane 6&7 from day 2, and lane 8&9 from day 3 larvae, respectively. Arrow indicates BeK gene.

3-49 Effect of Locally Targeted Heavy-ion Microbeam Irradiation on Root hydrotropism in Arabidopsis

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Roots respond to a number of environmental cues such as gravity, light, and moisture gradients with gravitropism, phototropism and hydrotropism, respectively. Root hydrotropism is most likely to play an important role in the acquisition of water.¹⁾ Studies using seedling roots of pea, cucumber, and maize have shown that de-tipped roots of pea and maize seedlings display a remarkable reduction in hydrotropism, and that an auxin-responsive gene from cucumber, CsIAA1, are asymmetrically expressed in the elongation zone during the root hydrotropic response of cucumber.^{2,3)} More recently, we established an experimental system of hydrotropism using Arabidopsis seedling roots, and showed that the requirement of root cap cells as well as auxin response in root hydrotropism also seemed to be true for Arabidopsis,^{4,5)} however, there has been no confirmation that columella cells or elongation zone are necessary for hydrotropism. In this study, we investigated the role of root cap cells and elongation zone on root hydrotropism by positional irradiation of heavy-ion and laser microbeam. In addition, we examined the auxin-responsiveness by monitoring the induction of de novo IAA5 expression after systemic irradiation of heavy-ion broad-beam.

First, to investigate the role of columella cells and elongation zone in the root hydrotropic response of Arabidopsis seedlings, columella root cap and elongation zone proximal to the root tip were locally irradiated with 500 Gy of 180-µm-diamter carbon-ion microbeam. Control Arabidopsis seedling roots responded to a water potential gradient and developed orthotropic curvature. In contrast, when cells of the elongation zone proximal to the root tip were exposed to carbon-ion microbeam irradiation, the hydrotropic response was significantly repressed, particularly in the early phase.⁶⁾ The overall root growth during the corresponding period was less affected by irradiation. Unexpectedly, localized carbon-ion irradiation of the root cap cells did not affect the development of hydrotropic curvature or root growth. Because this result was somewhat contradictory to our previous results that suggested the necessity of root cap cells for the hydrotropic response,⁴⁾ we further investigated the functional role of the columella cells by laser ablation. Laser ablation of stories 1 and 2 (the first and second proximal cell layers, respectively) of the columella cells severely repressed the development of hydrotropic curvature for as long as 12 h post-irradiation, indicating that columella cells play an important role in the early phase of the hydrotropic response.⁶⁾ Laser ablation of root cap cells did not affect the overall root growth during this period.

We further examined changes in auxin-responsive gene expression in the presence of exogenously applied auxin to determine whether systemic heavy-ion irradiation with broad-beam inactivates irradiated cells, so as to confirm the effectiveness of heavy-ions on suppression of de novo gene expression. We chose IAA5 gene as a probe, for its expression is highly sensitive to exogenously applied auxin among the IAA genes.⁷⁾ In the following experiments, we used seedlings that did not receive irradiation (cold-run seedlings) as a control, to take into account possible effects of the experimental procedure on gene expression levels and hydrotropism. When control Arabidopsis seedlings were treated with 1 µM IAA for 1 h, there was a 43.3-fold increase in the mRNA of the auxin-responsive gene IAA5 compared to mock-treated cultures. In contrast, systemic irradiation of Arabidopsis seedlings with 500 Gy of carbon-ion broad-beam caused a 8.3-fold increase in the level of IAA5 mRNA, which was significantly low when compared to control cultures.⁶⁾ We also monitored MIZ1 gene whose product is responsible for root hydrotropism in Arabidopsis.⁸⁾ In contrast to the IAA5, 500 Gy of carbon beam irradiation caused neither increase nor decrease in mRNA level.⁶⁾ Considering that MIZ1 is MIZ1 predominantly expressed in columella cells, and that MIZ1 is not an auxin-responsive gene, our present result suggests that the decrease in root hydrotropic curvature by heavy-ion beam irradiation is not due to the decrease of MIZ1 mRNA level, rather de novo expressions of auxin-responsive IAA genes at elongation zone might be involved. However, our present data do not completely explain the necessity of de novo gene expression at the elongation zone during root hydrotropism, and thus efforts should be made to verify gene expression profiles of several genes to explain the necessity of *de novo* gene expression in root bending in the future

In summary, our present results indicate that both the root cap and elongation zone have indispensable and functionally distinct roles in root hydrotropism, and that *de novo* gene expression might be required for hydrotropism in the elongation zone, but not in columella cells.

- 1) Takahashi, J. Plant Res. 110 (1997) 163-169.
- 2) Takano et al., Planta 197 (1995) 410-413.
- 3) Mizuno et al., Plant Cell Physiol. 43 (2002) 793-801.
- 4) Takahashi et al., Plant Physiol. 132 (2003) 805-810.
- 5) Kaneyasu et al., J. Exp. Bot. 58 (2007) 1143-1150.
- 6) Miyazawa et al. J. Radiat. Res. doi:10.1269/jrr.07131.
- 7) Abel et al., J. Mol. Biol. 251 (1995) 533-549.
- Kobayashi et al., Proc. Natl. Acad. Sci. U.S.A. 104 (2007) 4724-4729.

3-50 Characterization of *C. elegans F49F1.6* Gene Coding for a Secreted Surface Protein in Response to Ionizing Radiation

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In this study, to clarify the effects of ionizing radiation on individuals at the molecular level, we monitored global transcriptional alterations in the nematode Caenorhabditis elegans, which is easy to handle and is genetically well characterized, and whose development and life cycle are well understood. In addition, approximately 40% of the C. elegans genes show good homology to mammalian genes, and transcriptional profiles of individual cells or tissues at different developmental stages as determined by DNA microarray analysis have been reported¹⁾. We therefore compared the transcriptional profiles of nematodes at adult stage that were exposed to 100 Gy of whole-body (TIARA HY1 port of AVF cyclotoron) with ¹²C⁵⁺ ions, or 100 Gy of Co-60 γ -ray irradiation using the entire-genome C. elegans GeneChip array. We compared the relative signal strengths estimated from single chips corresponding to 4 h after 100 Gy ¹²C⁵⁺ ions broad-beam irradiation / control for 4 h. In all (signal detection P value: P < 0.05), 106 transcripts were significantly (2-fold or greater) upregulated and 104 transcripts were significantly downregulated (0.5-fold or less) 4 h after IR exposure. Combining the data for both $^{12}C^{5+}$ ions broad-beam and γ -ray irradiation showed that 38 genes were significantly (2-fold or greater) upregulated under both conditions. The cadmium responsive and glutathione S-transferase-like genes cdr-2 and -4, some C-type lectin genes, and the secreted surface protein genes F01D5.1 and F49F1.6, were specifically induced by IR. Especially, we observed the largest alteration of the F49F1.6 gene expression.



To confirm the microarray expression results, we performed real-time RT-PCR to quantitatively measure the expression levels of F49F1.6 using the translation elongation factor 2 gene *eft-2* as an internal standard against which to normalize these PCR data. The real-time PCR results confirmed the results of the DNA microarray analyses, and the expression levels were increased by 18-fold and 30-fold in L1 larvae and adult hermaphrodites, respectively (Fig. 1). In addition, 4 h after irradiation of 100 Gy of body-surface at 10 μ m depth with ${}^{12}C^{5+}$ ions (TIARA 3MV tandem accelerator), the expression level in L1 larvae was increased by 2-fold (Fig. 1). Its upregulation was transiently, and the expression became steady-state level 24 h after IR irradiation².

F49F1.6 gene product has a signal peptide and Metridin-like ShK toxin domain and shows good similarity to the N-terminal region of mammalian Mucin-2 precursor. It has been reported that the gene expression is upregulated by infection of *Psudomonas aeruginosa* and the regulation is required for p38 MAPK pathway³). In addition, our preliminary analysis indicates that the gene expression increases in the defect of mitochondrial DNA replication. These results suggest that F49F1.6 gene may be involved in immune response genes controlled by pathogen response, oxidative damage and ionizing radiation. We are now attempting to study biological function and transcriptional regulation of F49F1.6 gene.

- S. J. McKay et al., Cold Spring Harb. Symp. Quant. Biol. 68 (2003) 159.
- 2) T. Kimura et al., Bioelectromagnetics (2008) in press.
- 3) E.R. Troemel et al., PLoS Genet. 2 (2006) e183.

3-51 Quality Control in Positron Emitting Tracer Imaging System (PETIS) Examination for Quantification Analysis of Plant Physiological Functions

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The positron emitting tracer imaging system (PETIS) is the most promising approach for *in vivo* plant molecular imaging that facilitate an understanding of the dynamics of water, nutrients and pollutant in plants. Furthermore, the PETIS and its quantitative tracer images have the potential to analyze the plant physiology of a test plant. In order to acquire the analyzable dynamic data of PETIS images quantitatively, a scheduled work for constant quality controls is absolutely essential.

We have performed experiments of quarterly maintenance of the uniformity and sensitivity correction of this system. The system uniformity and sensitivity on the focal plane of field of view (FOV) were measured using a flat uniform phantom¹⁾ in an environmental control chamber. The flat phantom has inner dimensions of 3 mm thickness, 210 mm width and 330 mm height, contains a radioactive solution and is made of three acrylic plates (each 3 mm thick). Figure 1 shows an example of the focal plane image measured by the flat phantom filled with ¹⁸F solution, where the image is corrected for the density of coincidence lines back-projected on the FOV. The phantom image on the left in Fig. 1 has a sharper pattern that is acquired at high activity, on the other hand the image on the right at low activity has more uniformity at the same time noisier. These patterns depend on the target activity on the FOV. This

crosshatching pattern on the image indicates that the peripheral regions of detector unit generated the low sensitivity area. As a result of the activity dependence, we made a modification to the PETIS software in the sensitivity correction, and we have recommend the following algorism; in some measurements for the plant study having very low-activity tracer, it may be preferable to correct the sensitivity with low-activity phantom data.

At the same time, the quantitative performance of PETIS has been checked with a time activity curve (TAC) of the ¹⁸F phantom images. Figure 2 shows an example of a TAC (black dot) generated with the dynamic phantom data without physical decay correction and an ideal TAC (extrapolated straight line) estimated with the physical decay of ¹⁸F (109.8 min). The two TACs represented the PETIS linearity was kept for approximately 10 times of the half-life, that is the quantitatively dynamic range of PETIS had between from 20 kBq to 20 MBq.

These works of the maintenance for constant quality controls support the many other plant physiological experiment of PETIS studies and guarantee quantitative kinetic analysis.

Reference

1) H. Uchida et al., Nucl. Instr. Meth. A 516 (2004) 564.



Fig. 1 Phantom images at the high activity (left) and at low activity (right). The bottom plot profiles indicate its sharpness and uniformity.





3-52 Imaging of Translocation of ⁶²Zn from the Roots to the Shoots in Zn/Cd Hyperaccumulator *Thlaspi caerulescens* by PETIS

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The heavy metal hyperaccumulation phenotype seen in Thlaspi caerulescens is the result of physiological differences at the cellular, organ and whole plant levels relative to non-hyperaccumulator plant species. These differences lead to the accumulation of zinc (Zn) and cadmium (Cd) as high as 40,000 ppm Zn and 10,000 ppm Cd dry weight in shoot tissues. Most non-hyperaccumulator plant species only accumulate 100-300 ppm Zn and 0.1-10 ppm Cd foliar concentrations. These differences in Zn and Cd content relative to non-hyperaccumulators is due, in part, to enhanced metal transport at a number of different sites within the plant, including the root surface, xylem loading and reabsorption of xylem-born metals by leaf cells. These differences are also seen at the molecular level with higher expression of metal transporter genes in both roots and shoots of T. caerulescens relative to T. arvense, a closely related non-hyperaccumulating plant species. For example, expression of the Zn/Cd transporter, ZNT1, is much greater in roots and shoots of T. caerulescens than in T. arvense and down-regulation of ZNT1 expression with increased plant Zn status requires accumulation of much higher levels of Zn in T. caerulescens plants. As a result, one of the characteristics for metal hyperaccumulators is the enhanced translocation of Zn and Cd from the roots to the shoots.

In the present study, the translocation of 62 Zn from the roots to the shoots in *T. caerulescens* (hyperaccumulator) relative to *T. arvense* (non-hyperaccumulator) was examined by the positron-emitting tracer imaging system (PETIS).

T. caerulescens and *T. arvense* were hydroponically cultured in a growth chamber. Plants were supplied with 20 mL of culture solution in polyethylene bags. The plants and the bags were fixed on an acrylic board and placed between a pair of PETIS detectors in a chamber. ⁶²Zn (about 25 MBq) were added to culture solution. The plants were monitored by PETIS for 300 min.

The serial images of translocation of 62 Zn from the roots to the shoots were obtained (Fig. 1). And time-activity curves for translocation of 62 Zn in the basal shoot and the root region were shown in Fig. 2.

The slopes of time-activity curves from 200 to 300 min. in the root and the basal shoot regions, implying the flow rate of 62 Zn in the region, were higher in *T. caerulescens* than those in *T. arvense*, although the transpiration rate in *T. caerulescens* (0.5 mL h⁻¹) was lower than that in *T. arvense* (0.75 mL h⁻¹). It was indicated that the translocation of Zn from the root to the shoots of *T. caerulescens*, that is a Zn/Cd-hyperaccumulator, would be enhanced.



300 min.

Fig. 1 The serial images of translocation of 62 Zn from the roots to the shoots in *T. caerulescens* and *T. arvense*.



Fig. 2 The time-activity curves in the root and the basal shoot region in *T. caerulescens* and *T. arvense*.

3-53 Noninvasive Analysis of Photoassimilate Translocation in Tomato Leaves under the Dark Condition by Using ¹¹CO₂ and a Positron-Emitting Tracer Imaging System

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Recently, Kawachi et al. $(2006)^{11}$ established a method for the kinetic analysis of ${}^{11}CO_2$ for studying photoassimilate transport in a leaf, using PETIS. The method quantitatively analyzes the photoassimilate transport under various light conditions. However, it is important to analyze photoassimilate transport under both dark and light conditions because leaves transport carbon under both these conditions (Mullen et al., 1988)²). In the present study, we developed a new method to analyze photoassimilate translocation, using PETIS, and ${}^{11}CO_2$. This novel method clarified the mechanism underlying photoassimilate translocation from a leaf under the dark condition by noninvasive, repeated imaging experiments, using the same specimen.

Tomato plants were placed in the growth chamber equipped with PETIS. ¹¹CO₂ was fed to the fifth matured leaf for 5 min under artificial irradiance of 150 μ mol·m⁻²·s⁻¹. Figure 1 illustrates the study protocol. The same plant was fed with ¹¹C and monitored 3–4 times consecutively. For the first to third feeding experiments, γ -ray correction was conducted under the dark condition, except in 1 case of temporal light condition. In contrast, the fourth feeding was conducted under an irradiance of 500 μ mol·m⁻²·s⁻¹. The leaf blades were intensely labeled with absorbed ¹¹CO₂. To analyze the kinetics of the ¹¹C-labeled photoassimilate, 15 regions of interest (ROIs) were manually drawn on the integrated ¹¹C PETIS images around the leaf segments of 3 plants.

The equation for calculating the amount of ¹¹C activity remaining in the fed leaf is as follows.



Fig. 1 Time schedule followed for the preparation and feeding of ¹¹C.

In the above equation, C(t) is the amount of ¹¹C activity remaining in the leaf, C_0 is a constant value, τ shows the proportion of decline per hour as a percentage (b \times 100%), and *t* is the time after monitoring was initiated. In the above equation, to quantities carbon kinetics the export time constant *t* is determined to the best possible extent by means of least-square fitting.

The calculations provided below are for the 4 sequential feeds supplied to Plant 1 at ROI 1.

 $C(1) = 9.45 \times 10^{3} \times \exp(-5.41 \times 10^{-3}...t)...$ first feeding. $C(2) = 8.57 \times 10^{3} \times \exp(-5.49 \times 10^{-3}...t)...$ second feeding. $C(3) = 8.80 \times 10^{3} \times \exp(5.24 \times 10^{-3}...t)...$ third feeding. $C(4) = 17.7 \times 10^{4} \times \exp(3.17 \times 10^{-3}...t)...$ fourth feeding.

Time constants of photoassimilate export for all ROIs are summarized in Table 1.

The technique presented in this paper involves the use of PETIS for measuring carbon translocation in plants by examining the principal C fluxes in the leaf continuum of intact plants with fine temporal resolution.

These results indicate that plants possess potentially high carbon translocation activity during night.

The proposed methods may be used to elucidate the mechanism and relationship between leaf carbon fixation and translocation under various conditions.

- N. Kawachi et al. IEEE Transact. Nucl. Sci (2006) 53: 2991-2997.
- 2) Mullen et al. Plant Physiol. (1988) 880-884.

Table 1 Estimated all time constant of photoassimilate exporting of all ROIs.

		1 st feeding	2 nd feeding	3 rd feeding	4 th feeding
Plant No.	ROI	2 h dark	5 h dark	8 h dark	light
Plant 1	ROI 1-1	50.4	51.9	48.6	24.4
	ROI 1-2	53.9	60.1	57.7	27.7
	ROI 1-3	56	57	51.5	32.7
	ROI 1-4	53.2	59.1	55.1	28
	ROI 1-5	56.6	58.6	50.9	29.3
	ROI 2-1	54.1	54.9	52.4	31.7
	ROI 2-2	53.6	50	48.3	33.7
plant2	ROI 2-3	43	38.1	38.8	31.8
	ROI 2-4	55.1	55.8	54.4	30.2
	ROI 2-5	62.2	59.8	56.7	44.5
	ROI 3-1	41.8	45.4	45.2	n.a
	ROI 3-2	29.2	32.3	32.1	n.a
Plant 3	ROI 3-3	31.1	36.9	36.8	n.a
	ROI 3-4	37.6	44.2	40	n.a
	ROI 3-5	28.6	34.1	30.5	n.a

3-54 Visualization of Photoassimilate Translocation in Eggplant Fruit Using ¹¹CO₂ and a Positron-Emitting Tracer Imaging System

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To clarify the mechanism of the growth and development of fruits such as eggplant and tomato in relation to plant growth and yield, examination of the import of dry matter into fruits from other organs such as leaves is essential. Positron-emitting tracer imaging system (PETIS) noninvasively visualizes movement of ¹¹C-labeled photoassimilate in plants. So we fed ¹¹CO₂ to a leaf and monitored the translocation of ¹¹C-labeled photoassimilate into the eggplant fruit by PETIS.

PETIS can obtain two-dimensional serial images of the distribution of tracers. Thin organ such as leaf and stalk is suitable for localization analysis by PETIS. But it is difficult to analyze the three dimensional fruit by PETIS. Then, we placed fruit by acrylic board at the early stage of the fruits development. As a result, we succeeded in the creation of fruits whose thickness is about 1.5 cm. The photoassimilate translocation inside fruit was visualized by using this thin fruit. The seventh leaf of the eggplant was covered with a gas cell and ¹¹CO₂ gas and ambient air were introduced into the cell and maintained at 200 mL/min. PETIS images were acquired every 10 s for 180 min.

Figure 1 gives examples of the test plants (a) and serial PETIS images (b) every 60 min after ¹¹CO₂ feeding; these images were generated from integration of the corresponding images collected every 10 s. [¹¹C]photoassimilates began to be translocated to the fruit via the leaf petiole about 60 min after feeding (Fig. 1b). Furthermore, we successfully observed the movement of

^{[11}C]photoassimilates inside the fruit. ¹¹C signal intensity in the fruit increased gradually, and its distribution was not uniform. We observed a similar translocation pattern in eggplant. Although a number of experiments have reported the translocation of photoassimilates from source leaf to sink fruit, the translocation of photoassimilates within the fruit has never been reported. We examined photographs of sliced eggplant fruit after PETIS measurement (Fig. 2a), together with BAS images (Fig. 2b). Traces of translocated ^{[11}C]photoassimilates inside the fruit were clearly shown on the BAS image. The images showed that the translocation of ^{[11}C]photoassimilates was localized. Similar PETIS and BAS images were obtained in other independent experiments using other plants (data not shown). In the tomato, it is suggested that phyllotaxis and the arrangement of the vascular system were related to the distribution of photoassimilates. Data indicating photoassimilate localization inside the eggplant fruit clearly demonstrated that photoassimilate translocation was related to phyllotaxis and the arrangement of the vascular system.

Knowledge of the distribution pattern of photoassimilates inside the fruit is important for our understanding of the morphogenesis, development, and maturation of fruit. The production of malformed fruits, such as those that are puffy or curved or that show defective development, is a very serious problem that reduces fruit yield and quality. Thus, PETIS may be a powerful tool for revealing the mechanisms of fruit malformation in future research.



Fig. 1 A plant photograph in the experiment. ${}^{11}CO_2$ received leaf was set outside the imaging region but the petiole was inside (a). Serial PETIS images of translocation of [${}^{11}C$]photoassimilates (b).



Fig. 2 Photographs (a) and BAS (bio-imaging analyzer system) images (b) of sliced eggplant fruit after PETIS measurement.

3-55 The Production of ¹³N-labeled Nitrogen Gas for Imaging of Nitrogen Fixation by Soybean Nodule

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The nodule is a symbiotic organ of leguminous plants with rhizobium. Soybean plants utilize nitrogen (N) fixed by nodules from atmospheric N₂. Until now, ¹⁵N, a stable isotope, has long been used for kinetics of N₂ fixation and dynamics of transport of fixed N. However, because this method is invasive, it has been difficult to analyze an instant response to environmental (eg. temperature, light conditions) changes with these methods because they are invasive.

We are planning to image and analyze such kinetics and dynamics quantitatively and noninvasively by using nitrogen gas labeled with ¹³N (half life: 10 min), a positron emitting isotope, and PETIS (positron imaging tracer imaging system). In this paper, we report the synthesis and purification of ¹³N-labeled nitrogen gas ($^{13}N_2$).

Soybean plants, *Glycine max* [L.] cv. Williams and En1282 (non-nodulated isogenic line) were inoculated with rhizobium (*Bradyrizobium japonicum* strain USDA 110) and were hydroponically cultivated with an N-free medium. The plants were used for the experiment 30 days after sowing.

 $^{13}N_2$ was produced with the $^{16}O(p, \alpha)$ ^{13}N reaction by bombarding CO2 containing 10% He with proton beam delivered from the TIARA AVF cyclotron. We collected the irradiated gas pressured by Ar gas into a cylinder (Fig. 1, without the tube filled with soda lime). A part of the collected gas was analyzed by Gas Chromatography (G.C.). We found four major peaks in the chromatogram and identified three of them as Ar, ¹³N₂, CO₂ by using standard gas analysis (Figs. 2A and 2B), but the fourth peak (indicated as X) has not been identified yet (Fig. 2 B). It is known that more than 10% CO2 in atmosphere inhibits N2 fixation¹⁾, so we tried to remove CO₂ from the irradiated gas. A stainless tube filled with granular soda lime was connected before the collection cylinder. The irradiated gas pressured by Ar gas was collected through this tube into the cylinder (Fig. 1). As a result, the second peak was disappeared. In other words, ¹³N₂ without CO₂ was obtained (Fig. 2 C and 2D).

We fed the ${}^{13}N_2$ to Williams and En1282 and visualized ${}^{13}N$ distribution in the roots and nodules. ${}^{13}N_2$ was mixed with ambient air and fed to the plants for 10 min. After ${}^{13}N_2$ was washed out with fresh air, the roots and nodules were contacted to imaging plates for the Bio-imaging Analyzer System (FUJIFILM, Tokyo, Japan) for 30 min. We obtained ${}^{13}N$ signal in both the nodules and roots near the nodules as shown in Figs. 3A and 3B. However, even in non-nodulated

soybean (En1282), approximately 50% signal compared with that of Williams was detected in the roots (Figs. 3C and 3D). We think the signal could be derived from the component X. Therefore, now we are trying to isolate and collect ${}^{13}N_2$ using G.C. from the irradiated gas.

Reference

1) N. Ohtake et al., TIARA Annual Report 2002 (2003) 114.



Fig. 1 Scheme of irradiation, collection and analysis of gas.



Fig. 2 Gas chromatogram of the collected gas.



Fig. 3 Scanner and BAS images of Williams (A and B) and En1282 (C and D).

3-56 Imaging of ¹⁰⁷Cd Translocation in Oilseed Rape Plants Treated with Different Cd Concentrations

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Cadmium (Cd) is one of toxic heavy metal element. Cd accumulation in the human body causes serious health problems when Cd is taken via the farm products. In order to decrease Cd accumulation in these products, it is necessary to elucidate mechanisms of Cd long-distance transport in the plant body. However, these mechanisms are not fully understood so far. In this research, we tried to elucidate mechanisms of Cd long-distance transport by visualizing Cd transport in the plant body using positron emitting tracer imaging system (PETIS).

¹⁰⁷Cd is used as a positron-emitter in the PETIS experiment. 107Cd was produced according to the method established by Ishioka et al.¹⁾. Oilseed rape plants (Brassica napus L.) were grown hydroponically for two weeks after sowing. We selected this plant for our experiments because it is expected to be Cd tolerant and a Cd accumulator. PETIS experiments were performed in the growth chamber where the growth conditions of plants were controlled. After setting plants in the chamber, PETIS experiments were started by adding purified ¹⁰⁷Cd in hydroponic solutions. In this study, different concentrations of Cd (0.1 μ M and 10 μ M) were added as a carrier to hydroponic solutions. 0.1 µM is comparable to the Cd concentration in the soil solution from non Cd-polluted soils²⁾. 10 μ M is comparable to the toxic Cd concentration for oilseed rape plants (unpublished data). Time-series images of the ¹⁰⁷Cd distribution were obtained with the PETIS apparatus.

We succeeded to obtain fine serial images of Cd transport in oilseed rape plants (Fig. 1). Strong ¹⁰⁷Cd signals were seen in the basal region of the shoot. We also could see strong signals of ¹⁰⁷Cd in the node. In these experiments, the pattern of Cd accumulation in the oilseed rape plants was similar in the different Cd concentrations (0.1 μ M and 10 μ M). These results indicate that, at least in the young stages, Cd concentration in the hydroponic solution don't have effects on Cd absorption in the root systems.

- N.S. Ishioka et al., JAEA Takasaki Ann. Rep. 2005 (2006) 162.
- 2) G.J. Wagner, Adv. Agron. (1993) 51, 173-212.



Fig. 1 Accumulation of ¹⁰⁷Cd signals in oilseed rape plants treated with Cd of 0.1 μ M and 10 μ M. Each image is integration of 90 original frames, corresponding to 6 hour.

3-57 Cyclotron Production of PET Radionuclide: No-carrier-added Bromine-76 with Protons on ⁷⁶Se-enriched Cu₂Se Target

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In recent years, Positron Emission Tomography (PET) has become established as particularly attractive tracer for cancer detection using positron-emitter. The increasing use of PET for clinical diagnosis, drug development and biological research has prompted many chemists to develop new labeling and purification methods. The PET radionuclides such as ¹¹C, ¹³N, ¹⁵O and ¹⁸F have been used for imaging agent, which are short half-lives; ¹⁸F with the longest half - life is 110 minutes. On the other hand, the positron-emitter with a longer half-life that can be applied to the PET diagnosis based on a nuclear characteristic is proposed besides above-mentioned nuclides. Bromine-76 is of potential interest in nuclear medicine: ⁷⁶Br has been suggested as a halogen label with a convenient half-life ($T_{1/2}$ = 16.1 h) and a sufficient β -branching ratio (57.2%) in PET. Here we present the development of a production method for no-carrier-added ⁷⁶Br with protons on ⁷⁶Se-enriched Cu₂Se target.

 $^{76}\mathrm{Br}$ was produced at the JAEA-TIARA AVF cyclotron. Cu₂ $^{76}\mathrm{Se}$ disk target, which is prepared by mixing $^{76}\mathrm{Se}$ metal powder enriched up to 99.67% with Cu metal powder was bombarded at 5 $\mu\mathrm{A}$ to make $^{76}\mathrm{Br}$ via the (p, n) reaction. The $^{76}\mathrm{Br}$ is removed from the irradiated target by dry distillation and from the bulk of selenium traces by thermal chromatography (Fig. 1). The target was placed in a platinum ship inside a quartz tube, and positioned in the

 $\begin{array}{c} \leftarrow 1000^{\circ}\text{C} \longrightarrow 200^{\circ}\text{C} \longrightarrow \\ (30 \text{ ml/min}) & (7^{6}\text{Br}\uparrow, \text{Se}\uparrow) & (\text{Se}\downarrow) \\ \hline \\ Cu_2\text{Se} & \downarrow \\ Platinum & {}^{65}\text{Zn}, {}^{71.72.74}\text{As}, {}^{75}\text{Se}, {}^{77}\text{Br} & (7^{6}\text{Br}\downarrow) \\ \end{array}$

Fig. 1 Schematic view of the ⁷⁶Br separation equipment.

middle of a furnace. In an adjacent auxiliary furnace (30 cm), a polytetrafluoroethylene (PTFE) tube (I.D. 2 mm) was inserted into the quartz tube. The end of the PTFE tube was immersed into a H₂O (15 mL). Under a constant stream of argon gas (30 mL/min) through the tubes, the temperature in the furnace containing the target pellet and the second furnace were raised respectively to 1030 °C, 200 °C.

Separation yields were typically 20-50% and the separation time of 2-3 hours. Conditioning of the oven (heating up to the working temperature) took most of the separation time. When at operation temperature there was a rapid release of radioactivity from the target. The production yield in the present target was found to be about 20 MBq/ μ Ah (EOB). In the final solution, we could no detect except for the ⁷⁶Br.

To evaluate the efficiency of the thermochromatographic separation, the deposition pattern of bromide and selenium was studied by utilizing ⁷⁷Br and ⁷⁵Se, formed via the Se(p,xn)⁷⁷Br and ⁷⁶Se(p,pn)⁷⁵Se reactions after irradiating a Cu₂Se target of natural isotopic composition. After the end of separation, the PTFE tube was cut in steps of 10 mm, and measured for ⁷⁷Br and ⁷⁵Se radioactivity using an HPGe detector (Fig. 2). It was shown that the ⁷⁷Br was deposited with the ⁷⁵Se. For increasing the separation yield, we have to find the condition of extraction of only radiobromine from the PTFE tube.



Fig. 2 Separation of selenium and bromine by thermal chromatography, showing the deposition areas of ⁷⁵Se and ⁷⁷Br along the PTFE tube. The radioactivity was normalized to maximum values.

3-58 Production of Radioisotopes for Nuclear Medicine Using Ion-beam Technology and Its Utilization for Both Therapeutic and Diagnostic Application in Cancer

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Optical imaging is recently developed for in vivo molecular imaging. It requires only a simple system, and has advantage of relatively low cost. Furthermore, optical imaging probes can provide the highest signal-to-noise ratio for molecular targeting, so optical imaging is an ideal candidate for molecular imaging. In this study, we prepared a probe, which labeled with both radioisotope and fluorescent dye, evaluated the difference of images from PET and optical imaging, and compared the characteristics of both imaging methods. From our results, multimodal imaging system can provide complementary information about the functional status of various tissues, and can improve the accuracy of tumor diagnosis.

臨床画像診断に用いられる代表的な手法としてCT、 MRI、PET・SPECTやUSなどがあげられるが、近年こ れらに加えてOptical Imagingへの関心が高まっている。 このOptical Imagingは最近になってインビボでの利用 が実用化された技術であり、PET、MRIのような大が かりな設備を必要とせず、比較的安価である利点を持 つ。さらに標的分子との相互作用によりシグナルを出 す分子型に変化する設計が可能であり、投与直後から 高い標的/非標的比が得られる可能性を有する。ただ し光は組織透過性が悪く、体内部のイメージングには 限界があるため、単純な臨床応用は困難と予想される。

一方で最近PET-CT、SPECT-CTなどが開発され、臨 床画像診断の新たな戦力として成果を上げおり、複数 の画像診断法を組み合わせることで臨床診断に有効な 情報をもたらすことが期待できる。そこで我々はPET と Optical Imaging を組み合わせたマルチモダリティ によるがん診断の有用性を明らかにするため、一つの 分子内に放射性核種(RI)標識部位と蛍光標識部位を 有するがん指向性probeを作製し、各々の画像を直接 比較することで両者から得られる情報の同一性、画像 の特徴を明らかにすることを計画した。

RIおよび蛍光色素で標識するprobeにはCD20を抗原 とする抗体:NuB2を用い、モデル動物としてヒトの び慢性大細胞性リンパ腫であるRamos細胞を移植した BALB/cヌードマウスを用意した。またPET用RIとし て⁶⁴Cuを用いることとし、抗体に⁶⁴Cuを結合させるた めのキレート部位として1,4,8,11-tetraazacyclotetradecan -N,N',N'',N'''-tetraacetic acid (TETA)を導入した。蛍光 色素には組織透過性を考慮してSAIVITM Alexa Fluor[®] 750 (Invitrogen Corp., Carlsbad, CA, USA)を利用し、 probe投与24時間後にPETおよび蛍光撮像を行うとと もに、臓器を摘出し、重量および放射能を測定した。 PETの撮影はSET-2400W (Shimadzu Corp., Japan)を用 いて行った。またOptical ImagingにはMaestroTM (CRi, USA)を利用した。

摘出した臓器の放射能と蛍光色素を検出・比較し た実験では、一部明らかに一致しない組織(血液、肝





臓)があったが、比較的一致した結果が得られた (Fig. 1)。一方イメージングにおいては組織透過性 の高いNIRを利用してもOptical Imagingでは体表面付 近しか検出できず、体内深部まで検出可能なPET画像 とは異なる画像を得る結果となった。摘出臓器への放 射能の集積結果と比較して蛍光の画像は必ずしも正確 な体内分布を反映しているとは言えないが、皮下に移 植した腫瘍の評価には有用であると思われた¹⁾²⁾。

今回の結果はPETとOptical Imagingのマルチモダリ ティによる画像診断の有効性を示しており、RI-蛍光 二重標識probeを用いたがん診断の可能性を示唆する。

- Y. Iida et al., "A Direct Comparison between In Vivo Optical Imaging and PET Imaging in Mice", Proc. AMI/SMI Joint Molecular Imaging Conference 2007, Providence, Rhode Island, USA.
- 2) 飯田靖彦、「がん診断におけるPETの有用性と新しいPET用放射性核種の利用」、群馬大学 生体調節研究所シンポジウム、前橋(2007).

3-59 Production Yield of Endohedral ¹³³Xe-Fullerene at Incident Energy of 5 keV by Ion Implantation

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Endohedral fullerenes encapsulating a radioisotope within a fullerene cage have a possibility of being new radiopharmaceuticals. We have prepared endohedral ¹³³Xe-fullerenes by ion implantation¹⁾. Xenon-133 ($T_{1/2}$ = 5.243 d) would be useful for cancer therapy because it emits 0.346-MeV β^- rays (maximum range in water: 1.0 mm) followed by 81-keV y rays and conversion electrons associated with the γ transition. We have studied the production yields of endohedral ¹³³Xe-fullerene at incident energies of 15 and 30 keV by ion implantation. The production yield at the incident energy of 15 keV was about 20% larger than that of 30 keV. In that experiment, implantation of ¹³³Xe⁺ ions into the targets was carried out with an isotope separator at acceleration energy of 30 keV. For retardation from 30 keV to 15 keV, high voltage of 15 kV was supplied from a retardation power supply to the fullerene targets mounted on a retardation system (Fig. 1). We wished to study the production yields of endohedral ¹³³Xe-fullerene at lower incident energies. However we could not supply more than 15kV because of electric discharge on the power terminal of the retardation system. In the present paper, the production yield of endohedral ¹³³Xe-fullerene at incident energy of 5 keV is investigated using a new retardation system that can supply high voltage of 25 kV without electric discharge.

Fullerene targets for ion implantation were made by vacuum evaporation of C_{60} on Ni foils. Implantation of $^{133}Xe^+$ ions into the targets was carried out with an isotope separator at acceleration energy of 30 keV. In the implantation, a new retardation system with a cover of the power terminal (Fig. 2) was used to avoid electric discharge. For retardation of incident energy, the high voltage of 25 kV was supplied to fullerene targets mounted on the new retardation system: incident energy became 5 keV. Doses were in the rage of 1×10^{12} to 1×10^{14} cm⁻². After ion implantation, the fullerene part on the target was dissolved

in *o*-dichlorobenzene. The solution was filtered through a membrane filter to remove insoluble materials. The ¹³³Xe radioactivity in the filtrate was measured by γ -ray spectrometry to determine the radioactivity of ¹³³Xe@C₆₀. In order to evaluate the efficiency of the formation of ¹³³Xe@C₆₀, the yield of ¹³³Xe@C₆₀ was defined as a percentage of the radioactivity of ¹³³Xe@C₆₀ to that of ¹³³Xe implanted into the target.

The yields of ¹³³Xe@C₆₀ were plotted as a function of ¹³³Xe dose in Fig. 3, together with the yields at incident energy of 15 keV. The solid and broken straight lines in this figure, which were fitted by the method of least-squares on log-log scales, are $y = (10.12\pm0.45) - (0.825\pm0.035)x$ and $y = (10.18\pm0.09) - (0.842\pm0.007)x$ on 5 and 15 keV, respectively. For both dose studied, the yields of ¹³³Xe@C₆₀ were found to decrease with increasing dose. Because an increase of the dose leads to an increase of the number of damaged fullerene molecules as reported by Kastner *et al.*²⁾ from their Raman spectra measurements of fullerene targets bombarded with energetic-ions, those dose dependences of the production yields can be ascribed to amorphization of ¹³³Xe@C₆₀.

From comparison between both lines one can note that the yield of 5 keV is about 50% larger than that of 15 keV for the same dose in the rage of 1×10^{12} to 1×10^{14} cm⁻². This energy dependence of the production yield can be also ascribed to the amorphization of ¹³³Xe@C₆₀. The yield of the ¹³³Xe@C₆₀ should be more increased by decreasing incident energy. We will study the yield at lower incident energy.

- S. Watanabe et al., J. Radioanal. Nucl. Chem., 255 (2003) 495.
- 2) J. Kastner et al., Appl. Phys. Lett. 65 (1994) 543.



Fig. 3 The yield of ¹³³Xe@C₆₀ as a function of ¹³³Xe dose.



3-60 **Effect of Different Immersion Solutions on Fluorine** Uptake into Enamel around Fluoride-containing Materials during pH-cycling

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Introduction

It is well established that the fluorine is a possible agent for caries prevention. The fluoride that contained some restorative material was also demonstrated as a great benefit for preventing dental caries. The purpose of this study was to determine the effect of immersion solutions on fluorine (F) uptake into enamel around fluoride-containing materials during pH-cycling¹⁾ using proton induced gamma-ray emission (PIGE) technique²⁾

Material and Methods

Class I cavities in the buccal surfaces of 7 extracted human teeth were drilled and filled with fluoride-containing materials; Fuji IX (FN) and Beautifil II with Fluoro Bond Shake One (BE). Four 300 µm sections through the material were obtained from each tooth. All specimens were polished to a thickness of 120 µm. All tooth surfaces, except the outer surface of enamel, were coated with a wax. In order to simulate daily acid challenges occurring in the oral cavity, the pH-cycling (pH 7.0-pH 4.5) was carried out for 5 weeks¹⁾. Two pairs of demineralizing and remineralizing solutions were prepared for the immersion solution of pH-cycling (Table 1). After pH-Cycling, F distribution of the outer lesion in each specimen was evaluated using PIGE technique²⁾. The amount of F in the outer 150 μ m of the lesion was then calculated. Two-way ANOVA was used for the analysis (p<0.05).

Results

F distribution of the specimens after 5-week pH-cycling showed in Fig. 1. For both FN and BE, the calculated amount of F had significantly higher value comparing to those before pH-cycling. There was no significant difference in the F amount between the immersion solutions, while there was significant difference between the materials. **Discussion:**

In a pH of 5.5-4.5, hydroxyapatite (main enamel component) dissolves, and by supplying fluorine from the material, precipitation of fluorapatite (FA) occurs because of the continuing supersaturation of FA. At the same time, adequate amounts of calcium (Ca) and phosphate (P) were necessary for precipitation of FA. The concentration of Ca and P in solution A which had been used in previous studies were at an equal or higher degree compared to those in other pH-cycling reports. The current study carried out to

compare different version³⁾ (i.e., solution B) of the pH-cycling solution. There was no significant difference between the immersion solutions. Our previous reports on the ability of fluoride in materials would therefore be compared with other reports. Moreover, the difference in the F uptake between the materials was due to supplying the amount of fluorine from each material, when the pH was in the low range.

Table 1 Composition of de- and remineralizing solutions.

Solutions	Demineralizing	Remineralaizing	
	(pH 4.5)	(pH 7.0)	
	0.2 M Lactic acid,	0.02 M HEPES, 3.0	
А	3.0 mM CaCl ₂ , 1.8	mM CaCl ₂ , 1.8 mM	
	mM KH ₂ PO ₄ .	KH ₂ PO ₄	
В	0.05 M Acetic acid,	0.02 M HEPES, 1.5	
	1.5 mM CaCl ₂ , 0.9	mM CaCl ₂ , 0.9 M	
	mM KH ₂ PO ₄	KH ₂ PO ₄ , 0.13 M KCl	



Fig. 1 Fluorine uptake into the outer lesion after during pH- cycling.

Conclusion

There was no effect of F uptake by the difference of immersion solutions used in this study during pH-cycling. Among fluoride-containing materials, there existed some difference in F supply when the pH was in the low range.

- 1) Y. Matsuda et al., Dent. Mater. J. 24 (2006) 280.
- 2) H. Yamamoto et al., Nucl. Instr. and Meth. B 210 (2003) 388.
- 3) J.M. ten Cate et al., Eur. J. Oral Sci. 103 (1995) 362.

3-61 Innovation of Microcapsules That Release Their Core Contents through Radiation

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In the first symposium of Takasaki's particle research and its application, we reported the innovation of microcapsules for anticancer drug targeting through radiotherapy sessions¹⁾. These microcapsules release their core contents with response to clinical radiotherapy, and they are prepared by polymerizing alginate and hyaluronic acid by Ca-Y polymerization^{1),2)}. However, microcapsule's releasing efficiency (64.2 \pm 4.3%) was lower than that expected. In this study, we improved microcapsule's efficiency through polymerizing alginate and hyaluronic acid by Ca²⁺ and Fe²⁺.

The mixture of 0.2% alginate (weight/volume) and 0.1% (weight/volume) was mixed in agate pestle. To this mixture, 0.2 mmol carboplatin (a platinum (Pt)-containing anticancer drug) was added. A droplet of this mixture was sonicated using an ultrasound disintegrator and atomized to yield a solution of CaCl₂ that was supplemented with FeCl₂ at concentrations ranging from 0 to 100%. The polymerization was completed within 5 min to yield microcapsules. The microcapsules thus generated were purified using a Nalgene disposable filter kit (8-0301-84 DP591) and resuspeneded in 0.1 mmol Tris hydroxymethyl aminomethane (THAM) buffer. Resulting microcapsules were irradiated with each doses of 0.5, 1.0, 1.5, 2 or 2.5 Gy 60 Co γ -radiation. Then a frequency of microcapsule's rupturing were tested by micro PIXE camera.

The generated microcapsules were $20.3 \pm 3.8 \ \mu m\Phi$ in size, with a liquid core of $19.7 \pm 1.2 \ \mu m\Phi$. The distribution of the microcapsular core contents (carboplatin) was imaged using a micro PIXE camera that detected platinum (Pt) signals (Fig. 1 A–H). The contours of the microcapsules were identified by detecting the distribution of Ca and ion beam-induced charge (IBIC) (Fig. 1 B, D, F, and H). Before radiation, the contours of microcapsules were smooth and their liquid cores were clearly observed. After 1.5 Gy of ⁶⁰Co γ radiation, the size of some microcapsules decreased and their liquid cores could not be detected (Fig. 1 E-H).

The frequencies of microcapsules' rupturing were shown in Fig. 2. The increment in the frequencies of microcapsule decomposition was dependent on the final concentrations of the added F (Fig. 2). When the concentration of added Fe was ranging from 40.0 to 70.0%, the rupturing of unirradiated microcapsules was less than 5 % and that of irradiated was more than 80.0 % with 2.0 or 2.5 Gy. 2.0 or 2.5 is the dose that is used in the conventional fractionated radiotherapy. Therefore, our microcapsules may have possibility to be used in combination with clinical radiotherapy. However, microcapsules must be improved for better releasing of their core contents through radiotherapy.

- 1) Harada et al., TIARA annual report 2003 (2004) 217.
- 2) Harada et al., Nucl. Instr. Meth. B 260 (1) (2007) 16.



Fig. 1 Distribution of Ca, Fe, Pt, and IBIC.



Fig. 2 Frequency of ruptured microcapsules vs. final concentration of added Fe^{2+} (%).

3-62 Investigation of Cisplatin Sensitivity in Esophageal Squamous Cancer Cell Lines Using In-Air Micro-PIXE

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The purpose of this study was to elucidate the involvement of CDDP in ESCC using in air-micro PIXE (Particle Induced X-ray Emission). Two human ESCC cell lines–TE-2 and TE-13–were examined for their response to CDDP. TE-2 cells were more highly sensitive to CDDP than TE-13 cells in MTT assay. Dynamic state of element was measured in CDDP treated cancer cells using air PIXE. Using air PIXE, we observed distribution of P and K were localized at cytoplasm, and Br was localized at nuclear site. Air PIXE may be one of the new methods to measure CDDP combined with existing methods. In our study, in cytoplasm level and nuclear level, the cisplatin was included in TE-2 higher than TE-13. However, the ratio to be present in nuclear for a cell of cisplatin did not have a difference with two cell lines. Our results suggest that cisplatin sensitivity using air PIXE is one of the methods of anticancer drug sensitivity test.

近年、様々な癌において数多くの遺伝子の研究 が行われているが、いまだに不明な点が多い。特 に元素レベルにおいて、その動態や分布はほとん ど知られていない。そこで我々は、イオンの同定 可能な大気マイクロPIXEを用いて、食道扁平上皮 癌におけるシスプラチン(Pt)の取り込みの検討を 行った。食道癌は独特な生物学的特徴を有する癌 である¹⁾。化学放射線療法が効果的な癌の一つであ り、そのresponderとnon-responderを選別することは、 個別化治療において重要な課題になっている。 我々はいままで分子生物学的アプローチによる食 道癌患者における化学放射線療法の感受性評価法 を検討し報告している2)。そこで既存の感受性評価 法にPIXEの技術を融合させることにより、さらに 精度の高い感受性評価法を開発できないかと考え た。

我々はこれまでの検討で、シスプラチン処理し た食道癌細胞株を試料として、大気マイクロPIXE を用いた測定により、細胞内プラチナ元素の検出 が可能であることを見出した。一方、感受性の相 違によるシスプラチン投与後の細胞内元素動態を 比較するため、基礎実験として食道癌細胞株を用 いてシスプラチン感受性試験(MTT assay)を行い、 高感受性株(TE-2)と低感受性株(TE-13)を見出した。 この差異につき細胞周期およびアポトーシスの面 からも、FACSやDNA fragmentation assayを行い確 認している。シスプラチンを細胞内のPt分布として 評価するには、通常感受性評価に用いられるIC50 より、かなり高濃度での投与が必要であった。た だし、この濃度での長時間投与ではほとんど全て の細胞がviabilityを失うため、高濃度短時間投与、 具体的には、マイラー膜上に細胞を生着させ、2.0 mMのシスプラチン含有メディウムで1時間培養し、 洗浄後凍結乾燥したサンプルにて、比較的短時間 の照射でPt分布の安定した検出が可能であった。ま た、シスプラチンは核内に作用することに着目し、 細胞培養の段階でBrdUを用いることで、細胞内に



Fig. 1 TE-2 was high sensitive to cisplatin compared with TE-13. In cell level and nuclear level, the cisplatin was included in TE-2 higher than TE-13. The ratio to be present in nuclear for a cell of cisplatin did not have a difference with two cell lines.

おける核の局在を確認した。大気マイクロPIXEの 実験では、カリウム(K)とリン(P)の分布より細胞そ のものの形態の確認を行った。また、シスプラチ ンを定量的に評価する為に、群馬大学の腫瘍放射 線学教室にて見出した検量線を用いて評価を行っ た³⁾。すると、高感受性株(TE-2)において細胞内の Pt濃度が低感受性株(TE-13)より有意に高いことが 判明した。さらに、核内に存在するPt濃度も高感受 性株で高い結果がえられた。また、単一細胞にお ける核内へのPtの移行割合は両細胞株で有意な差を 認めなかった (Fig. 1)。これは、感受性が核レベル ではなく、細胞膜レベルで制御されている可能性 を示唆するものであった。

- Kuwano H., et al., Ann Thorac Cardiovasc Surg. 2003 Feb; 9(1): 6-13.
- 2) Sohda M., et al., Int J Cancer, 2004. 110(6): 838-44.
- 3) Sakurai H, et al., Cancer Sci, 2008. 99(5): 901-4.

3-63 The Changes of Heavy Metal in Sertoli Cells Induced by Cadmium Exposure

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Cadmium (Cd) is an industrial and environmental pollutant that is toxic to cells and organs. Cd is also well known to induce apoptosis and necrosis in liver, edema in lungs, and hemorrhagic necrosis in testis. To clarify the mechanism of acute Cd toxicity in Sertoli cells, we have analyzed distribution of metal elements such as Cd, Fe and Zn using in-air micro-PIXE (Particle Induced X-ray Emission) in Sertoli cells. Evaluation by in-air micro-PIXE method revealed that Cd and Fe distribution was increased in Sertoli cells, and Zn distribution was decreased.

1. 緒言

カドミウム(Cd)の精巣に対する毒性に関しては組織 の壊死やアポトーシスを引き起こすことが報告されて いるがその機序に関しては不明な部分が多い。現在ま での研究においてCd投与後の精巣組織において鉄と 銅の分布が増加しメタロチオネインのセルトリ細胞に おける発現が増加しCdの毒性緩和に寄与している可 能性が示された¹⁾。

本報告に於いてはCdの毒性緩和に関与していると考 えられる精巣のセルトリ細胞の単離培養系を用いて Cd曝露後のセルトリ細胞内の金属元素分布の変動を 大気マイクロPIXE法を用いて分析をおこなった。

2. 実験方法

Wistar ラット(350-400 g)の精巣をエーテル麻酔下に て摘出後、ollagenase I、hyaluronidaseによりセルトリ 細胞を単離した。マイラーフィルム上への細胞培養法 は硫酸処理を行い、その後細胞を培養する方法が行わ れてきたが、この方法では接着力が弱い初代培養系の 細胞の培養は困難であった。本研究では接着力の弱い 細胞をマイラーフィルムに接着させる方法としてマイ ラーフィルムにCellmatrix Type I-C (新田ゼラチン株 式会社、ブタ皮由来のペプシン可溶化Type-Iコラーゲ ン)をコーティングすることにより細胞を接着させ分 析を行った。コーティングされたマイラーフィルムを 用いてセルトリ細胞の培養を行い、Cd曝露後のセル トリ細胞における金属元素分布の変動を日本原子力研 究開発機構高崎量子応用研究所イオン照射研究施設 (TIARA)の大気マイクロPIXE分析システムを用い て測定した。なお、測定条件は3MVシングルエンド 加速器より発生させた加速電圧2.9 MeVのプロトンマ イクロビームを用い90分間照射を行った。

3. 結果及び考察

マイラーフィルムへのCellmatrix Type I-C による処 理によりマイラーフィルムへ接着が困難な細胞の大気 マイクロPIXEを用いた培養細胞の分析が可能になっ た。また、Cellmatrix Type I-C による処理によるバッ クグランドの元素分布の増加も認められなかった。

大気マイクロPIXEによる分析からセルトリ細胞にCd の曝露が生じるとセルトリ細胞質においてCdの分布 が増加しセルトリ細胞の細胞質がCdの毒性のターゲットで ある可能性が示された。また、Cdの曝露によるセルトリ細胞 内のFeの分布の増加とZnの分布の減少が確認された (Fig. 1)²⁾。



Fig. 1 Heavy metal maps of Sertoli cells was analyzed by in air micro-PIXE. Sertoli cells were exposed to 10 μM Cd for 24 h (B). A. Control, B. Cd 10 μM.

- T. Kusakabe, et al, "The changes of heavy metal and metallothionein distribution in testis induced by cadmium exposure." Biometals. (2008) Feb;21(1):71-81.
- T. Kusakabe, et al, "Changes of heavy metal, metallothionein and heat shock proteins in Sertoli cells induced by cadmium exposure." Toxicology in Vitro. (2008) May 10.

3-64 In-air Micro-PIXE Analysis of Asbestos in Lung Tissue and Asbestos Fibers *in vitro*

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We demonstrated that in-air micro-PIXE analysis is useful for assessing the distribution of asbestos bodies and other metals in lung tissue section, and also for analysis of standards asbestos fibers *in vitro*. This new system could detect the spatial distribution and quantitative analysis of asbestos and other metals in lung tissue section. In-air micro-PIXE system is expected to be used in examination of occupational particles exposure in the lungs or screening of particles.

緒言

アスベストの吸入は肺線維症、肺癌及び中皮腫を引き起こすと報告されている。アスベストは、クリソタイル(白石綿)、クロシドライト(青石綿)、アモサイト(茶石綿)などの種類が工業製品として使用されてきた¹⁾。このアスベストの肺組織内における局在とアスベスト組成をIn-air micro-PIXE (Particle Induced X-ray Emission)を用いて解析するとともに、アスベスト 粉末の解析も試みた。

方法

アスベスト吸入歴があり、肺癌を合併した患者肺の 癌以外の部分と、アスベスト吸入歴がなく肺癌を合併 した患者肺の癌以外の部分の肺組織中の微量元素を、 原子力機構高崎量子応用研究所のIn-air micro-PIXE分 析装置を用いて測定し比較した。

結果

In-air micro-PIXEによる観察で、アスベスト吸入歴 のある肺組織では、アスベスト吸入歴のない肺と比較 しSi, Mg, Feの沈着が明らかに多い様子が観察された (Fig. 1)。 アスベスト吸入歴のある肺組織ではSi, Mg, Feからなるアスベスト小体が組織間隙に刺さったよう になっている様子が二次元的に観察できた。

また、アスベスト肺の中にはTiやNiを吸入している ものが観察された。アスベスト粉末の解析では、クリ ソタイル(白石綿)、クロシドライト(青石綿)、ア モサイト(茶石綿)を構成するSi, Mg, Feの相対比が 検出可能であった。

考察

In-air micro-PIXEを用いたアスベスト肺の解析で、 吸入され、肺組織に沈着したアスベストを同定できる ことがわかった。 またアスベスト以外の職業性肺疾 患における吸入沈着物の同定にも応用できる可能性が あると考えられた。



Fig. 1 Distributions of Si, Mg and Fe in normal lung and asbestos lung.

Reference

 Beckett WS. Occupational respiratory diseases. New Engl J Med (2007); 342: 406-413.

3-65 The Optimum Condition and Present Situation of the Analysis of Boron Micro-distribution in Tumor Cells Using PIGE

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We have applied micro particle induced X-ray emission (micro-PIXE) to determine inter- and intracellular distribution of boron (¹⁰B). Because the energy of PIXE from ¹⁰B is too low, we employed a method to detect gamma-ray produced by the nuclear reaction of ¹⁰B (p, γ) ⁷Be, namely particle induced gamma-ray emission (PIGE). Micro-PIGE imaging showed the distribution of boron elements in tumor tissue. We compared various conditions of sample preparation to look out optimum conditions for clear distribution images. The result shows that the tumor BSH distributions were clearly demonstrated at 250 ppm of boron concentration. The best measurement ranges were from 50×50 µm to 100×100 µm, and the measurement time was from 30 to 60 minutes to make the clearest image using PIGE.

However, intracellular micro-distribution of boron could not detect clearly in our analysis. It seems to need for the improvement of the technical methods of cell fixation and upgrading micro-PIXE analyzing system itself and so on.

悪性脳腫瘍に対する硼素中性子捕捉療法(BNCT)で はその効果は硼素元素の細胞内分布状態に大きく影響 を受けるものと考えられている。これまでにも硼素元 素の腫瘍内での分布やその動態の研究が報告されてき たが、画像的に明らかにしたものはなかった^{1,2)}。

本研究の目的は腫瘍細胞内での硼素元素の分布状態 を測定・画像化することである。そのためTIARAのマ イクロPIXE分析システムを用い、特にparticle induced γ -ray emission (PIGE)法を用いることでこれまでに硼素 $10(^{10}B)$ の画像化に成功した。今回は実験における最適 条件をさらに追求することを主目的とした。

細胞レベルでの分布状態を明らかにするIn vitro実験 として、9L gliosarcoma腫瘍細胞にBSHが集積している ことを確認後、まず9L gliosarcoma腫瘍細胞を5 µmの厚 さのポリカーボネート膜上で4日間培養する。3日目お よび4日目にBSH (BSH: sodium borocaptate, Na2B12H11SH) 100 ppm、250 ppm、1000 ppmをそれぞ れ投与し、5日目にこれらを固定した。そして出来上が った試料をマイクロPIXE分析システムで30分から60分 で、また測定範囲は25×25 µmから100×100 µmの範囲 で測定・画像化し、硼素元素の分布状態を比較した。

種々の測定結果を比較すると、BSH濃度は測定感度 や細胞の耐容能などから250 ppmが、また測定範囲は 50×50から100×100 μm範囲で測定時間は30分から60 分がもっとも鮮明な画像が得られることが確認された。

まず硼素元素測定では周辺バックグランドと比べて 測定スペクトル上小さなピークが認められ、硼素元素 (¹⁰B)が測定されていることが確認できた。Fig. 1左は9L gliosarcoma腫瘍細胞の分布状態を硫黄(S)の分布を指標 に測定画像化したもの、また右は硼素元素(¹⁰B)チャン ネルにおける測定分布図である。ここで硫黄の分布か らマスクを作成し、適当なしきい値で二値化、その後 マスクで指定された部分のPIGEスペクトルを作成し全 体のスペクトルとマスクされたスペクトルを比較する と、マスクされたスペクトルでは硼素10(¹⁰B)のピーク がよりはっきり現われることから、硫黄の分布と硼素 の分布には相関があることが確認できた。つまり Fig. 1の2つの画像を重ね合わせると腫瘍細胞分布に一致し て硼素元素(¹⁰B)分布も多く測定されていることがわか り、硼素元素(¹⁰B)は腫瘍細胞周辺に集積していること が認められた。



50 µm

Fig. 1 Distribution of tumor cells(S) and ¹⁰B.

しかしながら本来の目的である細胞レベルでの硼素 元素分布の明確な画像化は、上記最適条件の下でも困 難であった。

原因としてこれまでにも報告してきたような試料作 成、9L gliosarcoma腫瘍を固定する際の技術的・手技的 問題、特にその再現性の問題があることに加え、マイ クロPIXE分析そのものの測定限界・試料のビーム耐容 限界に起因することも考えられ、これらの改善を今後 進める予定である。

これらの問題点を改善するためには、更なる細胞固 定方法・手技の向上、またマイクロPIXE分析システム 自体のバージョンアップなどに期待するところも大き いものと思われる。

- 1) Coderre JA, et al., Radiat. Res. 149 (1998) 163-170.
- Wegden M, et al., Nucl. Instrum. and Meth. in Phys Res B 219-220 (2004) 67-71.

3-66 Measurement of Bromine in Social Isolation Mice Brain Slices by In - Air Micro-PIXE

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To examine the change of the distribution on trace metal in a cell, organs or tissues using a various kind of disease model, it might be useful to establish a new pharmaco-therapeutics. PIXE (Particle Induced X-ray Emission) is a powerful method for the analysis of trace elements. In-air micro-PIXE was developed at the Takasaki Ion Accelerators for Advanced Radiation Application (TIARA) facility in the Japan Atomic Energy Agency (JAEA). Micro-PIXE allows analyzing the spatial distribution of the elements quantitatively. Our previous studies show that we might be got useful data on distributions of trace elements in the brain slices using the in-air micro-PIXE analysis.

In this study, we analyzed a distribution change of trace metals in brain slices which were subjected early isolation stress and socially reared mice. We are usually exposed to stress and the protection of the brain from stress is one of our big interests in our life.

Male histamine H1 receptor gene knockout mice (H1-KO) and their wild type (WT) mice were bred in our laboratory. After weaning (21 days old at the beginning of the experiment, 12-20 g) H1-KO and their WT mice were divided into two groups: socially reared mice kept in groups of four in plastic cages ($32 \times 22 \times 15$ cm) with sawdust bedding and non-socially reared (isolated) mice kept in one mouse per cage. The mice were divided into two groups: socially reared mice kept in plastic cages ($32 \times 22 \times 15$ cm) with sawdust bedding and non-socially reared (isolated) mice kept in one mouse per cage. The mice were divided into two groups: socially reared mice kept in groups of four in plastic cages ($32 \times 22 \times 15$ cm) with sawdust bedding and non-socially reared (isolated) mice kept in one mouse per cage. The mice were given a daily 50 mg/kg of 5-bromo-2'-deoxyuridine (BrdU) injection for five consecutive days. After 2-h final injection, mice brain were removed and immediately quick-frozen in dry ice to -40 °C. Each brain was



Fig. 1 Beam irradiation image of brain slices.

Fig. 2 Image of bromine $(90 \times 90 \ \mu m^2)$.

cryosectioned sagittally at 16 μ m thickness. The brain slices were mounted on polycarbonate films, which were processed with 5N sulfuric acid. The brain slices were examined with beam currents of almost 120 pA. PIXE data were obtained using incident 3 MeV proton microbeams.

It is well known that sedative antihistamines induce cognitive decline in humans through blockage of H1 receptor1), both facilitatory and inhibitory effects of neuronal histamine on learning and memory have been described in animal behavioral studies. Interestingly, we recently reported that blockage of histamine H1 receptor improves the impaired cognition by social isolation stress in mice²⁾. Early social isolation rearing from the age of weaning (typically 21- 28 days) into adulthood has been often used as an appropriate environmental factor to produce animal models with negative symptoms of schizophrenia³⁾. Figure 2 shows the beam irradiation image in region of $90 \times 90 \ \mu m^2$ in the brain slice of Fig. 1. The amount of bromine in the slices of Wild type, H1-KO mice and their subjected stress mice are shown in Fig. 3. The amount of bromine in brain slice of socially isolated WT mice was lower than that of WT social group mice. However, in the case of H1KO mice, bromine level was no change in the socially and isolation-reared mice.

These results suggested that the neurogenesis of H1-KO mice might be lower than their WT mice. And isolation-stress might caused neurodegenenesis in WT mice. **References**

- 1) Sakurai E et al., JAEA-review 2007(2006) 182.
- Yanai K et al., Histamine: biology and medical aspects (2004)251-263.
- 3) Dai H et al., Psychopharmacology 183 (2005) 285-293.



Fig. 3 Amount of bromine in $1 \,\mu\text{m}^2$ of brain slices.

3-67 Direct Visualization and Quantification of Anticancer Agent cis-diamminedichloro-platinum(II) in Human Lung Cancer Cells Using In-air micro-PIXE Analysis

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We designed an elemental analysis system using an ion micro-beam combined with a particle-induced X-ray emission (PIXE) method for the analysis of biomedical samples in air with a spatial resolution of 1 µm (in-air micro-PIXE system). We used this system to develop an imaging and quantification method for intracellular cis-diamminedichloro-platinum(II) (CDDP) in a human lung cancer cell line¹⁾. A human lung adenocarcinoma cell line, A549, was cultured and nuclear labeling was carried out by incubating the cells with BrdU. The cells were then exposed to CDDP at concentrations ranging from 1 µM to 1 mM, for 30 min to 24 hr. After drug treatment, samples were washed and frozen with liquid nitrogen, and freeze-dried for 24 hr. Standard samples were made using agar containing several concentrations of CDDP. Experiments using standard samples showed a linear correlation between CDDP

concentration and platinum signal strength. No clear platinum signal was detected after exposure to CDDP for 24 hr at doses between 1 and 100 μ M. However, significant Pt signals were observed at 1 mM. In that case, the Pt image of in-air micro-PIXE had enough quality to visualize nucleus and cytoplasm (Fig. 1). The detected signals of CDDP were stronger in the nucleus than in the cytoplasm (Fig. 1). A time-course study showed increased CDDP uptake in cells after longer drug exposure periods (Fig. 2). The use of this system enables the high-resolution visualization of intracellular CDDP distribution and measurement of intracellular CDDP concentrations.

Reference

1) H. Sakurai et al., Cancer Science 99 (2008) 901-904.



Fig. 1 Elemental maps of phosphorus and bromine using image analysis software. Intracellular CDDP is also estimated by measuring platinum count/nC in a selected area of the cells. Intracellular and intranuclear platinum are counted in individual cells.



Fig. 2 Intracellular and nuclear platinum count/nC after exposure to 1 mM CDDP. Increased CDDP uptake is observed in cells with linear correlation after longer drug exposure. CDDP uptake is higher in the nucleus than in the cytoplasm.

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4-01 Preparation of Epitaxial WO₃ Thin Films by Pulsed Laser Deposition

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Recently, the gasochromic materials, coloration by gases, have considerable promise as the optical hydrogen sensing materials. Tungsten trioxide (WO₃) thin films coated with noble metal (Pd, Pt) catalysts can be used for optical hydrogen sensors with an optical fiber network for the continuous monitoring of hydrogen leakage. In the present study, structural properties of epitaxial WO₃ thin films were investigated.

The growth of epitaxial WO₃ films on α -Al₂O₃(0112) substrate was performed by pulsed laser deposition (PLD). During the deposition, the substrates temperature was changed from 432 to 538 °C, and the pressure of oxygen gas was varied from 0.57 to 1.20 Pa. Typical thickness of WO₃ films was about 740 nm after 3 hours deposition. The crystallographic relationships between WO3 films and substrates were determined by x-ray diffraction measurements, using a high-resolution diffractometer (X'Pert, PANalytical). The RBS/channeling analysis using a 3MV single-stage-accelerator at JAEA/Takasaki was employed to characterize the epitaxial films. The analyzing 2.0 MeV ⁴He⁺ ions were incident, and backscattered particles were detected at 165° scattering angle.

Figure 1 illustrates a typical 2.0 MeV ⁴He⁺ RBS spectra from the WO₃ film taken under the random and the WO₃<001> aligned condition. The film was deposited on an α -Al₂O₃ (0112) substrate at 538 °C up to 740 nm thickness, under the 1.2 Pa O₂ gas pressure. The minimum yield, χ_{min} value, the ratio of the aligned spectrum to the random one at the fixed depth near the surface region, gives a measure to



Fig. 1 2.0 MeV 4 He ${}^{+}$ RBS/channeling spectra from a WO₃ film on the α -Al₂O₃ (0112) substrate. The film was deposited at 538 °C.

evaluate the degree of disorder in crystalline solids. The χ_{min} value in the WO₃<001> aligned spectrum is 5.7 % at the just area behind the surface peak of the W component, which suggests that a high quality epitaxial WO₃ film was grown on the α -Al₂O₃ (0112) substrate. Figure 2 shows the crystal quality of the films changes as a function of the substrate temperatures, by referring to χ_{min} values at the surface region. The results of RBS show that the crystal quality of WO₃ films is improved with an increase in the substrate temperature and the optimum temperature for the growth of high quality epitaxial WO₃ (001) on α-Al₂O₃ (0112) substrates is higher than 500 °C. The influence of O₂ gas pressure on the crystal quality of WO₃ films was examined. The WO₃ films were deposited on α -Al₂O₃ (0112) substrates in an O_2 gas pressure range of 0.7 to 1.3 Pa at a constant substrate temperature of 501 °C. The results indicate that the crystal quality of WO₃ films is improved with an increase of the O2 gas pressure during the deposition. The high quality epitaxial films were obtained above 1.0 Pa O₂ gas pressure at the substrate temperature of 500 °C. To confirm the in-plane orientations of epitaxial WO₃ films on α -Al₂O₃ (0112) substrate, pole figure measurements were carried out. The growth of WO₃ (001) films on the α -Al₂O₃ (0112) and the in-plane orientation with WO₃ [011] $//\alpha$ -Al₂O₃ [100] was confirmed. RBS and XRD results demonstrated that monoclinic WO₃ (001) films were successfully grown on the α -Al₂O₃ (0112) substrate. The crystal quality was improved by increasing both the oxygen pressure and the substrate temperature.



Fig. 2 The minimum yield for 2.0 MeV ${}^{4}\text{He}^{+}$ RBS from the W component of the WO₃ film as a function of α -Al₂O₃ (0112) substrate temperature during the deposition.

4-02 Effects of Temperature on the Gasochromic Behavior in Amorphous WO₃ Films

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Gasochromic behavior in amorphous WO₃ films was investigated by varying temperature of the films. Amorphous WO₃ films covered with 15 nm palladium layer were prepared by a reactive sputtering method. It was observed that the films colored from transparent yellowish to dark blue while in exposure of 1% hydrogen in a temperature range from 30 to 400 °C. The films, colored below 250 °C, bleached during exposed to air at room temperature. The hydrogen concentration in the films increased along with the coloration. However, the films, colored higher than 250 °C, could not be bleached by air exposure and did not incorporate hydrogen. These results suggested that the gasochromic mechanism changes around 250 °C that is a critical point of gasochromic behavior.

水素分子を解離させる触媒(Pt、Pd など)を表面に 堆積させた三酸化タングステン(WO₃)は、水素に曝 されて着色する(ガスクロミズム)ため、水素検知材 料への応用が期待されている。我々は、解離した水素 が WO₃中へ侵入し、水素タングステンブロンズ (H_xWO₃)が形成されることで着色すること、この H_xWO₃が酸素と反応し WO₃に戻ることで脱色するこ とを実験的に確認したが¹⁾、室温以上の温度における 着色・脱色挙動については未確認のままとなっていた。 そこで本研究では、30~400 ℃の温度範囲における非 晶質 WO₃膜のガスクロミック着色特性を調べた。

初めに、反応性スパッタリング法を用いて作製した 非晶質 WO₃ 膜試料¹⁾を、1%の水素を含んだ窒素ガス (N₂-1%H₂) に曝しながら透過光強度を計測し、着色特 性を評価した。その後、着色した試料を室温で大気に 曝し、同様な手法により、脱色特性を評価した。着色 前後の膜中水素濃度の定量には反跳粒子検出法 (ERDA)を用い、2.8 MeVのHeイオンの照射により 試料中水素をはじき出し(反跳させ)、その反跳水素量 を反跳水素エネルギーに対して計測した。

Fig.1は、水素曝露前後における波長 650 nm の透過率 と水素曝露時の試料温度の関係を示している。水素曝露 時の温度が 250 ℃ 以下では、試料は水素曝露によって着 色し、その後室温において大気に曝すと脱色した。一方、 水素曝露時の温度が 250 ℃ 以上になると、水素曝露によ る着色は 250 ℃ 以下の場合と同様な挙動を示したが、室 温で大気曝露を行っても脱色は起こらなかった。

次に、Fig.2に、水素曝露による試料中水素濃度の変 化を水素の反跳エネルギーに対して示す。反跳エネル ギーは水素が存在する試料深さに相当するため、30℃ で水素曝露させた試料では、着色に伴って膜内部で一 様に約0.27 H/W(6.2 at.%)の水素濃度が増加したこと になる。300℃で水素曝露した試料では、着色に伴う 水素濃度の変化は見られなかった。

以上の結果から、250 ℃以下の WO3 膜では、水素曝 露により膜中水素濃度が増加しながら着色し、大気曝 露により脱色したことから、室温における着色メカニ ズムと同様、H_xWO3の形成が着色の原因であると考え られる。一方、250 ℃以上では着色に伴う水素侵入が 見られないことから、H_XWO₃の形成とは異なる着色機 構の存在が示唆される。これは H_xWO₃の熱分解に関係 していると考えられ²⁾、分解時に WO₃中に酸素欠陥が 生成されたために着色したと推定している。



Fig. 1 Light transmittance at 650 nm of WO₃ films after hydrogen exposure (■) and after subsequent air exposure (○) plotted as a function of temperature for hydrogen exposure.



Fig. 2 Changing of hydrogen distribution in WO₃ films before and after hydrogen exposure at 30 °C (\blacksquare) and 300 °C (\circ)

- 1) A. Inouye, et al., Nucl. Instr. Meth. B 266 (2008) 31-307.
- 2) P. Dickens, et al., J. Solid State Chemi. 7 (1973) 241-244.

4-03 Gas Permeation Property of SiC Membranes Deposited on SiC Intermediate Layer

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Silicon Carbide (SiC) is a promising material for a hydrogen separation membrane because of its high mechanical strength and corrosion resistance at high temperature. To separate hydrogen molecule (H₂) from other gaseous species such as nitrogen (N₂), carbon mono oxide (CO) and hydrogen iodide (HI), dense SiC thin film was coated on porous alumina support¹⁾. However, preparing a crack-free thin film on such porous materials is technically difficult. To avoid crack formation, an intermediate layer is often added to fill pores of the support surface or release stress at the interface between the film and support during pyrolisis²⁾. In this paper, we describe the fabrication of a novel SiC hydrogen separation membrane used with a SiC intermediate layer.

SiC films were prepared on porous alumina supports by a dip-coating method with a cycrohexan solution of polycarbosilane (PCS) of 3 mass%. In order to prepare a SiC intermediate layer, the precursor film was immersed in cycrohexan after dipping, cross-linked at 453 K for 1 h in air, then pyrolyzed at 1123 K for 1 h. After the pyrolysis, another coating, cross-linking and pyrolysis was carried out for this support coated with the intermediate layer by using 10 mass% PCS solution. Hydrogen (H₂) and nitrogen (N₂) permeance of the membrane was measured at 523 K and the selectivity (H₂/N₂) was calculated as the ratio of the amount of H₂ to N₂ which passed thorough the membrane.

The surface of the SiC intermediate layer without the immersion is shown in Fig. 1(a). Black spots, which might

be SiC, appeared on the surface. It indicates a small amount of the precursor solution penetrated pores of the support. Figure 1 (b) shows the surface of the immersed intermediate layer. The outline of spots became dull, which indicates cycrohexane penetrate the support and dissolve PCS in pores during the immersion.

The dependence of H₂ and N₂ permeance of the SiC films on the immersion time is shown in Fig. 2. The H₂ permeance decreased with increasing the immersion time up to 20 seconds. It is supposed that PCS in pores is dissolved by cyclohexane and pores are filled with PCS solution during the immersion. As a result, the diameter of pores becomes smaller and smaller to decrease the permeance. For the immersion time longer than 20 seconds, the H_2 permeance increased. This result can be explained by the molecular sieving properties of the film, which leads to an improvement of the selectivity¹⁾. The N₂ permeance of the film decreased by the immersion, probably because the diameter of pores becomes small by the process. Here, a large value of the N₂ permeance at immersion time of 20 seconds is attributable to the formation of defects in the film such as pin-holes or cracks. Further investigations are necessary to clarify the mechanism behind the improvement.

- 1) T. Nagano et al., J. Ceram. Soc. Japan. 114 (2006) 533.
- 2) Y. Gu et al., J. Membr. Sci. 306 (2007) 216.



Fig. 1 Surface images of the intermediate layer. (a) un-immersed, (b) immersed for 20 seconds. Arrows point at black spots.



Fig. 2 Dependence of the permeance and selectivity of the SiC film on the immersion time.

4-04 Radiation Oxidation of Silicon-based Polymer Blends (II)

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Introduction

We have developed silicon carbide (SiC) coating for hydrogen gas separation membrane from a polymer blend of polycarbosilane (PCS) and polyvinylsilane (PVS). In the synthesis process of SiC coating, the PCS-PVS polymer blend is coated on the substrate, cured by irradiation in air, and pyrolyzed in an inert gas atmosphere. It has been found that the PCS-PVS polymer blend can be cured by the half dose of irradiation of pure PCS in air. However, the mechanism of radiation oxidation curing of the PCS-PVS polymer blend has not been cleared. We have investigated the radical behavior of the PCS-PVS polymer blends, and found that a tendency that the radical concentrations in PCS-PVS polymer blends were low as compared with those in pure PCS and PVS¹). The decay of radicals in case of the irradiation in air is considered to be attributed by (1) the reaction with oxygen, and (2) the recombination of radicals. In this work, we focused on above-mentioned (1), the amount of oxygen incorporated into PCS-PVS polymer blends by the irradiation was investigated.

Experimental procedure

Figure 1 shows the chemical structure of PCS and PVS. The PCS is a solid polymer at room temperature with the number average molecular weight of 2000. The PVS is a viscous liquid polymer at room temperature with the number average molecular weight of 960.

The PCS-PVS polymer blends and the predetermined quantity of oxygen gas were enclosed into a glass tube. The samples were irradiated by gamma rays with dose of 1 MGy at room temperature, and then the gases remained in the glass tube were analyzed by a gas chromatograph.



Fig. 1 Chemical structure of polycarbosilane (PCS) and polyvinylsilane (PVS).

Results and discussion

The gases evolved from the PCS-PVS polymer blends by the irradiation were composed of hydrogen (H_2), carbon dioxide (CO_2), carbon monoxide (CO), and methane (CH_4). The amount of oxygen incorporated into the PCS-PVS polymer blends was calculated by the subtraction of the amounts of oxygen remained and consumed as CO_2 and CO in the glass tube from the initial amount of oxygen. Figure 2 shows the oxygen concentration in the PCS-PVS polymer blends after the irradiation. As increasing in the PVS content, the oxygen concentration increased from 10 to 20 wt%. This indicates that the PVS promotes the oxidation of PCS-PVS polymer blends. This result explains well the reason of the decay of radicals as mentioned above.



Fig. 2 The oxygen concentration in the PCS-PVS polymer blends after the irradiation of gamma rays at room temperature in oxygen atmosphere.

Conclusions

In this work, the amount of oxygen incorporated into PCS-PVS polymer blends by the irradiation of gamma rays was investigated. It was found that the PVS promotes the oxidation of the PCS-PVS polymer blends.

Reference

1) A. Idesaki, et al., JAEA-Review 2007-060 (2008) 146.

4-05 Ceramic Nano Fiber Synthesis by Ion Beam Irradiation on Thin Film of Si-based Precursor Polymer

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Nano-structure materials with quite large surface area show excellent characteristics of filtering, absorption and reaction activity compared with bulk materials. Recently, nano fiber has been successfully obtained by the irradiation of ion-beam that produces cross-linking reactions within the ion beam track in polymer molecules.¹⁾ Therefore it is considered that precursor nano fiber is able to be obtained in ceramic precursor polymers by similar ion beam irradiation. If the obtained fiber is converted into ceramics by high temperature firing, ceramic nano fiber with high thermal and corrosion resistances is expected to be fabricated. However the synthesis method has not completed at present. In this research, we report the synthesis processes of silicon carbide (SiC) ceramic nano fiber by ion beam irradiation.

Polycarbosilane (PCS) as a ceramic precursor polymer, which was purchased from Nippon Carbon Co. LTD., was spin-coated on a Si substrate. The coated PCS was irradiated using 388 MeV 58Ni15+, 450 MeV 129Xe23+ and 500 MeV ¹⁹⁷Au³¹⁺ ion beams. The loss of kinetic energy of ions due to penetration through the films was estimated using the SRIM2006 code. The irradiated films were treated by toluene and insoluble in the solvent was developed as PCS nano fiber, then the nano fiber was fired at 1273 K in Ar. Direct observation was conducted using probe microscope (SII SPA-400). The PCS nano fiber was observed on the surface of substrate and remained after firing. Moreover, the shape of the nano fiber did not change by heating up to 1573 K in argon or at 1273 K in air. This indicates that the fired PCS nano fiber has good thermal and oxidation resistances. It is considered that PCS changes into amorphous SiC ceramics via an organic-inorganic conversion reaction similar to that of SiC fiber.



Fig. 1 The number density of PCS and SiC nano fibers. These are formed from PCS thin films with 400 nm thick on a Si substrate by 450 MeV Xe ion beam irradiation.

Figure 1 demonstrates the changes in the number density of isolated PCS nano fibers and SiC nano fibers in a unit area at various fluences. The number density of SiC nano fibers is almost the same as that of PCS nano fibers, and therefore the PCS nano fibers were not detached from the substrate during the conversion of PCS into SiC.

Radii of PCS and SiC nano fiber formed by ion beam irradiation with various LET are presented in Fig. 2. The solid line in Fig. 2 indicates the theoretical curve derived from Eq.(1) 1 ;

$$r^{2} = \frac{LET \cdot G(x)mN}{400\pi\rho A} \left[\ln \left(\frac{e^{1/2}r_{p}}{r_{c}} \right) \right]^{-1},$$
(1)

where A is Avogadro's number, m is the mass of the monomer unit, and N is the degree of polymerization. G(x) denotes the efficiency of cross-linking reaction. The value of mN/pA reflects the effective volume of a polymer molecule. The value of r_c or r_p represents the extent of cylindrical core or penumbra area, respectively. We had reported that Eq.(1) is corresponding to the experiment results of several kinds of polymer nano fibers well²⁾. In the case of the PCS nano fiber in Fig. 2, Eq. (1) also gives good interpretation to the experiment results. In the ceramization reaction of the PCS, the PCS nano fiber shrinks at a constant ratio, therefore the radii of the SiC nano fiber was closely controlled by the ion beam irradiation.

References

1) S. Seki et al., Adv. Mater., 13 (2001) 1663.

2) S. Seki et al., Macromolecules, 38 (2005) 10164.



Fig. 2 Radii of PCS and SiC nano fibers formed by ion beam irradiation with various LETs in PCS thin films.

4-06 Multi-Functional Polymer Nanowires by Single Particle Nano-fabrication Technique

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High energy charged particles have often been characterized by their "hard" interaction with the organic materials, giving cylinder-like nano-space of "ion tracks" along the particle trajectories where reactive intermediates are produced non-homogeneously with extremely high density. The chemical reactions within the limited nano-space are feasible to produce 1-D nano-materials, and we have successfully produced 1-D nanowires based on the crosslinking reactions in the polymer thin films as target materials for high energy charged particles. The present nano-scaled negative tone imaging technique (single particle nano-fabrication technique: SPNT) is applicable to a variety of polymers with fairly controlled sizes.¹⁻⁷⁾ According to these features of the SPNT, the characteristics of resists materials bearing high aspect-ratio tolerance have been examined in the present paper. The 1-D nanowires with ultra-high aspect-ratio as much as $\sim 10^3$ are demonstrated based on the commercially available SU-8 polymers, intending to a practical application as nano-sensing materials with extra-wide surface area.

High energy particles penetrating into the thin film of SU-8 promote effectively cross-linking reaction along the trajectories. The development procedure isolated the nanowires on the substrate as shown in Fig. 1, giving the nanowires with uniform length and thickness. All the nanowires are no longer standing, collapsed onto on the substrate observed as the 2-D images. The size (radius of cross section) of nanowires has been revealed to be in good



Fig. 1 AFM micrographs of nanowires with ultra-high aspect ration based on SU-8 produced by SPNT. Images (a) – (c) were observed in the films of SU-8 at ~10 μ m thick after irradiation of 400 MeV Kr ion beam at the fluence of 1.0×10^8 ions cm⁻².

correlation with the spatial distribution of deposited energy in an ion track by a penetrating particle and the parameters of the target polymers. The efficiency of cross-linking reactions has been also crucial parameter for the sizes, and the semi-empirical modeling¹⁻³⁾ gives the estimates of the crosslinking efficiency: G(x) as > 3.5 (100 eV)⁻¹ which is an appropriate value as the reaction promoted by chain reactions in the chemically amplified resists.⁵⁻⁷

The nanowires with ultra-high aspect ratio were demonstrated by an irradiation of thick SU-8 films (~ $10 \mu m$ thick). Fairly long nanowires are observed clearly on the substrate, and the number density of the nanowires matches exactly with the number of incident particles. In spite of same development protocol applied to isolate the nanowires, the length of the nanowires also reflect completely the initial film thickness. This is suggestive that the nanowire based on cross-linked SU-8 has sufficient mechanical strength bearing the solvation of surrounding uncross-linked molecules. The radius of the cross-section of the nanowires was estimated as 7.5 nm which is also in good agreement with the predicted value by the semi-empirical formulation and G(x) > 3.5. The radii of the nanowires were controlled from 4.5 nm (produced with 520 MeV Kr) to 17 nm (with 400 MeV Xe), and the maximum length of the nanowires reaches up to 12 µm. Fragmentation of the nanowires during development processes was not observed for all cases (variation of incident particles and film thickness), thus length of the nanowires was uniform and controlled perfectly by varying the film thickness.

The maximum of the aspect ratio of the nanowires estimated from their radii and the length reaches up to ~800, which is unlikely high value compared with those observed for the other polymeric materials. The previous studies on the cross-linked polymer nanowires prepared by SPNT have suggested the typical values of the aspect ratio as ~50 at maximum,^{8,9)} which is suggestive of ultra-high tolerance of SU-8 for nano-structure formation with extraordinarily high aspect ratio.

- 1) S. Seki, et al., Phys. Rev. B, 70 (2004) 144203.
- 2) S. Seki, et al., Adv. Mater., 13 (2001) 1663.
- 3) S. Tsukuda, et al., J. Phys. Chem., 108 (2004) 3407.
- 4) S. Tsukuda, et al., J. Phys. Chem. B, 110 (2006) 19319.
- 5) S. Seki, et al., Macromolecules, 39 (2006) 7446.
- 6) S. Tsukuda, et al, Appl. Phys. Lett., 87 (2005) 233119.
- S. Seki, et al., Surf. Coat. Technol., 2007, 201, 8486; ibid., 2007, 201, 8526.
- 8) D. Stumbo, et al, J. Vac. Sci. Technol. B, 11 (2003) 2432.
- 9) H. Lorenz, et al., J. Micromech. Microeng, 1997, 7, 121.

4-07 Effects of Ion Irradiation on Microstructure and Photoluminescence of SiC Nanotubes

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Since the discovery of carbon nanotubes (CNTs) in 1991, significant numbers of researchers have prepared new one-dimensional nanostructured materials such as nanotubes, nanorods and nanowhiskers for potential applications. Some of them have reported that many nanomaterials such as TiC, NbC, BN, C/C/N, SiO₂ and GaN nanostructures are synthesized from CNTs as the template. We reported that the C-SiC coaxial nanotubes, which were CNTs sheathed with SiC, were formed. Furthermore, the single-phase SiC nanotubes were successfully synthesized by heating the C-SiC coaxial nanotubes in air^{1,2)}. It is well known that SiC is the most important semiconductor because it has excellent stability against radiation. SiC nanotubes are, therefore, expected to be used for electrical and optical devices under environments. However, radiation-induced radiation microstructural change of SiC nanomaterials has not been well investigated. The purpose of this study is, therefore, to investigate the of ion-irradiation-induced microstructural change and photoluminescence of SiC nanotubes.

CNTs (GSI Creos Corporation, Tokyo, Japan) was used as the template. The C-SiC coaxial nanotubes were synthesized by heating CNTs with Si powder (The Nilaco Corporation, Tokyo, Japan) at 1200 °C for 100 h in a vacuum. Single-phase SiC nantoubes were formed by the heat treatment of C-SiC coaxial nanotubes at 600 °C for 2 h in air. Thin films of single-phase SiC nanotubes were prepared on the alumina plates by embrocating with the single-phase SiC nanotubes dispersed in ethanol. These thin films of single-phase SiC nanotubes were irradiated with

SICNTs as received SICNTs irradiation at 300 °C SICNTs irradiation at 900 °C SICNTs irradiation at 900 °C 290 288 286 284 282 280 Bidling energy (eV)



3.0-MeV Si^{2+} ions at 300 °C and 900 °C. The ion fluence was 6.4×10^{20} ions/m². Photoluminescence of SiC nanotubes dispersed in ethanol were measured at room temperature by YAG laser with the wavelength of 266 nm.

The XPS spectra of the C 1s bands for the SiC nanotubes before and after ion irradiation at 300 and 900 °C are shown in Fig. 1. There are two peaks representing Si-C and C-C in all specimens before and after ion irradiation. The energy difference between the peaks representing Si-C and C-C decreases with increasing the irradiation temperature. This result indicates that the binding state of SiC nanotubes changed by the ion irradiation at higher temperature.

Figure 2 shows the room temperature photoluminescence spectrum of SiC nanotubes dispersed in ethanol. A broad PL emission peak is observed at 390 nm. The ultraviolet-blue emission properties of SiC nanotubes are of significant interest for their potential ultraviolet-blue emitting device application. In near future, effect of ion irradiation on photoluminescence of SiC nanotubes will be investigated.

- T. Taguchi, N. Igawa, H. Yamamoto, S. Jitsukawa, J. Am. Ceram. Soc. 88 [2] (2005) 459-461.
- T. Taguchi, N. Igawa, H. Yamamoto, S. Shamoto, S. Jitsukawa, Physica E 28[4] (2005) 431-438.



Fig. 2 Photoluminescence spectrum of the SiC nanotubes dispersed in ethanol.

4-08 Preparation of Ion-Track Membranes of Poly(*p*-phenylene terephthalamide)

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There have been numerous studies on the ion-track membranes of poly (ethylene terephthalate) and polycarbonate, which are easily prepared by ion beam irradiation and the subsequent alkaline etching. However, the ion-track membrane of polyamide (PA) has not yet been reported because the etching sensitivity is inversely related to polymer stability. We report herein, for the first time, the investigation of ion-track etching of poly (*p*-phenylene terephthalamide) (PPTA), which is one of PAs with the highest mechanical strength as well as chemical or thermal stability.

Sixteen µm-thick PPTA films were bombarded by swift heavy ions from the TIARA cyclotron of the JAEA and the UNILAC linear accelerator of the GSI. The ion fluence was 3×10^7 ions cm⁻². The other irradiation conditions are listed in Table 1. The irradiated PPTA films were etched in a sodium hypochlorite (NaClO) solution at 40 °C without stirring, whose pH value was adjusted at pH 9±0.5 by adding a 2 mol dm⁻³ hydrochloric acid (HCl) solution. The etched sample was washed with a large amount of water and dried at 40 °C for 2 hours in vacuum. The surface and cross-section of the membranes were observed with a scanning electron microscope (SEM) after their Au coating. The conductometric measurement was applied to determine a track etching rate, V_t¹⁾. A bulk etching rate, V_b, was estimated at 0.05 µm h⁻¹ by taking the decrease in film thickness during one-hour etching. Using the ratio of the two etching rates, we obtained the track formation sensitivity, Q, according to the following simple equation:

 $Q=(V_t/V_b)-1$

Figure 1 (a) shows the SEM photographs of the surface and cross-section of the membranes, which were obtained by the irradiation with ²³⁸U ions and the 12-hour etching. This clearly confirms the perfectly cylindrical pores with a 0.3-µm diameter. The cylindrical shapes were also observed for the ¹⁹⁷Au ion irradiated films. In contrast, as shown in Fig. 1 (b), the tracks of ¹²⁹Xe ions were etched for 18 hours, leading to the observable tracks with a surface diameter of 0.8 µm. The cross section appeared to be not cylindrical all over the thickness of the membrane. The films irradiated

Table 1 Irradiation conditions and all the obtained results.

lon	Energy (MeV n⁻¹)	LET (MeV μm⁻¹)	Q	Pore shape
⁸⁴ Kr	6.2	6.7	2.4	Funnel
¹⁰² Ru	3.1	9.8	6.8	Funnel
¹²⁹ Xe	3.5	12.2	14.2	Funnel
¹⁹⁷ Au	11.1	15.4	62.4	Cylindrical
²³⁸ U	11.1	18.3	63.4	Cylindrical

with ⁸⁴Kr and ¹⁰²Ru ions provided such funnel-shaped pores, too.

All of the results, including the linear energy transfer (LET) evaluated by a TRIM code as well as the Q value and shape of the etched pore, are also shown in Table 1. The latent tracks of the ⁸⁴Kr, ¹⁰²Ru and ¹²⁹Xe ions exhibited lower sensitivity to the etching. This is probably why funnel-shaped pores appeared in the membranes irradiated with these relatively lighter ions, where the pore diameter at the film surface increased before the formation of through-holes. However, the track sensitivity of the heavier ¹⁹⁷Au and ²³⁸U ions were found to be about 4.5 times larger under the same etching conditions. The sharp increase in the Q value between the ¹²⁹Xe and ¹⁹⁷Au ions should be the origin of the transformation of the etched track from the funnel to cylindrical shape. This is a striking result, because the difference in the LET between these ions, which is just as much as that between the ¹⁹⁷Au and ²³⁸U ions, is considered to be very minor. In this case, the LET does not seem to be the only significant factor determining the sensitivity. It was reported that the damage distribution in the latent track depended on the beam energy even at a fixed value of the LET²). This so-called "velocity effect" in the damage creation might be manifested for the present etched track^{3,4)}.

References

(1)

- 1) P. Apel et al., Nucl. Instr. and Meth. B, 131 (1997) 55.
- 2) A. Meftah et al., Phys. Rev. B, 48 (1993) 920.
- 3) Y. Suzuki et al., Nucl. Instr. Meth. B, 260 (2007) 693.
- 4) T. Yamaki et al., GSI Scientific Report 2006 (2007)322.



Fig. 1 SEM photographs of the surface (top) and cross-section (bottom) of the PPTA-based ion-track membrane, which was prepared by (a) ²³⁸U and (b) ¹²⁹Xe irradiations.

4-09 Epitaxial Nitriding Processes of Titanium Thin Films due to N Implantation

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It has recently been reported that properties of non-stoichiometric titanium nitrides (TiN_v) such as electrical conduction, diffusion barrier, wear resistance, catalysis, etc. depend not only on chemical composition, but also on orientation relationships between TiNy films and substrates. Therefore, much interest has been focused on studying atomistic growth processes of TiN_v films¹⁾. The purpose of the present paper is to clarify atomistic growth processes of TiN_v films due to ion implantation by using in-situ transmission electron microscopy (TEM) and electron energy-loss spectroscopy (EELS), along with composition analysis and with the characterization of the electronic structure by molecular orbital calculation. The ions of N_2^+ with 62 keV are implanted into deposited Ti films in the 400-kV analytic high-resolution TEM combined with ion accelerators installed at JAEA-Takasaki²⁾.

Nitrogen ions (N_2^+) with 62 keV were implanted into the as-deposited Ti film composed of mainly (110)-oriented TiH_x and (03.5)-oriented hcp-Ti at room temperature, which results in the epitaxial formation of (110)-oriented and (001)-oriented TiN_v, respectively. In order to elucidate the atomistic nitriding processes of the epitaxial transformation of Ti thin films due to N implantation in detail, DV- $X\alpha$ MO calculations have been performed for the Ti_{19} cluster and $Ti_{19}N$ cluster models shown in Fig. 1(a). The Ti_{19} cluster of Fig. 1(a), which does not include a nitrogen atom indicated by an open circle G, corresponds to a part of the hcp-Ti structure. The Ti-Ti distances are taken to be 0.29238 nm, corresponding to those of the ideal bulk crystal structure. Figure 1(b) shows the contour map of the electron density of the Ti₁₉ cluster, which are drawn for the $\{\overline{2}1 \cdot 0\}$ plane including Ti atoms denoted by A, L, F, and J in Fig. 1(a). The site indicated by G in Fig. 1(b) is the central position (O-site) of the octahedron with larger space as formed by A-F atoms in the Ti₁₉ cluster, and has lower electron density (~1/5000 of electron density of A-site). Thus, O-sites have smaller repulsion for electrons of N atoms, and admit the invasion of N atoms, which leads to the formation of a $Ti_{19}N$ cluster as also shown in Fig. 1(a). Bonds between Ti atoms of the octahedron occupied by a N atom at its center, as the octahedron ABCDEF in Fig. 1(a), weaken, whereas bonds between Ti atoms of the octahedron not occupied by a N atom, as the octahedron ACDHIJ, do not change, or rather strengthen a little. On the other hand, the Ti-N bonds as A-G and F-G bonds indicated by dotted lines become relatively strong compared with the Ti-Ti bonds in the octahedron ABCDEF for Ti₁₉N. Thus, it can be considered that the strengthening of the A-G and F-G

bonds promotes the shear in the FL $<01\cdot0>$ direction indicated by an arrow on the (00•1)-plane including B, E and F atoms for Ti₁₉N. In order to obtain an fcc sublattice by the hcp-fcc transformation, the atoms on the (00•1)-plane including B, E and F atoms for Ti₁₉N have to be shifted. After the shift, the F atom, for example, has to be at the center of gravity of the BEF triangle. The projected line of the line FA to the (00•1)-plane including B, E and F atoms for Ti₁₉N is on the line FL. Furthermore, the inheritance of square atomic arrangement and the movement of the N atom to other neighboring O-site (M site in Fig. 1, for example), in the transformed fcc-Ti sublattice, is responsible for the epitaxial growth of TiN_y.

References

- S. Hao, B. Delley and C. Stampfl, Phys. Rev. B 74 (2006) 035424-1.
- 2) H. Abe et al., JAERI-Research 96-047 (1996) 1.



Fig. 1 (a) Schematic illustration of the Ti₁₉ cluster without a nitrogen atom indicated by an open circle G, and Ti₁₉N cluster with a nitrogen atom, G. (b) The contour map of the electron density of the Ti₁₉ cluster, which are drawn for the plane including Ti atoms denoted by A, L, F and J in (a).

4-10 Structural Change in Si Induced by Ion Bombardments and Its Application for Nano-Fabrication (II)

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In the previous study¹⁾, we investigated Ar⁺ and Si⁺ ion-beam-induced shrinking effects of the holes made in the 180nm-thick Si layers on silicon-on-insulator (SOI) substrates. In the present study, we have further investigated the chemical influences on the shrinking effects by using B⁺ and P⁺ ion implantation. The initial µm-size holes were prepared by ~30 keV Ga⁺ focused-ion-beam irradiation. B⁺ and P⁺ ions with the energy of 10 keV were implanted into the hole region at room temperature with doses of 4.4×10^{16} /cm² and 0.9×10^{16} /cm², respectively. In order to avoid possible carbon contaminations on the sample surfaces, the irradiation chamber was kept cooled by surrounding cold shrouds.

Figure 1 shows the transmission electron microscopy (TEM) photographs obtained from the SOI samples before and after B^+ ion irradiation. The structural changes induced by ion bombardments were analyzed based on the dark field (DF) and bright field (BF) images combined with the selected-area-diffraction (SAD) patterns. The shape of each hole was determined by using the BF image, while the Si crystallinity of the hole region was judged by the DF image using the Si(004) diffraction spot as well as the SAD patterns with the resolution of approximately 500 nm. In the P⁺ ion irradiation case, the TEM results were essentially the same as the Si⁺ ion irradiation case previously reported¹⁾.



Fig. 1 TEM photographs of the hole made in the 180 nm-thick Si layer on a SOI substrate before and after 10 keV B⁺ ion irradiation with the dose of 4.4×10^{16} /cm⁻².

The shrinking effects for the four ion species, Ar^+ , P^+ , Si^+ , and B^+ are summarized in Table 1. It should be noted that the SAD shows the crystalline Si spots and the shrinking effect is very weak in the B^+ ion bombardment case.

According to SRIM simulations for Ar^+ , Si^+ , B^+ and P^+ ion bombardments on Si at 10 keV, the projected ranges were estimated to be ~180 nm for Ar^+ , Si^+ , and P^+ , and ~450 nm for B^+ . Based on these experiments and simulations, we summarize our results as follows:

(1) Since there is no significant difference in the shrinking effect between Si^+ and P^+ ion bombardments, chemical effects may well be small.

(2) The shrinking effects can be attributed to dilation of crystalline Si upon amorphization induced by ion beam bombardments. The observed difference in ion species may be simply due to the difference in the numbers of the ion-beam-induced Frenkel-pairs, which is consistent with the D-D pair model for ion-beam-induced amorphization of Si proposed previously^{2,3)}.

- 1) T. Motooka, et al., JAEA-Review 2007-060 (2008) 169.
- 2) T. Motooka, Phys. Rev. B 49 (1994) 16367.
- 3) T. Motooka et al., Phys. Rev. Lett. 78 (1997) 2980.

Table 1	Ion-beam-induced	shrinking	effects	of	Ar ⁺ ,
P^+ , Si	$^{+}$, and B^{+} .				

	Ar^+	Р+	Si ⁺	B+
Shrinked Area (µm²)	0.83	0.17	0.76	0.23
Dose (10 ¹⁶ cm ⁻²)	1.5	0.9	4.3	4.4
Shrinked Area per Dose (10 ⁻²⁴ cm ⁴)	0.55	0.19	0.18	0.05

4-11 Electron Irradiation Effects on Heavily Boron-Doped Diamond Thin Films

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Heavily boron-doped diamond thin films show superconductivity around 2 K¹⁾. After the discovery, the importance of the crystal growth direction of the film was out²⁾. pointed The (111)-oriented films show superconductivity above 10 K. However, the mechanism of the superconductivity of this material is still unclear. In order to research the origin of the superconductivity, we have investigated the influence of point defects on the film with high energy electron irradiation. The sample was synthesized with high-frequency plasma chemical vapor deposition (CVD) method. The 2-MeV electron irradiation was performed at room temperature using the electron accelerator of JAEA-Takasaki up to the dose of 1×10^{18} e/cm². The defects introduced by the irradiation were estimated to be about 100 ppm. No significant change, however, was observed on superconducting properties, such as the transition temperature Tc, critical current densities Jc, and the upper critical field Hc, at this dose. Further irradiation on this material is needed. We are going to continue the irradiation furthermore in next year.

Here, we will introduce another attempt to synthesize B-doped diamond films using mega-gravity (MG) treatment. Figure 1 shows the setup of our centrifugal apparatus. Nano-scaled diamond colloid with fine boron particles was settled under 0.6 MG (1 G = 1 gravitational acceleration) centrifugal field at 350 °C for 19 hours, and deposited on a hydrophilic sapphire substrate.

After the mega-gravity treatment, a nano-diamond film was formed on the substrate. To evaluate the film thus obtained, ac magnetization and Raman spectra were



Fig. 1 A photo of our ultracentrifugal machine generating over one million-g. In the main apparatus shown in the left hand of the picture, a $\Phi 80$ titanium-alloy rotor is settled inside. It rotates more than 170,000 rpm by controlled compressed air flow.

measured. Unfortunately, superconductivity of the film was not observed around 2 K. The synthesis conditions must be optimized. Furthermore, the adhesiveness between the film and the substrate was not good enough, and the film exfoliated easily. This is another point to be improved on the synthesis of the nano-diamond films. It is possible, however, to compare the crystallinity between the front and the back of the film, i.e. the lower-G side and the higher. From the Raman spectrum measurements using an argon ion laser with a 488 nm wavelength, more intense peak around 1330 cm⁻¹ (green line in Fig. 2) originating from sp³-bonding of diamond is observed on the high-G side than on the low-G side. Two peaks in Fig. 2 around 1360 cm⁻¹ and 1580 cm⁻¹ originate from the sp²-bonding of graphite (black lines). Considering the fact that the excitation efficiency for the sp²-bonding with the Ar laser is more than several ten times larger than that of the sp3-bonding, it is possible to conclude that the diamond phase is more dominant at the higher-G side. In the present condition, however, the carrier doping in the diamond film is not achieved yet. And we will continue the efforts to find the optimum conditions to synthesize B-doped diamond thin films.

This research was performed in collaboration with Dr. T. Nishizaki of the Institute for Materials Research (IMR), Tohoku University.

- 1) E. A. Ekimov et al., Nature 428 (2004) 524.
- 2) Y. Takano et al., Appl. Phys. Lett. 85 (2004) 2851.



Fig. 2 The raman spectra of the lower-G (front) side and the higher-G (back) side of a mega-gravity treated nano-diamond thin film.

4-12 Ion-induced Self-organized Ripple Patterns on Graphite and Diamond Surfaces II

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The irradiation with heavy ions leads to various morphological evolutions of material surfaces as a result of a balance between roughening and smoothing processes. It is known that self-organized ripple patterns are formed on the surfaces in the case of off-normal incidence of ions. We concentrate on the study of ripple formation of carbon allotropes after irradiation under identical conditions. The ripples formed on carbon materials would be a key issue to discuss the mechanism of ion-induced liquid crystal alignment on diamond-like carbon. Further, periodical ripple patterns are expected to enhance a field emission property of diamond surface. Datta et al.¹⁾ observed ripples on diamond surface after irradiation with focused 5-50 keV-Ga⁺ ions at incident angles larger than 40°. Adams et al.²⁾ also demonstrated that focused 30 keV-Ga⁺ ions in the presence/absence of H2O formed periodical ripple patterns on diamond. According to the linear erosion theory developed by Bradley and Harper,³⁾ similar features should be observed for all different carbon materials since all surfaces become amorphous already after low fluence irradiation. Recently, Takahiro et al.4) experimentally showed that different features appeared on graphite, diamond and amorphous carbon surfaces. In the present work, highly oriented pyrolytic graphite (HOPG) and single crystalline diamond were irradiated under identical conditions in order to investigate the allotropic effect on ripple formation.

The HOPG (0001) sample $(1 \times 1 \times 0.1 \text{ cm}^3 \text{ in size})$, manufactured by Molecular Device Tools for Nano Technology, Russia, was cleaved with a Scotch tape just before irradiation. The diamond (001) sample $(0.3 \times 0.3 \times 0.05 \text{ cm}^3)$ is a commercially available Ib-type diamond. The samples were irradiated with Xe⁺ ions of 10–200 keV at an incident angle of 60° with respect to the sample normal at room temperature. The ion current density



Fig. 1 AFM images of HOPG (right) and diamond (left) irradiated with 200 keV-Xe ions at 60° to 2×10^{17} cm⁻². Arrows indicate the projection of ion-beam direction.

and ion fluence were 2–4 μ A·cm⁻² and 2×10¹⁷ Xe⁺·cm⁻², respectively. The surface topography of the eroded sample surfaces was examined with an atomic force microscope (AFM) in a contact mode.

Figure 1 shows AFM images taken for HOPG and diamond irradiated with 200 keV-Xe⁺. Ripples are clearly seen on the irradiated HOPG and diamond surfaces, and they are aligned perpendicular to the ion-beam projection. The averaged ripple spacing for HOPG is found to be approximately 500 nm, much larger than that for diamond (320 nm). The line profile analysis reveals that the amplitudes of ripples, defined as height differences between hills and valleys, are typically 105 nm and 38 nm for HOPG and diamond, respectively. Thus, large differences in ripple wavelength and amplitude between HOPG and diamond were recognized. As seen in Fig. 2, the irradiation energy dependence of ripple amplitude for diamond is similar to that of sputtering yield calculated by the SRIM code, indicating that sputter erosion predominates in ripple formation of diamond. On the other hand, ripple amplitude for HOPG increases almost linearly with irradiation energy, quite different from the energy dependence of sputtering yield. This tendency suggests anomalous erosion in roughening process of HOPG, probably due to the layered structure of this material.

- 1) A. Datta et al., Phys. Rev. B 63 (2001) 125407.
- D. P. Adams et al., J. Vac. Sci. Technol. B 21 (2003) 2334; Appl. Surf. Sci. 252 (2006) 2432.
- R. M. Bradley and J. M. E. Harper, J. Vac. Sci. Technol. A 6, 2390 (1988).
- 4) K. Takahiro et al., Nucl. Instr. Meth. B 256 (2007) 378.



Fig. 2 Ripple amplitudes as a function of Xe⁺ energy for HOPG (○) and diamond (●). The energy dependence of sputtering yields (▲) is also shown.

4-13 Interdiffusion Behavior at Spin-Injection Interfaces of Ferromagnetic Fe₃Si/Semiconductor Heterostructures

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Interdiffusion behaviors at Fe₃Si/Ge heterostructures were investigated by Rutherford Backscattering spectrometry (RBS). Annealing experiments revealed that the interdiffusion of Fe and Ge atoms was caused above 673 K. We confirmed that the interdiffusion through the interface changed the film composition close to the equilibrium line between FeGe and FeSi phases.

強磁性体 Fe₃Si の規則構造である DO₃相は、バンド 計算によって大きなスピン偏極度を持つと予想されて おり、またキュリー点が室温より十分に大きいため (840 K)、スピン注入デバイスへの応用が期待されてい る。われわれは、分子線エピタキシー(MBE)法を用い てエピタキシャル成長させた DO₃-Fe₃Si/Ge の界面の軸 配向性が、成膜温度に依存した Fe と Ge 原子の相互拡 散に大きく影響されることを明らかにした¹⁾。

本研究では、エピタキシャル界面を介した Fe と Ge 原子の相互拡散による界面構造・組成の変化を明らか にするため、不活性ガス(Xe)マーカーを用いて各原子 の拡散流束および拡散経路を検討した。

 $Fe_3Si/Ge(111)$ 試料に Xe^+ をイオン注入(230 keV, 10¹⁵ cm⁻²)して、界面より深い位置に Xe 分布を形成し た。赤外線ランプアニールを行った試料の Fe, Ge, Si および Xe 原子の深さ分布を RBS により測定した。イ オン注入により Fe_3Si 層は非晶質化したが、イオンミ キシングによる組成の変化は起こらなかった。

Fig. 1 に(a)イオン注入後と(b)アニール(400 °C, 30 min)後の各原子の深さ分布を示す。アニール後、Fe 原子のGe基板側への拡散とGe原子のFe₃Si層への拡 散が確認され、Si原子はほとんど拡散していないこと が分かった。アニール後のFeとGe原子のMatano界 面がほぼ一致していることから、FeとGeの相互拡散 を実証できた。またアニールによりMatano界面はGe 基板方向に34 nm移動し、Ge基板内にあったXeマー



Fig.1 Depth profiles of Fe, Ge ,Si and implanted Xe atoms in (a) the as-implanted sample and (b) the annealed sample.

カーも同じ方向に 40 nm 移動した。これは、Ge 基板 側から Fe₃Si 層への拡散流束がその逆方向より大きい ことを示している。

次に、各原子の相互拡散流束 \tilde{J}_i と3元系相互拡散係数 \tilde{D} を測定した各原子の深さ分布 $C_i(x)$ から以下の式(1)-(3)を用いて計算した²⁾。

$$\widetilde{J}_{i}(x) = \frac{1}{2t} \int_{C_{i}(-\infty)}^{C_{i}(x)} (x - x_{o}) dC_{i}$$

$$\tag{1}$$

$$\widetilde{J}_{i}(x) = -\widetilde{D}_{i,Fe}^{Si} \frac{\partial C_{Fe}}{\partial x} - \widetilde{D}_{i,Ge}^{Si} \frac{\partial C_{Ge}}{\partial x} \quad (i=Fe, Ge)$$
(2)

$$\widetilde{D} = \sqrt{(\widetilde{D}_{FeFe}^{Si} - \widetilde{D}_{GeGe}^{Si})^2 + 4\widetilde{D}_{FeGe}^{Si}\widetilde{D}_{FeGe}^{Si}}$$
(3)

Fig. 2 に Fe₃Si/Ge 拡散対の組成変化と拡散経路およ び代表的な組成で求めた 3 元系相互拡散係数 \tilde{D} を示 す。本実験では,拡散経路を決定するために 400 °C よ りも短時間で組成変化の大きい 450 °C で試料をアニ ールした。Fe₃Si/Ge 拡散対では Si 濃度はほとんど変化 せず、主に Fe と Ge 原子の相互拡散により組成が変化 し、FeSi と FeGe を結ぶ相平衡線上に近づくことが確 認できた。さらに相平衡線での 3 元系相互拡散係数は 他の組成に比べて 2 桁ほど小さいことが分かった。

以上の結果より、Fe₃Si/Ge ヘテロ構造は Fe と Ge 原 子の相互拡散により2相平衡(FeGe+FeSi)した界面 構造に最終的に変化することが明らかになった。

- 1) Y. Maeda et al., Appl. Phys. Lett. 91 (2007) 171910.
- 2) K.M. Day et al., J. Phase Equilib. Diffus. 26 (2005) 579.



Fig. 2 Ternary interdiffusion coefficients and diffusion path in the isothermal section of the Fe-Ge-Si phase diagram at 450°C.

4-14 Control of Superconducting Properties of Y and Gd Series Tapes due to High-energy Xe Ion Irradiation followed by Thermal Annealing

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Thickness and element dependence of pinning properties of superconducting tapes prepared by pulsed laser deposition (PLD) method was investigated after ion irradiation and thermal annealing treatment in order to control their superconducting properties. In this study, YBCO tapes (layer thickness: 0.8 and 2.25 μ m) and GdBCO tapes (layer thickness: 2.25 μ m) were irradiated with 450-MeV Xe²³⁺ from 1.1×10^{10} to 7.6×10^{10} ions/cm². As a result of ion irradiation, the enhancement of J_c was observed at the liquid nitrogen temperature of 77 K. After the ion irradiation, the samples were annealed at the temperature of 473 K or 673 K in the oxygen atmosphere for 2 h. From this study, it was clarified that the ion irradiation is effective in enhancing J_c especially for superconducting tapes which have thick YBCO layer, and that J_c lowers by the annealing after irradiation because radiation-induced defects are partly recovered by the thermal annealing.

銅酸化物高温超伝導体(HTSC)は液体窒素温度であ る77 Kにおいて超伝導性を示すため、その超伝導応用 へ向けて種々の研究開発が行われている。HTSCは第 2種超伝導体に分類され、その超伝導臨界電流密度(J_c) を向上させるためには、超伝導体内に侵入した磁束を 捕捉するための常伝導領域、すなわち格子欠陥などの ピニングセンターを導入することが必要不可欠である。 量子線照射は、超伝導材料の製作過程とは独立してピ ニングセンターの形状や密度を制御できるメリットが ある。そこで本研究課題においては、現在開発が進め られている超伝導線材である、Pulsed laser deposition 法 (PLD法)により作製されたYBCO線材(Y系PLD線 材)へ重イオンビーム照射を用いて照射欠陥を導入し、 J_cなどの超伝導特性の変化を調べている。

線材を対象としたこれまでの研究では、同一の超 伝導線材に対して照射イオン種やフルエンスを変えて 高エネルギー重イオン照射を行い、超伝導特性の変化 を観察した¹⁾。そこで本研究では、厚みや組成の異な る高温超伝導線材に対してXeイオン照射および照射 後アニールを行い、線材の超伝導特性を制御するため の情報を得ることを目的とした。

照射用試料は(財)国際超電導産業技術研究センター 超電導工学研究所において作製されたYBCO超伝導線 材(YBCO層:0.8, 2.25 μ m)およびGdBCO線材(GdBCO 層:0.8 μ m)を2 mm×2 mm×0.5 mm程度に切り出して 作製した。高エネルギー重イオン照射にはTIARAの AVFサイクロトロンを使用した。照射は保護層である Ag層をケミカルエッチングによって除去した試料に、 放射化を抑えるために10 μ mの銀箔をのせ、超伝導体 のc軸に平行に照射した。照射イオン種は超伝導層を 貫通する条件として、450 MeVのXe²³⁺を選択し、照射 フルエンスは1.1×10¹⁰~7.6×10¹⁰ [ions/cm²]の範囲で 行った。 J_c はPPMS (Physical Properties Measurement System: Quantum Design社製)を用いて照射前後で測定 した。アニール実験は照射後の測定を行った試料に対 し、473 Kまたは673 Kの温度について酸素雰囲気中で 2時間保持した後銅板上で急冷し、その影響を調べた。 Fig. 1に77 K、印加磁場 4 Tにおけるアニール前のJ。 向上幅とXe照射フルエンスの関係を示す。まず、 YBCOの0.8 µm厚と2.25 µm厚を比較すると、2.25 µm の試料の向上幅は、低いフルエンスから大きくなる傾 向がある。この結果は超伝導薄膜においては薄膜の厚 さが厚いほどピニング力が強くなるという理論と一致 した傾向である²⁾。また、Y系線材とGd系線材を比較 すると、Gd系線材はLが低下する傾向にあることが分 かる。これは重イオン照射により生ずる円柱状欠陥の 径がGd系線材ではY系線材より小さく、磁束をピン止 めする力が弱いため、照射損傷による超伝導性の劣化 の方が影響するためであると考えられる。本研究にお ける照射後アニール実験では、アニールにより欠陥径 が小さくなることで、ほぼ全ての試料でJ。が低下した。 本研究から、膜厚の厚い線材の方が照射によるJ。の向 上が大きい傾向にあること、Y系線材の方がGd系より も照射の効果が大きいことが分った。



本研究の一部は超電導応用基盤技術開発業務の一環とし て新エネルギー・産業技術総合開発機構(NEDO)の委託に より実施された。

- 1) K. Nakashima et al., Physica C 463-465 (2007) 665.
- 2) H. Wada et al., Physica C 378-381 (2002) 570.

4-15 Effects of Ion Irradiation on Hydrogen Storage Characteristics of Alkaline Pretreatment Mm Alloy

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Mm (Misch metal) based alloys have been applied to the negative electrodes of Ni-H batteries in recent years because of their excellent hydrogen storage properties. In order to apply such alloys to hydrogen storage system in automobiles, further improvement of their hydrogen storage properties is indispensable. In previous studies^{1,2)}, we have reported that the alkaline pretreatment of the alloy surface using LiOH, NaOH or KOH accelerates the rate of the initial activation. In this study, we investigate the effects of K ion (K⁺) irradiation on the electrochemical hydriding rate of the alkaline pretreatment Mm based alloys³⁾.

The samples used in this study were $MmNi_{3.48}Co_{0.73}Mn_{0.45}Al_{0.34}$ (Mm = $La_{0.35}Ce_{0.65}$) pellets. The weight, diameter, and thickness of the pellets are 1.08 g, 12 mm and 1.2 mm, respectively. K⁺ irradiation into the Mm pellets was made at an acceleration energy of 350 keV with a dose of 1×10^{16} cm⁻² using the 400 kV ion implanter The samples were soaked in 6M-KOH at TIARA. solutions at 373 K for 30 min as the alkaline pretreatment after the ion irradiation. In order to reduce the hydroxide layers formed by the pretreatment, the surface of the pretreated samples was rinsed in fresh distilled water at room temperature. For the electrochemical measurements of the hydriding rate, either un-irradiated or ion irradiated Mm pellet was used as an anode. An Hg/HgO electrode was used as the reference electrode^{1,2}. For comparison, the hydriding rate was examined for the samples in which pretreatment was not performed.

Figure 1 shows hydrogen absorption curves of ion irradiated/un-irradiated Mm pellets with/without 6M-KOH pretreatment. The hydrogen absorption rate of Mm pellet with 6M-KOH pretreatment increased as compared that of alloy without pretreatment. Since the concentration of K atoms at the surface of Mm pellet becomes high by 6M-KOH pretreatment^{1,2)}, this result suggests that the hydriding rate is improved by K atoms at the surface.

Figure 2 shows the change in the hydrogen absorption rate (change rate) for Mm pellets with/without K^+ irradiation. The change rate is defined as the ratio of hydrogen absorption rate for 6M-KOH pretreatment sample to that for non-pretreatment one. The change rate for K^+ irradiated Mm pellet was higher than that for un-irradiated Mm pellet, for example the value for the ion irradiated Mm pellet was more 2 times higher than that for un-irradiated one at 1 min. This result indicates that the effect of 6M-KOH pretreatment on hydrogen absorption rate was enhanced by K^+ irradiation. The mechanism behind the improvement of the hydrogen

absorption rate by K^+ irradiation has not yet been clarified. Further investigations are necessary to reveal this mechanism.



Fig. 1 Hydrogen absorption curves of K⁺ irradiated and un-irradiated MmNi alloys with KOH pretreatment or without pretreatment.



Fig. 2 Change rate curves of H/M for K+ irradiated MmNi alloys and un-irradiated ones.

- H. Uchida, K. Yamashita and M. Goto, J. Alloys comp. 330-332 (2002) 662.
- H. Uchida, K. Yamashita, T. Tabata, H. Uchida, M. Iwase and Y. Katoh, J. Alloys and Comp. 293-295 (1999) 751
- 3) H. Abe et al., J. Alloys and Comp. 408-412 (2006) 348.

4-16 Atomic Displacements, Electrical Resistivity and Lattice Defects in FeRh Induced by Electron Irradiation

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For last three years, we have performed the electron irradiation experiments on Fe 50%Rh intermetallic compound at the 5th target room of TIARA single-ended accelerator. Here the experimental results are summarized. The most fundamental and important parameter for estimating the amount of defects induced by charged particle irradiation is a threshold energy of atomic displacements. We have therefore determined the threshold values for both of Fe atoms and Rh atoms. A sheet of FeRh alloy was irradiated with 0.5-2.0 MeV electrons near 20 K and the change in electrical resistivity was measured in-situ as a function of electron fluence for each electron energy. Figure 1 shows the dependence of electrical resistivity change per unit fluence (resistivity change rate) on the electron energy. Below the energy of 0.5 MeV, resistivity change rate is zero. This means that neither Fe nor Rh atom can be displaced by electron with the energy of <0.5 MeV. Above the energy of 0.5 MeV, a finite value of resistivity change rate appears, implying that Fe atoms can be displaced above 0.5 MeV. Around 1 MeV, a kink is observed in the experimental curve of resistivity change rate. This shows that in addition to Fe atoms, Rh atoms also get displaced from this energy. As can be seen in Fig. 1, data analysis by using the formula of McKnley and Feshbach, the values of threshold energy for atomic displacements have been determined as 30 eV for Fe atom. The same analysis has been done after the subtraction of the contribution of Fe atom displecements (Fig. 2). From the curve fitting, we have determined the threshold energy for Rh atom displacemens as 45 eV.

Next, the effect of energetic electron irradiation on the magnetic transition of FeRh alloy was studied. It is well known that Fe-50%Rh alloy shows ferromagnetic(FM)antiferromagnetic(AF) transition near the room temperature and that this magnetic transition is accompanied by the change in electrical resistivity. Therefore, by measuring the electrical resistivity change in-situ during the electron irradiation, we can observe the change in magnetic transition temperature by electron irradiation. The value of electrical resistivity for unirradiated sample decreases suddenly near 300 K, corresponding to the FM-AF transition. After the irradiation, the temperature where the value of electrical resistivity changes shifts to a lower temperature side. It shows that the AF-FM magnetic transition temperature decreases by electron irradiation. This phenomenon has been confirmed by the magnetization measurements using a SQUID magnetometer.

Finally, we studied electron irradiation induced lattice defects in FeRh by means of positron annihilation technique. The relationship between S-parameter and W-parameter for FeRh alloy irradiated with 0.8 MeV and 2 MeV electrons shows that the electronic structure around irradiation-induced vacancy-type defects depends on the energy of electrons.



Fig. 1 Resistivity change rate as a function of electron energy. Results of curve fitting for Fe atom displacement are also shown.



Fig. 2 Results of curve fitting for Rh atom displacement.

4-17 Intrinsic Defect Formation in Amorphous SiO₂ by ⁶⁰Co γ-ray Irradiation

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Fluorine-doped synthetic amorphous SiO_2 (a-SiO₂) is suitable for investigating intrinsic defect processes in a-SiO₂ because the concentrations of extrinsic defect precursors are lower than those in fluorine-free a-SiO₂ commonly used for telecommunication fibers and ultraviolet optical components. In ⁶⁰Co γ -ray-irradiated fluorine-doped a-SiO₂, oxygen vacancies (Si-Si bonds) are the most abundantly formed. The concentrations of Si–Si bonds and interstitial oxygen molecules (O₂) increase almost linearly with the γ -ray dose. These observations indicate that the primarily γ -ray-induced intrinsic defect process in a-SiO₂ is not a simple cleavage of an Si–O bond into a pair of dangling bonds, but the formation of the Frenkel pairs (oxygen vacancy and interstitial), demonstrating a similarity of intrinsic defect processes between amorphous and crystalline materials, but the structural disorder of a-SiO₂ most likely enhances the formation of the Frenkel pairs.

アモルファスSiO₂(a-SiO₂)の真性欠陥過程には、ダングリングボンド対の形成

$$\equiv$$
Si-O-Si $\equiv \rightarrow \equiv$ Si[•] + [•]OSi \equiv (1)

と、Frenkel対の形成

 \equiv Si-O-Si $\equiv \rightarrow \equiv$ Si-Si $\equiv + O^{0}(2)$

の二種類あることが知られている。反応(1)は、生成 種のダングリングボンド対(E'中心 ≡Si[●]、NBOHC ≡SiO[●])が、電子常磁性共鳴(EPR)という、微量常磁性 種の解析に適した方法よって鋭敏に検出できるため、 非常に良く研究されている。反応(2)は、架橋酸素の 変位による酸素欠陥(Si-Si結合)と格子間酸素原子

(O⁰)の形成であり、アモルファスにおけるFrenkel 欠陥過程とみなせる。しかし反応(2)は、生成種が非 磁性であるためEPRを用いて研究できず、さらにO⁰は 光学的手法による検出も難しいため、見過ごされがち である。そのうえ、気相法で得られた合成a-SiO₂中に 残留しやすいSiOH基、SiCI基、SiH基などの官能基は、 光や放射線照射によってE中心やNBOHCへ変化しや すく、反応(1)以外の外因的ダングリングボンド源と なる。このように、これまでの研究では反応(1)のみ が注目されていたため、反応(1)と(2)のどちらが主過 程であるのかは明らかではなかった。

本研究の目的は、反応(1)と(2)のどちらが、a-SiO₂の 主な真性欠陥過程であるかを調べること、また、a-SiO₂の構造の乱雑さが、反応(1)と(2)とにどのように 影響するかを調べることである。

外因性欠陥の形成を極力避けるため、試料として、 SiOH基、SiCl基、SiH基に比べてはるかに分解しにく いSiF基を含むフッ素ドープa-SiO₂ (SiF ~4×10¹⁹ cm⁻³、 SiOH <10¹⁷ cm⁻³)を用いた。試料を900または1400[°]C で熱処理した後、核的衝突効果を考慮する必要のない ⁶⁰Co γ線を励起源とし、最大~ 5.3×10^7 Gyまで照射を 行った。生成したNBOHC、E'中心、Si-Si結合の濃度 をそれぞれ4.8、5.8、7.6 eV帯の光吸収測定によって、 またO⁰が二量化して生成した格子間酸素分子(O₂)の濃 度を、波長765 nmで励起した一重項O₂の0.97 eV (1272 nm)の発光測定によって決定した。 Fig. 1に欠陥濃度の変化を示す。照射によってSi-Si 結合が最も多く生じた。Si-Si結合の濃度はダングリン グボンド(E'中心とNBOHC)の濃度よりも1桁弱大きく、 またO₂として検出されたO⁰の濃度(O₂濃度の2倍)も、 高線量域でダングリングボンド濃度を上回った。さら に、低線量域では、Si-SiおよびO₂濃度は線量にほぼ 比例して増大した。以上の結果は、式(1)でなく、式 (2)が主な真性欠陥過程であることを示している。 Frenkel対濃度は、吸収線量に対して次第に飽和した。 また、熱処理温度を高めて構造の乱雑さ(安定角から 大きく歪んだSi-O-Si結合の濃度)を増すことで増大し た。Frenkel対は、構造乱れのない結晶での真性欠陥 であることが知られているが、今回の結果は、アモル ファスの構造乱れは、Frenkel対の前駆体を提供し、 その生成効率を高めうることを示している。

Si-Si結合および格子間O₂の生成の量子効率はそれ ぞれ~2×10⁻⁵、~1×10⁻⁶と見積もられ、前者はプロト ン照射によって高密度電子励起したa-SiO₂での報告値 (~1×10⁻⁵)と良く一致した。



Fig. 1 Dependence of the concentrations of NBOHC, E' center, Si-Si bond, and interstitial O_2 on absorbed γ -ray dose for fluorine-doped a-SiO₂ thermally annealed at 900 or 1400 °C prior to the γ -ray irradiation.

4-18 Observation of Ultraviolet-Light Emission from Si-Ion Implanted Fused-Silica Substrates

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Various works on silicon (Si)-based luminescent materials utilizing the quantum confinement effect, such as Si nanocrystals (Si-nc's), have been reported. Typical fabrication methods of Si-nc's are co-sputtering of Si and SiO_2 ,¹⁾ laser ablation,²⁾ and Si-ion implantation into SiO_2 plates or thermally oxidized Si wafers.³⁾ We previously fabricated Si-nc's including fused-silica substrates by using the Si-ion implantation method with subsequent annealing and observed blue photoluminescence (PL) from the substrates.⁴⁾ In this work, we observed ultraviolet (UV)-light emission from Si-ion-implanted fused-silica substrates under other implantation conditions.

Si ions were implanted into a fused-silica substrate $(10 \text{ mm} \times 10 \text{ mm} \times 1 \text{ mm}^{t})$ at room temperature in TIARA. The implantation energy was 80 keV, and the implantation amount was 2×10^{17} ions/cm². The Si-implanted substrate was cut into four pieces $(0.5 \text{ mm} \times 0.5 \text{ mm} \times 1 \text{ mm}^{t})$ using a diamond-wire saw, and the pieces were annealed in air for 25 min at 1100, 1150, 1200, and 1250 °C in a siliconit furnace. PL spectra were measured at room temperature with excitation using a He-Cd laser (λ =325 nm). A monochromator, a photomultiplier, and a lock-in amplifier were used in our measurements. Figure 1 presents the PL spectra of the four samples. UV-PL spectra having peaks around a wavelength of 370 nm were observed from all the samples. The UV-peak wavelengths of the samples are almost the same in spite of the various annealing temperatures. In our experiments, the UV-PL peak had a maximum intensity after annealing at 1250 °C, and the longer wavelength PL peak around 800 nm observed from the sample annealed at 1100 °C disappeared.

It is well known that UV-light emission can be obtained from Si-based materials fabricated by various methods. UV PL having wavelengths of from 310 to 370 nm was observed from thin films containing Si-nc's prepared by reactive laser ablation of Si targets.⁵⁾ UV-PL spectra having a peak wavelength of around 370 nm were obtained from sputtered SiO₂ films including Si-nc's produced after thermal annealing at 800 to 1100 °C.⁶⁾ The UV-light emission might originate from emission centers of interface layers between Si-nc's and SiO₂ matrices, and its intensity might be affected by sizes of Si-nc's.⁶⁾ Sample A of oxidized porous Si in ref. 7 emitted UV light around a wavelength of 370 nm after thermal oxidization at 700 to 1150 °C, and it was also explained that the UV-light emission seemed to originate from Si-nc's and interface layers between Si-nc's and SiO₂ produced in the thermal oxidization process.⁷⁾ Therefore, it seems that Si-nc's and interface layers are formed in the samples.

In addition to a blue PL peak, a PL peak around a wavelength of 800 nm in a wavelength range from red to infrared was observed for the samples annealed at 1100 and 1150 °C. Such longer-wavelength peaks have been observed from Si/SiO₂ multilayered films and were described as originating from interface layers between Si and SiO₂.⁸⁾ From this consideration, the two PL peaks of our samples may also originate from interface layers between Si-nc's and SiO₂ media. However, we successfully obtained only the UV-light emission peak from our Si-ion-implanted samples after optimizing of the implantation conditions.

UV-light-emitting materials are expected to be useful as light sources for optical pick-up systems. Therefore, we are trying to optimize the conditions of Si-ion implantation and annealing to improve the emission intensity and evaluate optical gains of the Si-implanted samples.

- 1) O. Hanaizumi et al., Appl. Phys. Lett. 82 (2003) 538.
- 2) Y. Yamada et al., Jpn. J. Appl. Phys. 35 (1996) 1361.
- 3) L. Pavesi et al., Nature 408 (2000) 440.
- K. Miura et al., Nucl. Instr. and Meth. in Phys. Res. B 263 (2007) 532.
- 5) I. A. Movtchan et al., Thin Solid Films 255 (1995) 286.
- 6) H. Z. Song et al., Appl. Phys. Lett. 72 (1998) 356.
- 7) J. Lin et al., Solid State Commun. 97 (1996) 221.
- 8) K. Miura et al., Thin Solid Films (2008), in press.



Fig. 1 Measured PL spectra of Si-ion-implanted fused-silica pieces annealed at 1100, 1150, 1200, and 1250 °C.

4-19 RBS Study of Axial Orientation of Fe₃Si Epitaxially Grown on Ge(111) and Its Low Temperature Epitaxy

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Ordered Fe₃Si (DO₃ structure) has been attracting much attention as a highly spin-polarized ferromagnetic material that can be adapted to a few spin-injection devices. Fe₃Si can be classified as a Heusler alloy and can be expected to have highly spin polarized properties and a high Curie temperature of 567 °C, both of which are advantageous in enhancing spin-injection efficiency. The molecular beam epitaxy (MBE) growth of Fe₃Si on Si, Ge, or SiGe substrates could further enhance its applicability to IV-group-based spin-electronic devices. For Ge substrates, we have successfully accomplished high-quality epitaxial growth only on the (111) planes^{1,2}. However, the dominant factor influencing the epitaxial growth of Fe₃Si on Ge and Si has not yet been investigated in detail.

In this study, we report the RBS characterization of $Fe_3Si/Ge(111)$ hybrid structures synthesized by MBE and discuss the crucial factors for the realization of high-quality epitaxial growth on Ge(111) substrates.

Ferromagnetic Fe₃Si layers were grown on Ge buffer layers (thickness: 20 nm) grown epitaxially on Ge(111) substrates by employing a solid-source MBE process using Fe and Si co-evaporation (deposition rates: 0.12-0.16 nm/s for Fe and 0.04 nm/s for Si). Rutherford backscattering spectroscopy (RBS) using 2 MeV-⁴He⁺; random spectra for determining the depth profiles of the concentration and aligned spectra for evaluating the quality of the axial orientation of Fe₃Si along the Ge<111> crystal orientation were measured. The backscattering angle was set to 165°.

Figure1 shows the Fe-channel random and aligned spectra



Fig. 1 RBS spectra of both compositions Fe₃Si prepared at growth temperatures of 200 and 300 °C. The left and right dotted lines correspond to the Fe₃Si/Ge interface and Fe₃Si surface, respectively.

along the Ge<111> axis for the (4:1) and (3:1) composition films, which were prepared at growth temperatures of 200 and 300 °C, respectively. In this growth temperature range, we observed spectrum changes for both the films. The aligned spectra near the surface were affected by the presence of the surface oxide layer; therefore, we neglected the analysis of these surface regions in the calculation of the χ_{min} values.

Figures 2(a) and 2(b) show the changes in the Fe and Ge concentrations near the interface and χ_{min} as a function of growth temperature, respectively. It indicates that highquality epitaxial growth can be realized when χ_{min} is smaller than 4% at 130 °C and below 200 °C for off-stoichiometric (4:1)- and stoichiometric (3:1)-Fe₃Si, respectively. In the case of stoichiometric (3:1)-Fe₃Si grown below 130 °C, we succeeded in realizing highly axial oriented crystal growth for the smallest χ_{min} value of 2.2% at such low growth temperatures. However, the increase in χ_{min} was induced by the concentration change above 200 or 300 °C. We found that this change in concentration was due to the interdiffusion between Ge and Fe atoms at the interface. This interdiffusion of Ge and Fe atoms at the Fe₃Si/Ge interface strongly induces pronounced degradation in the epitaxial growth with the axial orientation of Fe₃Si.

- 1) T. Sadoh et al., Appl. Phys. Lett. 89 (2006) 182511.
- 2) Y. Maeda et al., Appl. Phys. Lett. 91 (2007) 171910.



Fig. 2 Concentrations of the Fe and Ge atoms of the silicide layers at the interfaces and χ_{min} as a function of growth temperature.

4-20 Evaluation of Radiation-damaged Halo in Quartz by Cathodoluminescence

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Cathodoluminescence (CL) microscopy and spectroscopy were conducted on natural low-quartz samples after He^+ ion implantation in order to quantitatively evaluate the relationship between dose and CL halo development.

He⁺ implantation experiments were performed with a 3MV-tandem ion accelerator at the Takasaki Advanced Radiation Research Institute of Japan Atomic Energy Agency, where the condition was set at 4 MeV with a dose density over 1.77×10^{-5} C/cm². The homogeneous He⁺ ion beam was irradiated perpendicularly on the polished surface of the quartz chips, which were prepared from quartz monocrystal from Minas Geras, Brazil. CL imaging and spectral measurements were carried out using scanning combined with electron microscope а grating monochromator (SEM-CL).

CL halos were observed in all samples after He⁺ ion implantation while no feature of radiation-damage was detected in SEM image (Fig. 1a) nor in optical observation under a polarized microscope. Figure 1 shows SEM and SEM-CL images of the sample irradiated by He⁺ ion with a dose of 3.544×10^{-4} C/cm². CL halos were recognized in SEM-CL image as a luminescent band with a width of about 14 µm paralleled to the irradiated surface (Fig. 1b). The width of CL halo is consistent with theoretical range of α -particles from disintegration of ²³⁸U in quartz¹ and agrees closely with that previously observed in synthetic quartz after He⁺ ion implantation under the same condition with a CL microscope, LUMINOSCOPE ².

CL spectra with two peaks around at 400 nm (blue region) and 650 nm (red region) were obtained from the halo area at room temperature. These emissions might be assigned to $[AIO^4/M^+]$ center and nonbridging oxygen hole center (NBOHC), respectively. Further addition of He⁺ ion irradiation results in an increase in CL intensity of blue spectral peak whereas the intensity in red region is unchanged.

CL line analysis with panchromatic CL mode was carried out along the line X-Y shown on SEM-CL image (Fig. 1b) at 200 points in the range of 35 μ m at 1 point per second. The CL emission within the halo band gradually increased from the implantation surface to a depth of 14 μ m (Fig. 1c). Regardless of He⁺ ion dose, this feature of CL halos was almost same in all samples, whereas the brightness of CL halo emission increased with increase of He⁺ ion dose. The increase of He⁺ ion dose leads to an enhancement of the overall intensity of CL halo area as similar as shown in CL imaging. The distribution pattern of CL intensity as shown in Fig. 1c looks similar with the

Bragg's curve, which shows specific energy loss related to specific ionization along the track of a charged particle. This pattern of CL intensity is consistent with that previously obtained in the study of synthetic quartz using CL CCD camera method $^{2)}$.

The He⁺ ion dose is related to integrated CL intensity obtained by subtracting the intensity of host material from intensity of CL halo area (shaded area in Fig. 1c). The integrated CL intensity increases with dose density, indicating that the CL intensity is a function of the population of the lattice defects created by He⁺ ion implantation. The CL halos are expected to be used for the dosimetry, and could be applied to the dating using radiation-damage in quartz.

- 1) Owen M.R., Geology 16 (1988) 529-532.
- Komuro K., Horikawa Y. and Toyoda S., Min. Petrol. 76 (2002) 261-266.



Fig. 1 Secondary electron (a) and SEM-CL (b) images of natural quartz after He⁺ implantation with 3.544×10^{-4} C/cm². Scale bar indicates 10 µm. The change of CL intensity obtained by CL line analysis via implantation depth from the surface to the inside (c).

4-21 Ion Beam Analysis of Epitaxial Thin Films Treated with Centrifugal Force

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The mechanical responses to the applied stress in nano-sized systems can be different from those in bulky ones because the motion of transporting careers is confined by the limited scale. In amorphous Si films bombarded with energetic ions, the lateral, viscous flow was induced through the quasi one dimensional motion of atom strings¹⁾. In polycrystalline Cu films, the unusual "parallel glide" was observed when the thermal stress was applied²⁾. Now, it is inevitable to perform the systematic study under the well-prepared experimental conditions.

In the present study, the compression tests employing the centrifugal method were made on the Cu-based epitaxial films to understand the mass-transport mechanism specific in the thin films. The depth-sensitive information of the tested samples was obtained with the Rutherford Backscattering spectroscopy (RBS).

epitaxial "sandwich-structured" films The of Cu(200 nm)/Au-Cu(20 nm)/Cu(200 nm) were prepared at 350 °C under the UHV conditions. The Au/Cu ratio at the mid-layer was controlled to be less than 1/9 not to form the ordered alloys. The centrifugal treatments of the epitaxial films were made at 610 kG (1 G = 9.8 m/s²) as a function of time without the temperature rise. The RBS/ <111> axial channeling analysis was made to the obtain depth-distribution information of relevant elements and lattice disorders.

Figure 1 shows a set of 2.7-MeV ⁴He⁺ RBS spectra from the "sandwich-structured" films before(-)and after the centrifugal treatments at 610kG for $15\min(\bullet)$, $30\min(\bullet)$ and 45 min.(•), respectively. In the initial treatment for 15 min., the film thickness became smaller by amount of ~5% but the further treatment for 30 min. resulted in some amounts of recovery in the film thickness. After 45 min. treatment, the film thickness recovered to the original one completely.



Most of the changes in thickness are suffered on the top Cu-layer judging from the corresponding shifts of Au peaks without the significant changes of distribution features.

The axial channeling alignments of the sandwich-structured films by detecting the planar channeling dips show the 6-hold symmetry, suggesting the coherently twinned growth along Cu <111>. Based on the detailed analyses of RBS/channeling data and X-ray diffraction data, the followings can be concluded:

1) The minimum yield at the top Cu layer is kept to be around 6%, which is good enough to assess the possible changes of crystal quality after the centrifugal treatments. The lower aligned fraction and the sharp distribution features at the mid Cu-Au layer also contribute to the easy assessment of the crystal quality change and/or the possible atomistic transport.

2) The application of the centrifugal forces (especially 15 min.) induces the apparent plastic deformation by around 5% that is supposed to be beyond the elastic limit, but judging from the aligned spectra,(not shown), it is difficult to find out the evidence of dislocation multiplication in the corresponding aligned spectra. It can be speculated that the centrifugal treatments may induce the quasi-elastic behavior reflecting the natures of twinned thin films.

3) The comparison among the aligned Au spectra normalized to the random ones, "aligned fraction", implies the systematic crystal-quality change by the centrifugal treatments. Before the treatment, "aligned fraction" is not uniform along the depth where the interface between the mid-Au-Cu layer and the top Cu layer is more disordered, but after the centrifugal treatment for 45 min., "aligned fraction" becomes uniform along the depth. This result can be explained by the strain relaxation associated with the quasi-elastic behavior in the "sandwich-structured" films by the centrifugal treatments.

The further crystallographic approach of the mechanical responses may provide us the conclusive results.

- T. van Dillen, A. Polman, P. R. Onck and E. van der Giessen, Phys. Rev. B<u>71</u> (2005) 024103.1 – 024103.12.
- H. S. Park, K. Gall and J. A. Zimmerman, Phys. Rev. Lett. <u>95</u> (2005) 255504-1 –255504-4.
- Fig. 1 RBS spectra from the "sandwich-structured" films under the random condition are illustrated semi-logarithmically, where the contribution from an α -Al₂O₃ substrate is not included for the simplicity.

4-22 X-ray Diffraction Study in Oxide Thin Film Irradiated with MeV Electrons

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One of the well-known standards for describing radiation damage density in solids irradiated with high energy (keV-MeV) particles is called dpa (displacements per atom). Radiation damage created by different radiation conditions can be compared respectively with the common standard, if dpa is adopted. Especially when the target material is a simple metal, this evaluation method is widely used. This is because relatively simple defects are created in an irradiated simple metal which consists of one atom species. Moreover, elastic displacement is the only origin of the defect formation for simple metal, and this facilitates the estimation of the dpa value.

However, it is not established for oxide ceramics materials that this method can be simply applied for the estimation of damage density. For oxide ceramics, the method for experimentally evaluating radiation damage has to be re-examined in the first place.

In this study 2.5 MeV electrons were irradiated to CeO₂ thin film at room temperature. For evaluating radiation damage in oxide sample, X-ray diffraction method is adopted because of the following reasons. The first reason is that the radiation damage can be quantified by the change in the intensity of diffraction peak as it reflects the damage created in crystal lattice structure. The second reason is that the atomistic defects, for example Frenkel pairs, can be detected by X-ray diffraction method, while microscopic observation method, such as transmission electron microscope, can hardly detect independent Frenkel pairs. The third reason is that X-ray diffraction method is applicable to all crystalline samples including electrically insulating materials, while electrical measurements cannot be applied to insulating materials. The energy of 2.5 MeV is adopted, since in this energy range the irradiation with electrons is known to cause elastic displacements of target atoms, and relatively simple defects are created.

Thin film of CeO_2 was irradiated with 2.5 MeV electron beam. The thickness of the films was about 0.3 µm which is thin enough for the electrons to pass through the film. The irradiations were performed at room temperature, and the X-ray diffraction pattern was measured before and after the irradiation.

In Fig.1 the XRD peaks corresponding to (002) reflection before and after the irradiation are respectively shown. Three data for unirradiated films are shown to demonstrate error originated from sample dependence. The first finding is that the intensity of the peak drastically decreases after the irradiation. This suggests that crystalline structure is disordered due to the

irradiation-induced point defects. Although the point defects are prone to annihilate by thermal energy, they survive even after the irradiation at room temperature. The second finding is that peak position hardly changes after the irradiation. This is clearly demonstrated also in Fig.2, where normalized XRD profiles are shown for the purpose of comparing the shape and position of the peaks. From this figure, it is found that the shape and position of the peaks hardly change after the irradiation. One of the possible interpretations of above results is as follows. Disordered region and undisordered region are clearly separated each other in the irradiated sample. The former region contributes to decrease in peak intensity, but does not contribute to change in the peak shape and position. The latter region is survived undamaged, and thus the shape and position of the peak is unchanged.



Fig. 1 XRD peaks corresponding to (002) reflection for unirradiated and irradiated CeO₂ thin films.



Fig. 2 XRD peaks corresponding to (002) reflection normalized by the maximum value of the peaks for unirradiated and irradiated CeO₂ thin films.

4-23 Characterization of Defects by the Helium and Hydrogen Implantation Using a Slow Positron Beam

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Behaviour of defects introduced by ion beam implantation to silicon carbide (SiC) has been investigated by positron annihilation spectroscopy¹). It is known that quantum structures, such as microvoids or bubbles, will be formed by high-dose implantation²). However, the interaction between positrons and quantum structures are not clarified in detail. We attempted to study the fundamental aspects of interaction between positrons and defects in SiC induced by implantation of hydrogen and helium ions.

Samples were formed by helium and hydrogen implantation to the *n*-type 4H-SiC substrate with the energy of $50 \sim 200$ keV to dose of 1×10^{17} cm⁻². High temperature implantation was carried out for avoiding amorphization. After implantation, the samples were annealed in an argon ambient at 100~1400 °C. Doppler-broadening of annihilation quanta was measured as a function of incident positron energy (*E*). The obtained Doppler broadening spectra were characterized by S-parameter which increases when positrons are trapped at vacancy defects. Also, S parameter tends to increase with increasing the size of defects. All the S parameters were normalized to the bulk value.

Figure 1 shows the energy dependence of S parameters of the helium implanted SiC for the various annealing temperatures. S parameter changes mainly at E=3~15 keV. This corresponds to the ion implantation region. Figure 2(a)shows the annealing behavior of the average S parameter at $E=5\sim10$ keV. The annealing process can be divided into three stages: (1) the decrease at 0- 600 °C; (2) drastically increase at 1000 °C and (3) recovery above 1400 °C. The stage (1) is interpreted as the trapping of helium ions to defects. The stage (2) corresponds to the microvoid formation by the agglomeration of small defects. Microvoid formation was also confirmed by the cross-sectional transmission electron microscope (TEM) observation (Fig. 3). The spherical structures with approximately 10 nm diameters were observed. This contrast corresponds to the microvoids. At the stage (3), S parameter does not decrease to the bulk value. This shows that microvoids do not disappear by high temperature annealing over 1400 °C due to the highly thermal stability. As mentioned above, positrons can interact with the defects introduced by the helium irradiation.

The annealing behavior of S parameter by the hydrogen implantation is also shown in Figure 2(b). S parameter decreases up to 600 °C as well as the case of the helium implantation. S parameter increases drastically at 800 °C and then decreases at 1000 °C. This corresponds to the formation and disappearance of microvoids. In addition, S parameter increases again with the increases of annealing temperature. This is because of flaking off the sample by the hydrogen implantation. Changes of S parameters are overall small unlike the case of helium implantation. This might be because the defect size becomes small due to the adsorption of the hydrogen on the surface of microvoids. Microvoid formation by hydrogen implantation is quite different from the case of the helium implantation.

In summary, defects in SiC formed by helium and hydrogen implantation were probed by positron annihilation spectroscopy. Clear differences of the microvoid formation by the ion species were observed.

References

- 1) A. Kawasuso et. al. Physica B 308-310(2001) 660.
- 2) C.H. Zhang et. al. J. Appl. Phys. 94(2003) 6017.



Fig. 1 S parameters for the bulk SiC and helium implanted SiC. *S* parameters are normalized to these for bulk SiC.



Fig. 2 Average S parameters at $E=5\sim10$ keV for the each annealing temperature.



Fig. 3 The cross-sectional TEM image of ion implanted region for helium implanted SiC.

4-24 Positron Energy Loss Spectroscopy in Reflection High-Energy Positron Diffraction

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When charged particles are incident on materials, various energy loss processes such as plasmon, phonon, and core-level electron excitations occur. In the case of the electron beam at grazing incidence, the surface plasmon excitation is known as a dominant process for the energy loss. According to the theoretical study¹, it was found that mean excitation number of the surface plasmon is proportional to $1/\sin\theta$ (θ : glancing angle of the incident beam) and the loss peak intensities are governed by Poisson distribution. Recently, by using reflection high-energy electron diffraction (RHEED), the excitation process of the surface plasmon has been extensively investigated²⁻⁴). On the other hand, the studies of the energy loss process by positrons are limited as compared to the studies using the electron beam.

The positrons are the antimatter of electrons, having the same mass as the electrons and positive charge. Owing to the positive charge, the total reflection for positrons takes place at grazing incidence⁵⁻⁶⁾. Under the total reflection condition, the energy loss process by the positron beam is considered to be different from that by electrons. In this study, we developed an energy analyzer for the reflection high-energy positron diffraction (RHEPD) and measured the energy loss spectrum of the intensities of the totally reflected spot from the Si(111)-7×7 surface.

The energy analyzer developed in this study is a retarding field type and composed of cylindrical lens and



Fig. 1 Energy loss spectrum of the intensity of the specular spot from the Si(111)-7×7 surface in the RHEPD under the total reflection condition.

retarding grid mesh⁷⁾. The positron beam was generated with a ²²Na positron source and tungsten moderator⁸⁾. The diffraction patterns passing through the energy analyzer were enhanced using a micro-channel plate (MCP) with a phosphor plane and taken by a charge coupled device (CCD) camera. The accelerating voltage of the positron beam was set at 7 kV. The retarding voltage was changed from 6975 to 7005 V at a step of 1 V.

Samples were cut from a mirror polished *n*-type Si(111) wafer with a resisitivity of 1-10 Ω cm. They were annealed at 400 °C over night and flashed at 1200 °C in a few seconds several times to produce the 7×7 reconstruction.

Figure 1 shows the energy loss spectrum of the specular spot intensities from the Si(111)-7×7 surface. The glancing angle is 2.2°, which satisfies the total reflection condition. The incident azimuth corresponds to 7.5° away from the $[11\overline{2}]$ direction. In this energy loss region, two prominent loss peaks are identified at around 10 and 20 V. Since the surface plasmon energy for the Si is estimated to be approximately 11 eV, these peaks correspond to single (denoted as SP1) and double surface plasmon losses (denoted as SP2), respectively. The peak intensity of the double surface plasmon loss is larger than the single one. Thus, the mean excitation number of the surface plasmon is larger than 1. Moreover, these loss peak intensities are much larger than the zero loss (elastic scattering) one. Therefore, totally reflected positrons mostly lose the energy due to the surface plasmon excitations.

In conclusion, the most of the totally reflected positrons excite the surface plasmons near the crystal surface.

- A. A. Lucas and M. Šunjić, Phys. Rev. Lett. 26 (1971) 229.
- Y. Horio, Y. Hashimoto, K. Shiba, and A. Ichimiya, Jpn. J. Appl. Phys. 34 (1995) 5869.
- H. Nakahara, T. Hishida, and A. Ichimiya, Appl. Suf. Sci. 212-213 (2003) 157.
- Y. Tanishiro, Hyomen Kagaku (Journal of The Surface Science Society of Japan) 24 (2003) 166.
- A. Kawasuso and S. Okada, Phys. Rev. Lett. 81 (1998) 2695.
- 6) A. Ichimiya, Solid State Phenom. 28&29 (1992) 143.
- P. Staib, W. Tappe, and J. P. Contour, J. Cryst. Growth 201-202 (1999) 45.
- A. Kawasuso, T. Ishimoto, M. Maekawa, Y. Fukaya, K. Hayashi, and A. Ichimiya, Rev. Sci. Instrum. 75 (2004) 4585.

4-25 Structure Analysis of Sn/Ge(111) Surface Studied by Reflection High-Energy Positron Diffraction

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The Ge(111)- $\sqrt{3} \times \sqrt{3}$ -Sn surface is formed by depositing 1/3 monolayer (ML) Sn atoms on a Ge(111)-c2×8 surface. The surface shows a $\sqrt{3} \times \sqrt{3}$ structure at room temperature, while below 220 K the surface undergoes the phase transition to 3×3 structure. This change of the surface structure has been described as a charge density wave transition¹⁾. Then, this transition was proposed to be thermally fluctuated rippled structure by angle-resolved photoemission spectroscopy²⁾. In 2006, Cortés et al. found that the 3×3 structure reverts to a new $\sqrt{3} \times \sqrt{3}$ structure below 30 K^{3} . They reported that the angle-resolved photoemission spectroscopy experiments show that, concomitantly with the structural phase transition, a metal-insulator transition takes place³⁾. The new $\sqrt{3} \times \sqrt{3}$ ground state is interpreted as a Mott insulator³⁾. However, the mechanism of the phase transition and the displacement of Sn atoms are not fully clarified. In this study, on the basis of the rocking curve analyses using reflection high-energy positron diffraction (RHEPD), we will report on the atomic displacement due to the phase transition.

Experiments were carried out in an ultra-high vacuum (UHV) chamber equipped with a positron source of ²²Na and magnetic deflection lenses. The energy of the positron beam was set at 10 keV. The rocking curves were obtained by changing the glancing angle of the incident beam from 0.7° to 6.0° with an interval of 0.1° . The azimuthal angle is 7.5°-off oriented from the $[11\overline{2}]$ direction. This is called the one-beam condition. In the one-beam condition, the influence of simultaneous reflections parallel to the surface is sufficiently suppressed. Hence, the rocking curve depends only on the atomic positions normal to the surface.

The sample was cut from a mirror-polished n-type Ge(111) wafer (~35 Ω cm). A clean Ge(111)-c2×8 surface was obtained by repeated sputtering (Ar⁺, 0.8 kV) and annealing cycles (800 K, 10 min) in an UHV chamber at a base pressure of 10⁻³ Pa. The formation of the Ge(111)-c2×8 surface was confirmed by reflection high-energy electron diffraction (RHEED). The Ge(111)- $\sqrt{3} \times \sqrt{3}$ -Sn surface was prepared by the deposition of 1/3 ML of Sn on a clean Ge(111)-c2×8 surface kept at 500 K using the electron beam evaporator. After the deposition, the surface was annealed at 500 K for 1 min so as to obtain sharp $\sqrt{3} \times \sqrt{3}$ pattern in RHEED.

Figure 1 displays the rocking curves of specular spots measured from the $\sqrt{3} \times \sqrt{3}$ (293 K) and 3×3 (110 K, 29 K) surfaces. The intense (111) Bragg peak and the distinct dip in the total reflection region can be seen in the curve. The profiles of the rocking curves do not change with temperature (293 K and 110 K). We found that the vertical

positions of the Sn atoms do not change during the phase transition⁴⁾. On the other hand, there are slight differences between the curves at 110 K and 29 K. The positions of the dip and the (111) Bragg peak shift to the low glancing angle. Moreover, the shoulder of the (111) Bragg peak becomes wider. Therefore, we found that the $3\times3-\sqrt{3}\times\sqrt{3}$ phase transition at 30 K occurs accompanied with their differences. However, the diffraction pattern showed still 3×3 structure at 29 K.

We also measured the temperature dependence of the totally reflected RHEPD intensity. The (0 0) spot intensity gradually decreases with increasing temperature for the $\sqrt{3} \times \sqrt{3}$ phase. In the $\sqrt{3} \times \sqrt{3}$ phase, the temperature dependence of the intensity is simply described as the thermal vibration. However, in the 3×3 phase, the spot intensity steeply increases with increasing temperature. We found that this extraordinary change arises from the softening of the phonon mode at Sn atoms. Therefore, the experimental result suggests that the order-disorder phase transition with the softening of the surface phonon takes place at 220 K. We consider that the softening of the surface phonon is closely related with the $3 \times 3 - \sqrt{3} \times \sqrt{3}$ phase transition at 30 K.

- 1) J.M. Carpinelli et al., Nature 381 (1996) 398.
- 2) R.I.G. Uhrberg et al., Phys. Rev. Lett. 81 (1998) 2108.
- 3) R. Cortés et al., Phys. Rev. Lett. 96 (2006) 126103.
- 4) Y. Fukaya et al., e-J. Surf. Sci. Nanotech. 4, 435 (2006).



Fig. 1 RHEPD rocking curves for the Sn/Ge(111) surface under the one-beam condition.

4-26 LET Effect on Irradiation of Hydroxymaleimide in N₂-saturated 2-Propanol (1)

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Hydroxymaleimide was irradiated in N_2 -saturated 2-propanol with 350 MeV Ne-ion, 100 and 50 MeV He-ion beams from the AVF cyclotron in the TIARA Facility. The apparent *G*-values of reduction of hydroxymaleimide obtained in N_2 -saturated system were 3-5 times larger than those in air-saturated system. The dose rate effect, that is, the degradation efficiency of irradiation at 2 nA was 1.6 times larger than that at 20 nA, was also found in this system.

<はじめに> マレイミド(C₂H₂(CO)₂NH)は、光化学 重合反応の開始剤として研究が行われてきた。一方、 ヒドロキシマレイミド(C₂H₂(CO)₂NOH)を窒素置換し た2-プロパノールおよびメタノール中でy線照射した ところ、溶媒分子のラジカルが付加した化合物やダイ マーが生成することがわかった¹⁾。また、ヒドロキシ マレイミドを含む空気飽和状態の2-プロパノール溶液 をイオン照射したところ、ヒドロキシマレイミド分解 の微分G値のLET依存性の傾向は、Heイオン照射 において、重イオン照射と比較して大きくずれること がわかった²⁾。今回は、窒素置換した2-プロパノール 溶媒中でヒドロキシマレイミドのイオン照射を行い、 LET効果について空気飽和状態での照射と比較検討 する。

<実験方法> ヒドロキシマレイミドを2-プロパノー ルに溶かして(0.1モル%)窒素置換し、窒素雰囲気 中で調製した試料を、原子力機構TIARA施設内AVFサ イクロトンのNeイオン(350 MeV)およびHeイオン (100 MeV, 50 MeV)を照射した。また、試料の上にア ルミ箔を置き、入射イオンのエネルギーを変化させ、 LET依存性を調べた。生成物およびヒドロキシマレ イミドは、液体クロマトグラフ(カラムODS、移動 相アセトニトリル:水=50:50)で分析した。分析試 料は、真空乾燥した残留物をアセトニトリルで溶解し たものを用いた。

<結果および考察> 吸収線量あたりのヒドロキシマ レイミドの減少割合から分解効率を求めた。得られた 分解効率をイオンの入射エネルギーあたりで規格化す ると、見かけのG値が得られる。Fig. 1-2にHeイオン 照射およびNeイオン照射における初期エネルギーに 対する見かけのG値をプロットする。空気飽和の系と 同様、初期エネルギーの増加に伴いG値は増加した。 窒素飽和の系でのG値は、空気飽和の系でのG値の3 ~5倍程度であった。 γ線照射における窒素飽和での 増倍率は2.5であり¹⁾、イオン照射では、酸素由来の活 性種による溶媒ラジカル付加反応の阻害効果が γ線照 射より大きいと推測される。

Heイオン(100 MeV)照射において、電流値を変化させて線量率効果について検討した。電流値20 nAおよび2 nAでの線量に対するヒドロキシマレイミドの濃度の減少をFig. 3にプロットする。線量率の低い方が分

解効率が高く、分解のG値は、2 nA照射では20 nA照 射の1.6倍であった。この傾向は、他のイオン種にお いても確認された。線量率効果はy線照射では認めら れず、イオン照射において、溶媒ラジカルの付加反応 と拡散・再結合反応との競争があるものと考えられる。



Fig. 1 Apparent *G*-value depending on the incident energy of He-ion.



Fig. 2 Apparent *G*-value depending on the incident energy of Ne-ion.



Fig. 3 Dose rate effect of degradation efficiency of hydroxymaleimide by He-ion irradiation in N_2 -saturated 2-proapnol solution.

- 1) S. Nakagawa, Radiat. Phys. Chem., accepted.
- 2) S. Nakagawa et al., Radiat. Phys. Chem., accepted.

4-27 Heavy Ion Pulse Radiolysis System Synchronized with AVF Cyclotron

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Introduction

High energy heavy ions have the unique irradiation effects, and are used for the various basic and application studies in chemical and biological fields. The irradiation effects are resulted in the high density and non-homogeneous distributions of initial transient species and subsequent reactions in track. Hydroxyl(OH) radical is the most important radical for oxidations in water. In a previous paper^{1,2)}, we estimated the formation yield of the OH radicals depending on the energy and mass of the incident ions, and on the elapsed time under the 18 MeV/u-energy heavy ion irradiations by the product analysis method. Time-resolved spectroscopy is a very effective method for observing chemical reactions directly under heavy ion irradiations. We constructed a highly sensitive transient absorption measurement system to investigate the effects of the heavy ion irradiations on chemical and biological substances.

Experimental

The heavy ion beam originated from an ECR ion source was a direct current, and pulsed by a beam chopper as shown in Fig.1. The time-width of the pulsed ion beam was controlled arbitrarily in the time range from ns to s by a pulse generator, which was synchronized with an AVF cyclotron. The pulsed ion beam then was accelerated by the cyclotron to a desired energy. Pulse width and electric charge contained in each ion pulse were estimated by measuring the output of a Faraday cup directly by an oscilloscope. The profile of the ion pulse was also monitored by the measurement of the 330-nm fluorescence from a plastic scintillator, which was put on the same position of the sample cell, by a photomultiplier tube assembly. A laser diode and Xe lamp were used as probe light sources. The probe light was introduced to the sample cell at about 30 degree against the axis of the pulse ion beam, and detected by a Si photodiode.

KSCN was chosen as a standard sample for the evaluation of the specification of the constructed transient absorption measurement system, because the rate constant of KSCN with OH radicals are large and formed radical, $(SCN)_2^-$, has the large molar extinction coefficient. The aqueous KSCN solutions were put into a stainless steal irradiation cell, which has a square structure of 20×2 mm. The cell has glass windows of 50-µm thickness at the top and bottom for the heavy ion irradiation and the optical measurements. This sample depth is sufficiently larger than



Fig.1 Schematic presentation of the time-resolved optical measurement system under the pulsed heavy ion irradiation synchronized with the AVF cyclotron.

the penetration range of the ions used.

Results and discussion

Basically, the ion beam from the cyclotron consists of the fine structure pulses with a few ns pulse width at a high frequency of 15-20 MHz. When the optical system had been not synchronized with the cyclotron, each fine pulse could not be observed and the time-jitter was larger than 100 ns. The time-jitter was, however, improved to a few ns by the synchronization and each fine pulse could be observed. The optical absorbance (OD) of $(SCN)_2^-$ produced in the aqueous solution was estimated as follows,

$$OD = \log(I_0/I)$$
(1)

where I_0 and I are optical intensity before and after the irradiation. A quite small absorbance measurement less than 10^{-4} was achieved by taking the differential of the probe light intensity before and after the pulsed ion irradiation. Four kinds of the ions, H (20 MeV/u), He (12.5 MeV/u), C (18.3 MeV/u) and Ne ion (17.5 MeV/u), are possible for the pulsed ion irradiation and transient absorption measurement at the present stage. The wavelength range, which can be observed, is 400 to 740 nm.

- 1) M. Taguchi and T. Kojima, Radiat. Res., 163 (2005) 455-461.
- M. Taguchi and T. Kojima, Nuclear Science and Techniques, 18 (2007) 35-38.

4-28 Extension of the Heavy Ion Beam Pulse Radiolysis Using Scintillators

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An ion-beam pulse radiolysis system was developing for elucidation of the initial process of ion-beam induced chemical reaction. Our unique measurement method was developing using a scintillator. An analysis light-source and excitation are arranged on a track of the ion-beam linearly strictly. Time synchronization between the excitation and the analysis light are also strictly same. The ion pulse radiolysis can realize high spatial resolution and high time resolution by our methods. In this study, we studied at the extension of the wavelength, extension of the time-domain, and extension in the LET-domain, of the heavy ion pulse radiolysis.

近年、加速器の発展により重イオンビームは様々な 分野で応用されている。重イオンビームは高LET放射 線であり、物質中を直進してある深さで大きなエネル ギーを与えて停止する。その結果、物質を局所的に高 密度に励起・イオン化できる。このイオンビームに特 徴的な効果が、物質改変、遺伝子切断による品種改良、 重粒子線がん治療などへ応用されている。応用の発展 にはイオンビーム誘起反応の機構を解明する必要があ る。しかし、基礎過程は完全には理解されていない。 パルスラジオリシスでは、物質にビームを照射し生成 した活性種を直接測定するが、イオンビームの場合、 物質中での短飛程、小ビーム強度、励起構造の特異性 等から、困難であった。我々はこれまで、固体シンチ レーターを用い、イオンビームがシンチレーターを透 過した時の発光を分析光として、イオンビームが試料 中で停止する時の高密度励起による活性種の光吸収を 測定する方法を提案してきた。この方法は、分析光源 と励起源が厳密にイオンビームの飛跡上に直線的に配 置され、時間同期も厳密であるから、時間空間的に高 分解能のイオンパルスラジオリシスを実現できる。-方、シンチレーターを使用すると発光波長と寿命がほ ぼ限定されるために測定に適した試料と活性種が限ら れるという問題点がある。イオンビームによるシンチ レーターの発光特性が明確でない事も問題点である。 本研究では、シンチレーターを利用した重イオンパル スラジオリシスの、波長領域の拡張、時間領域の拡張、 LET領域の拡張を目指した発展研究を行っている。本 研究では、サイクロトロンからの高速重イオンビーム はチョッパーで間引かれ、真空窓と空気層を通過し、 試料上に設置されたシンチレーターを通過した後に試 料に入射する系である。分析光とするシンチレーター の発光を、光ファイバーを通して光測定系に導いた。 フォトマルのシグナルを直接オシロスコープで測定し た。シンチレーターには、波長領域・時間領域の拡張 のために、CaF₂(Eu)(940 ns,435 nm),CsI(16 ns,315 nm), NE-102(2.4 ns,423 nm) (カタログ値)を用いた。各種シン チレーターの電子線励起発光スペクトルをFig. 1に示 した。これまでのCaF₂(Eu)と比べて、長波長側、短波 長側への拡張ができた。サイクロトロンからの重イオ



Fig. 1 Luminescence spectra of scintillators by electron beam.

ンビームは、20 MHz繰り返しであるため、ns-µs程度の 長寿命の反応活性種を測定するためには、パルスを間 引く必要がある。これまでSチョッパーを用いて、隣接 するパルスを1/5に間引いていたが、繰り返し周波数が 高すぎた。今回100 MeV-HeイオンをNE-102シンチレー ターに照射し、Pチョッパーを高度に制御することで、 準シングルパルスを作り出すことに成功した。その結 果をFig. 2に示した。測定システムの時間分解能は5 ns 程度であった。準シングルパルスの発生、発光波長・ 寿命の異なるシンチレーターの使用により、時間領 域・波長領域の測定性能が大幅に拡張された。



Fig. 2 Luminescence of the plastic-scintillator induced by semi-single shot 100 MeV He²⁺ ion beam.

4-29 Formation of Organic Compounds in Simulated Extraterrestrial Environments by High-Energy Particles

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We verified the possible formation of organic compounds in simulated extraterrestrial environment (interstellar dust particle and Titan atmosphere) and simulated primitive Earth atmosphere by irradiation of protons from the TIARA tandem accelerator and cyclotron. TEM observation of the tandem products showed that the structure consisted of globules was formed. Amino acids were detected after hydrolysis of all the products. It was shown that the large energy deposit made more amino acids. G-value of Glycine from CH₃OH-NH₃-H₂O by protons from the cyclotron was lower than heavy ions.

近年隕石や彗星など地球圏外に様々な有機物が検出 され、それらが原始地球に運び込まれ生命の素材と なった可能性が議論されている。検出される「複雑有 機物」は加水分解により、アミノ酸等生体関連物質を 生成することが知られているが、化学構造など未解明 の部分が多い。

本研究では模擬星間物質へ宇宙線の主成分である陽 子線を照射し、放医研HIMACで行なった重粒子線照 射実験と比較することで、アミノ酸生成量の線種によ る比較を行うことを目的とした。また模擬原始大気や 模擬タイタン大気に対して陽子線照射を行い、生成す る「複雑有機物」の構造やアミノ酸生成について明ら かにすることを目的とした。

Experimental

a: タンデム加速器

容量約400 mLのPyrex製容器に、模擬原始大気とし て一酸化炭素と窒素を350 Torrずつに水を5 mL、模擬 タイタン大気としてメタン5%窒素95%の混合ガスを 各700 Torr封入したものを作製し、タンデム型加速器 で3 MeV 陽子線を300 nAで60 min照射した。またイ オンビーム取り込みの窓としてカプトン箔(7.5 μm) を用いた。更に実験では発光分析を行い、模擬原始大 気試料についてはTEMによる観察を行った。

b: サイクロトロン

本研究で用いた陽子線照射用の容器はチタン製で、 内容積は7.6 mLである。照射口側は厚さ0.1 mmのチタ ン薄膜で蓋をした。各容器にメタノールとアンモニア、 水を1:1:2.8に調整し、7.6 mLずつ封入し、20 MeV, 1 µAの条件で1 minから40 min照射を行った。

作成した各試料に対してゲル濾過クロマトグラ フィー (GFC) による分子量推定を行った。また6 M HCl中で110 ℃, 24時間酸加水分解した後にアミノ酸分 析計 (Shimazu LC-10A) にてアミノ酸の同定・定量を 行い、同様な組成の混合物に放医研HIMACからのHe, Ne, Arビーム照射実験との比較も行った。

Results and Discussion

a: <u>タンデム加速器</u>

発光分析により窒素の励起とイオン化が確認された。 両系の生成物の分子量は数百から2千程度に推定され、 加水分解後にGlyやAlaが検出された。NH3を含む気体 への照射も行ったが、照射中にカプトン箔が破れてし まった。今後他の窓材の利用など照射方法について検 討する。

模擬原始大気試料の照射後のTEMによる観察結果 をFig. 1に示す。粒状の構造体が密集した数百nm程度 の大きさの集合体が観察された。



Fig. 1 TEM image of the complex structures like aggregated globules formed from simulated primitive Earth atmosphere by proton irradiation.

b: サイクロトロン

GFCから生成物の分子量は数百から3千程度と推定 された。また加水分解後のアミノ酸分析によりGlyや Alaなどの種々の生体アミノ酸の他に、無生物的生成 を示す α アミノ酪酸や β -Alaなどの非生体アミノ酸が検 出された。照射時間を伸ばすことによりアミノ酸の生 成量が増加した。生成したグリシンのG値(100 eVあ たりの生成分子数)は10 minの照射で6.8×10⁻⁵となり、 HIMACを用いた重粒子線照射実験の結果と比べ著し く小さい値を示した(G value: H<He<Ne>Ar)が、 γ 線 照射よりは大きい値であった。

4-30 Basic and Application Studies on Chemical Responses to Quantum Beams in Heterogeneous Systems

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We have recently reported on radiation-induced reactions in binary systems that in aqueous solutions recovery of platinum-group elements and non-toxic treatment of asbestos were each accelerated by adding solid oxides to the solution¹), and that the radiolytic behavior of amides in n-dodecane was dependent on aqueous solution contacting with the n-dodecane²). To elucidate their heterogeneous reactions, the primary processes of oxides involved in metal reduction or hydrogen production³) in aqueous solutions, and the LET effect of radiolysis of amides in n-dodecane were further investigated in fiscal 2007.

The samples of aqueous or organic solution systems were irradiated at ambient temperature by using electron beams (initial energy: 0.5-2.0 MeV) and gamma-ray (averaged energy: 1.25 MeV) at JAEA-Takasaki: the dosimetry of sample was made by using CTA-film and/or solution dosimeters. After the irradiation, ions or molecules formed in the liquid part of sample were measured by using spectrophotometry, ICP-AES or liquid chromatography; the gas part such as H_2 by using gas chromatography.

In order to observe the interaction between oxide particles and radiolysis products of water, reductions of dichromate Cr(VI) and ceric ions were studied in aqueous solutions containing colloidal silica. Figure 1 shows the reduction yields of Cr(VI) in the radiolysis of acidic aqueous solution, illustrating that the yield increased with increasing the silica concentration in the absence of t-BuOH, and that at the concentration higher than 7.5 wt% or in the



Fig. 1 Reduction yields of Cr(VI) ion in 0.1 mol/L HClO₄ containing colloidal silica. The yields in the absence of t-BuOH suggested the interaction between the colloidal silica and oxidizing species of water radiolysis.

presence of t-BuOH, it became constant (0.13-0.14 µmol/J).

Based on the facts that the constant value of yield can be explained by the primary yields of water radiolysis and that t-BuOH is ready to react with an oxidizing species of OH to reduce Cr(VI), the results suggested that the addition of colloidal silica brought about the reduction of Cr(VI) *via* the conversion of oxidizing species to reducing one.

We have been also studying H_2 production in aqueous solutions containing oxide particles to find out the effective condition for the production not only in kinds of solutions and oxides³⁾, but also in composition and structure of oxide. In the present study, the H_2 production was measured in acidic aqueous solution containing alumina powders with some crystal structures: α (trigonal corundum structure), γ (hexagonal spinel) or θ (monoclinic).

Figure 2 shows the typical results of production yields as the respective bar graphs, in which the specific surface area are indicated. When compared with the yields in aqueous solutions in the absence of alumina (AKP-50), those in the presence increased: the order was [water] < [0.4 mol/L H_2SO_4] < [water+alumina] < [H_2SO_4+alumina]. The yields for alumina powders with 3 crystal structures (TMD-100D, TMD-300D, TMD-DR) became different from each other, suggesting that the yields of H₂ production were dependent on the structures rather than the specific surface area.

References

- 1) R. Nagaishi et al., JAEA 2007-060 (2008) 161.
- 2) Y. Sugo et al., Radiat. Phys. Chem. 76 (2007) 794.
- 3) R. Yamada et al., Int. J. Hydrogen Ener. 33 (2008) 929.



Fig. 2 H₂ production in 0.4 mol/L H₂SO₄ solution containing various aluminum oxide powders. The production yield was enhanced by adding the powders and was dependent on the crystal structure of oxide.

4-31 Alpha-radiolysis of Organic Extractants for the Separation of Actinides

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For the purpose of reduction in the long-term environmental burden of high-level radioactive waste (HLW) from nuclear fuel reprocessing, we have developed a tridentate diamide. N,N,N',N'-tetraoctyldiglycolamide (TODGA)¹⁾ as one of the most promising extractants for the recovery of long-lived actinides such as americium and curium. In order to evaluate the applicability of TODGA to the process under high radiation exposure, the radiolysis of TODGA was investigated by the irradiation with 60 Co γ -rays and electron pulses in the previous study²⁾. It was obvious that *n*-dodecane, which was used as a diluent, had a sensitization effect on the radiolysis of TODGA. From the pulse radiolysis study, it was concluded that the sensitization effect of n-dodecane was mainly due to a charge transfer reaction from radical cations of n-dodecane to TODGA molecules in the primary process.

In the actual partitioning process of HLW, the organic extractants will be exposed to α -particles emitted by α -decay of the actinides. The common energies of the α -particles are approximately 5 MeV. Since the energy deposition of α -particles is quite different from that of γ -rays, it is essential to investigate α -radiolysis of the organic extractants. In contrast to the irradiation using an actinide radionuclide as an α -particles emitter, the irradiation with α -particles provided by an accelerator can be carried out in a reasonable timescale and without contamination with radionuclides.

In this study, α -radiolysis of TODGA in *n*-dodecane was investigated by the irradiation with helium ions accelerated by the AVF cyclotron using the HY1 irradiation apparatus in the TIARA facility. The solution of TODGA in *n*-dodecane was irradiated with helium ion beam according to the schematic diagram of the irradiation system shown in





Fig. 1. The area of the irradiated surface of the sample was 13.8 cm². The thickness of the sample was larger than the range of the ion beam. The incident energy of ions on the surface of the sample was attenuated by aluminium foils and determined by silicon semiconductor detector. The sample was also irradiated with ⁶⁰Co γ -rays for comparison of the radiation chemical yields between helium ions and γ -rays. The concentration of TODGA before and after irradiation was determined using a capillary gas chromatography equipped with a flame ionization detector.

Yields for the degradation of TODGA in *n*-dodecane by irradiation with helium ions and γ -rays are plotted against dose in Fig. 2. This figure indicates the both degradation yields increase with dose, and the yield by helium ions is not more than that by γ -rays. It was also observed the radiation chemical yield gradually increased with the incident energy of helium ions. These results suggest that the radiolytic degradation of TODGA in *n*-dodecane is reduced in the irradiation with high LET radiations such as α -particles emitted by the actinides.

References

- 1) Y. Sasaki et al., Solv. Extr. Ion Exch. 19 (2001) 91.
- 2) Y. Sugo et al., Radiat. Phys. Chem. 76 (2007) 794.



Fig. 2 Degradation of TODGA by irradiation with helium ions and γ-rays.

Sample: 10 mmol/L TODGA in *n*-dodecane, helium ions: incident energy of 16.8 MeV, fluence rate of 1.25×10^9 ions/cm²/s, γ -rays: dose rate of 4.8 kGy/h.

4-32 Helium Ion Beam Radiolysis of Sodium Bicarbonate Aqueous Solution

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In radioactive waste disposal system, after a failure of waste package, radiolysis of groundwater would occur on the vitrified waste matrix. While hydrogen is relatively inert and dissipated, oxidants like H_2O_2 and O_2 would make the surroundings an oxidizing environmental condition and eventually accelerate radionuclide migration in the near-field due to weaker sorption and higher solubility in oxidizing condition.

Solutes in groundwater would affect radiolysis of water. Typical groundwater contains bicarbonate or chloride as most abundant ions. However their effects on G-values and their reactions have not been systematically studied in the concentration range of typical groundwater composition. Thus we have studied gamma-radiolysis of bicarbonate aqueous solutions and measured concentration of molecular products (H₂O₂, H₂, and O₂) in solution after irradiation to clarify which reactions and rates constants of solutes should be included in modeling the radiolysis of aqueous solutions, since homogeneous reactions of radiolytic products in solutions are common in gamma- and alpha-radiolyses. Their changes upon irradiation time and solute concentration were successfully reproduced by homogeneous kinetic model calculation¹⁾.

In the current safety assessment of disposal system, it is assumed that overpack would be breached after 1000 years after disposal, and alpha decay would be dominant in terms of radiolysis on the waste surface. We have performed the homogeneous model calculation of alpha-radiolysis of those solutions²). In the case of alpha-radiolysis of water, it was predicted that H_2O_2 and H_2 were accumulated at the constant rate, and solutes reduced the accumulation rate of





 H_2O_2 .

To confirm the results of above model calculation on alpha-radiolysis, we have performed helium ion beam irradiation of sodium bicarbonate aqueous solutions, since significantly higher ion flux can be obtained compared to the case of typical alpha emitter (e.g.²⁴¹Am) that reduces irradiation time, and irradiation conditions such as acceleration voltage and ion flux can be controlled. Irradiation was performed with the AVF cyclotron in TIARA (Takasaki Ion Accelerators for Advanced Radiation Application), JAEA. Aerated sodium bicarbonate aqueous solutions were irradiated with ⁴He²⁺ at 20 MeV for 2, 5, and 10 minutes, and H₂O₂ concentrations were measured. As shown in Fig. 1, H₂O₂ concentrations increased almost linearly with time corresponding to dose in all cases. Their rates were reduced by increasing concentration of solutes.

Since the energy of the helium ion is higher than that of typical alpha-particles, *G*-values of water were assumed as shown in Table 1 based on the spur-diffusion model type calculation assuming cylindrical track structure with a LET value of 20 MeV ${}^{4}\text{He}^{2+}$ calculated by the SRIM-2006 code.

Figure 1 also shows calculated values of H_2O_2 concentration in water and sodium bicarbonate solutions by a homogeneous reaction model. A slow reaction of H_2O_2 with HCO₃⁻ was added to the model as lines. Calculated values of H_2O_2 concentrations increased almost linearly with reduced accumulation rates by addition of sodium bicarbonate, and in the case of 0.03% NaHCO₃ the agreement with experimental values is fairly good. Discrepancies at larger accumulated doses might be partly due to dissipation of radiolytic products to gaseous phase.

This study is a part of

the Project for assessment methodology development of chemical effects on geological disposal system funded by the Ministry of Economy, Trade and Industry, Japan.

References

- 1) M. Yamaguchi et al., 1st APSRC, PR22 (2006).
- M. Yamaguchi et al., ASR2007, PA22 (2007).

Species	G-value	
e	0.204	
Η	0.498	
H_2	0.763	
OH	0.458	
H_2O_2	0.832	
H^{+}	0.396	
OH-	0.173	
HO_2	0.027	
O_2	0.019	
O_2	0.013	
0	0.073	

Table 1 Calculated *G*-values of water by 20 MeV ${}^{4}\text{He}^{2+}$.

4-33 Neutron Production Double Differential Cross Sections of ⁹Be Bombarded with 10 MeV Protons and Deuterons

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Neutron production double differential cross sections (DDXs) on beryllium bombarded with low energy protons and deuterons are important as a source term for medical therapy and material irradiation facilities. Nuclear data provided by the ENDF/B-VII files and the calculated data by the PHITS code with the JQMD model are used for the source term calculation. PHITS code¹⁾ (PHITS/JQMD) in which the quantum molecular dynamics and statistic decay model are implemented. successfully generates charged-particle nucleon-induced and induced neutron-production DDXs in the energy region above 100 MeV/u^{2} . The accuracy of the ENDF/B-VII files and the PHITS code should be checked based on systematic experimental data in low energy region. In this report, we describe the experiment and comparisons between experimental data and the PHITS calculation results.

The experiments were carried out with 10 MeV proton and deuteron beams delivered to the HB-1 beam line at the AVF cyclotron facility. The neutron time-of flight (TOF) measurements were performed using NE213 organic liquid scintillators (5.08 cm diameter, 5.08 cm long). The detectors were placed in directions of 15, 30, 45, 60, 75, 90 and 120° with flight path lengths of 2.0 m. The thickness of the beryllium target was 15 μ m.

Neutron production DDXs from proton incidence are shown in Fig. 1. The experimental data showed two peaks corresponding to the ground-state neutrons (n_0) from the

 ${}^{9}\text{Be}(p,n){}^{9}\text{B}$ and a neutron energy group (n_1) which leaves ${}^{9}\text{B}$ in the 2.3 MeV first excited state. Continuum neutrons were also observed because thresholds for several three-body break-up reactions are low and these can give low energy continuum neutrons. Obtained results by ENDF/B-VII have some peaks which were not in experimental data. Figure 2 shows neutron production DDXs for deuteron incidence. The number labeled in this figure indicates the energy levels in ${}^{10}B$ by the ${}^{9}Be(d,n){}^{10}B$. The peak at the highest energy is corresponding to the population of the first few levels in ¹⁰B. These transitions appear to include considerable direct-reaction ("stripping") strength. The PHITS/JQMD reproduced the absolute value of DDXs for deuteron incidence, but it failed to reproduce maximum neutron energy because Q-values for nuclear reaction are not implemented. PHITS/JQMD describes the of distribution time evolution of nucleon many-body-systems in the phase space in the high-energy region, therefore it could not simulate the nuclear levels and low energy physics, correctly.

In future, we will measure neutron spectra from heavy-ion induced reaction at low energy and improve PHITS/JQMD.

- 1) H. Iwase et al., J. Nucl. Sci. Tech. 39 (2002) 1142.
- 2) K. Niita et al., Phys. Rev. C 52 (1995) 2620.



Fig. 1 Neutron production DDXs for the ⁹Be(p,xn) reaction.



Fig. 2 Neutron production DDXs for the ⁹Be(d,xn) reaction.

4-34 Feasibility Study of High Efficiency Neutron-Detection System for Measuring Elastic Neutron-Scattering Cross Sections in Intermediate Energy Region

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The purpose of this study is to develop a high efficiency neutron-detection system to measure the systematic data of the elastic neutron-scattering cross sections in intermediate energy region. It is required a neutron-detection efficiency of about 10^4 times higher than that of the conventional detection systems such as proton-recoil counter-telescope to obtain the sufficient data. We developed a high efficiency neutron-detection system based on a liquid organic scintillator by coupling with time-of-flight (TOF) technique. By using the system, the elastic neutron-scattering cross section was measured to examine its performance.

The experiment was carried out at the LC0 beam course of the TIARA cyclotron with the setup presented in Fig. 1. Quasi-monoenergetic neutrons of 75 MeV¹⁾ were produced by bombarding protons of 80 MeV on a Li target located 5 m upstream from the back end of the collimator. The intensity of the proton beam was set at 1.2 µA during the cross section measurement. A carbon rod whose size is 5.0 cm in diameter and 5.0 cm in thickness was hanged onto neutron beam as the scattering sample. A BC501A liquid organic scintillator was located in the direction of 15 degree away from the beam axis as the neutron detector. Both the diameter and thickness of the scintillator are 12.7 cm. The flight path between the sample and the detector is 400 cm. A shadow block composed of iron was mounted as covering the detector to prevent direct incidences of the neutrons scattered at the exit of the collimator. The size of the shadow block is 50 cm \times 50 cm \times 20 cm.

The experimental data were analyzed off line. Gamma ray events were distinguished from the neutron events by the two-gate integral pulse-shape discrimination. The elastic neutron events were picked out according to the TOF distribution. The fastest neutron events in TOF indicate the neutrons scattered elastically at the sample. Small correction was made based on the theoretical calculation with TALYS code²⁾ to eliminate the intrusion of the inelastic events from low-excitation states that are not able to be separated in TOF measurement due to low time resolution of the detection system. The uncertainty regarding to this intrusion was estimated below 5 % in the present experiment. The absolute value of the cross section was normalized by the incident neutron flux to the sample which measured independently on the beam axis.

Figure 2 shows the measured data of the elastic neutron-scattering cross section together with the values of evaluated nuclear data ENDF/B VII-0³). Our result obtained at 15 degree in laboratory frame agreed very well with the evaluated nuclear data. This indicates that the measurement

and analysis methods are appropriate, and the neutrondetection system developed in the present work is applicable to the measurement of the elastic neutron-scattering cross sections with higher detection efficiency compared with the conventional ones. We will extensively measure the systematic cross sections at the Research Center for Nuclear Physics of Osaka University in the energy region above 100 MeV where there is no available data to evaluate the nuclear data.

This work was partially supported by the Ministry of Education, Science, Sports and Culture by a Grant-in-Aid for Young Scientists (B) (no. 19760616, 2007).

- 1) Y. Shikaze et al., Radia. Prot. Dosim. 126 (2007) 163.
- A. J. Koning et al., "TALYS-1.0 USER MANUAL" (2007).
- M. B. Chadwick et al., Nuclear Data Sheets 107 (2006) 2931.



Fig. 1 Illustration of experimental setup.



Fig. 2 Neutron elastic-scattering cross section for carbon nucleus in 75 MeV.
4-35 Development of Dose Monitoring System Applicable to Various Radiations

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INTRODUCTION

For ensuring radiation safety in nuclear facilities as well as high-energy accelerator facilities, we have been developing a new device of dose monitoring system applicable to various radiations.^{1,2)} The system is composed of (1) a phoswitch-type scintillation detector, which consists of liquid organic scintillator coupled with ZnS(Ag) scintillation sheets doped with ⁶Li, and (2) a data acquisition (DAQ) system for digital analysis of the waveform of the scintillator signals. The system is capable of measuring doses from neutrons, photons and muons with energy ranges from thermal energy to 1 GeV, 150 keV to 100 MeV, and 1 MeV to 100 GeV, respectively.

In the fiscal year of 2007, we installed the function for estimating the energy spectrum from the light-output distribution of the phoswitch detector into this system. For this purpose, we improved the unfolding code MAXED,³⁾ and incorporated it into our developing system. The accuracy of high-energy neutron spectrum estimated by this system was examined in the quasi-monoenergetic neutron field at the LC0 beam course of the TIARA cyclotron.

EXPERIMENT

The experimental setup is depicted in Fig. 1. Quasi-monoenergetic neutrons with peak energy approximately 60 MeV were produced by bombarding protons of 65 MeV on a Li target. The phoswitch-type detector was placed behind the concrete and iron collimators at 12.9 m downstream of the target.

The waveform of the signal from the detector was digitized by the DAQ system, and analyzed by an off-line program. In the analysis, the neutron, photon and muon induced scintillations were distinguished by means of the pulse shape and height discrimination technique.¹⁾ The light-output distribution of neutron-induced scintillations was converted to the neutron energy spectrum, using our improved MAXED code. A simple 1/E spectrum was adopted for the initial guess of the neutron spectrum in the unfolding process.

RESULTS AND DISCUSSION

Figure 2 shows the estimated neutron spectra from the light-output distribution of the phoswitch detector, using our improved MAXED code. It is evident from the graph that the estimated neutron spectrum has a peak structure at energies around 60 MeV, indicating the applicability of our developed system to the neutron spectroscopy for such quasi-monoenergetic neutron fields. This is owing to the sophisticated unfolding algorithm employed in the MAXED code: the maximum entropy method. Note that conventional

unfolding techniques are generally not able to handle such peak structures because of the mathematical difficulty in making the solution convergent.

CONCLUSION

We installed the function for estimating the energy spectrum into our developing dose monitoring system applicable to various radiations. The accuracy of neutron spectrum estimated by this system was well verified in quasi-monoenergetic neutron fields of TIARA whose spectra are generally difficult to be estimated by conventional unfolding techniques.

Present study is the result of "Development of high-performance dose monitor applicable to various radiations" entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

- 1) T. Sato et al., J. Nucl. Sci. Technol. 42 (2005) 768.
- 2) T. Sato et al., Radiat. Prot. Dosim. 126 (2007) 501.
- M. Reginatto *et al.*, Nucl. Instr. and Meth. A476 (2002) 242.







Fig. 2 Measured neutron spectra at the 60 MeV quasi-monoenergetic neutron field of TIARA, using our developed dose monitoring system.

4-36 Evaluation of the Performance of a New Fluence Monitor for Quasi-monoenergetic Neutron Calibration Fields of Several Tens of MeV Range

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High energy neutron standard fields are necessary for calibrating area dosemeters and individual ones used in facilities of high intensity proton accelerator. For the neutron fields above 20 MeV, the neutron standard fields have not been established in Japan. Therefore, the development of the calibration fields and relevant calibration techniques has been in progress by using the quasi-monoenergetic neutron irradiation fields of several tens of MeV at TIARA¹⁾ of Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency.

At TIARA, neutrons induced by the nuclear reactions are transported into an irradiation room through a collimator. However, the existing neutron fluence monitors based on fission chambers are located around the neutron beam line (off-line) and ahead of the collimator. They cannot detect directly neutrons in the irradiation room. They are influenced by unwanted neutrons which are differently produced at every experiment from the beam transport line and from the materials around the monitors. Therefore, a transmission type neutron fluence monitor with a thin plastic scintillator (Fig. 1) has been developed in order to monitor directly the neutrons transported into the irradiation room (on-line). Performance of a prototype of the monitor has been evaluated by using three kinds of quasi-monoenergetic neutron fields with 45, 60 and 75 MeV peaks at TIARA.

The monitor has two photomultiplier tubes (PMTs) which read out the scintillation light yielded in the plastic scintillator $(130 \times 130 \times 0.5 \text{ mm}^3)$ through acrylic light guides attached on both sides of the scintillator. The scintillator and light guides coated by white reflection material are installed in the aluminum case with aluminum entrance windows $(120 \times 120 \text{ mm}^2)$. As for scintillator



Fig. 1 New neutron fluence monitor consists of a plastic scintillator, acrylic light guides, an aluminum case with aluminum entrance windows and two PMTs.

thickness, thicker scintillator has higher detection efficiency, but it causes lower transmission ratio, longer dead time and larger number of scattered neutrons. The scintillator thickness of 0.5 mm was selected so as to make the efficiency at the same level as the existing monitors and the transmission loss within 1 % as a result of the investigation using the MCNPX code.

In the performance tests, the monitor was set on the surface of the collimator exit. Data of individual pulse height for both PMTs and of the sum of them have been taken simultaneously. As linearity of count rate to beam intensity (beam current) was shown in Fig. 2, it was confirmed that the monitor had good linearity to beam intensity at the same level as those of the existing monitors. It was also found that there was good correlation with the count rate of the monitor and those of the existing monitors. However, sensitivity of the monitor was found to be at most 10 % of the existing monitor. Moreover, two-dimensional information of the pulse heights of both PMTs indicated that the monitor did not count the signals from all area of the scintillator in the irradiation field² because of rapid attenuation of the scintillator.

In the future, sensitivity of the fluence monitor will be improved by better light collection from the scintillator, based on direct light collection from the scintillator surfaces without reflection material.

References

- 1) M. Baba et al., Nucl. Instr. Meth. A 428 (1999) 454.
- 2) Y. Shikaze et al., JAEA-Review 2005-001 (2006) 337.



Fig. 2 Count rate of the fluence monitor v.s. beam current for neutrons with 75 MeV peak. Good linearity of the monitor's count rate to beam current was seen.

4-37 Properties of Radiochromic Film Dosimeters for Low Energy Electron Beam

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Introduction

Irradiation with electron beams in the energy range lower than 300 keV is used in the polymer industry for curing and crosslinking. Lately these beams have also been used in the pharmaceutical and medical device industries for sterilization of material surfaces. Radiochromic film dosimeters (RCD) have been used widely both in the industrial processes and research employing gamma-rays and electron beam irradiation¹⁾. These dosimeters have been calibrated commonly only for ⁶⁰Co gamma-rays or MeV electron beams, therefore the feasibility should be studied on the calibration also for low energy electron beams. In this fiscal year, we studied consistency in evaluation of 2-MeV electron dose and ⁶⁰Co gamma-ray dose, using RCD.

Experimental

Two different kinds of radiochromic dosimeters and cellulose triacetate (CTA) dosimeter were used in this study²⁾. The former and latter were independently calibrated for gamma-rays and electrons using an ionizing chamber and a calorimeter, respectively. One RCD is nylon-based FWT60 with thickness of 50 µm, containing hexahydroxyethyl pararosaniline cyanide as the radiation-sensitive component. Another RCD is GEX B3 radiochromic film dosimeter with thickness of 17 µm³⁾. The B3 radiochromic dosimeter contains a radiochromic dye precursor of pararosaniline cyanide in polyvinyl butyral. The gamma-ray irradiation was carried out using a 60Co plaque source (2.6 PBq) at a dose rate of 10 kGy/h, which was measured with the ionization chamber. Irradiation of 2-MeV electron beams was conducted using an accelerator with the 120 cm wide scanning beams at a current of 2 mA with the conveyor speed of 1.73 m/min, ultimately at a dose rate of 10 kGy/pass. Optical absorbance in terms of optical density (O.D) for radiation-induced coloration of dosimeters was measured using Hitachi spectrophotometer model U-3310. Wavelength of 600 nm and 554 nm was used for dose measurement of FWT60 and B3, respectively.

Results and discussion

Figures 1 and 2 show dose response curves of radiochromic film dosimeters, FWT60 and B3, for ⁶⁰Co gamma-rays and electron beams. Dose response value of FWT 60 for electron beam is nearly equal to dose response value for ⁶⁰Co gamma-rays. This result should enable us to achieve highly reliable dosimetry of electron beams on the basis of gamma ray dose reference. But the change in absorbance of B3 caused by electron beam irradiation is

much smaller than those by γ -irradiation. This difference is due to dose rate effect. In the case of electron beam irradiation, radiation-induced chemical reaction is not completed during irradiation. It is necessary to compare the response of B3 under various irradiation conditions to further clarify the dose rate dependence of B3.



Fig. 1 Dose response curves of radiochromic film dosimeter, FWT60, for ⁶⁰Co gamma-rays and electron beams.



Fig. 2 Dose response curves of radiochromic film dosimeter, B3, for ⁶⁰Co gamma-rays and electron beams.

- ISO/ASTM 51275 (2004) Standard Practice for Use of a Radiochromic Film Dosimetry System.
- 2) Z. P-Stuglik et al., Appl. Radiat. Isot. 66 (2008) 346.
- 3) J. H-Hansen et al., Radiat. Phys. Chem. 74 (2005) 341.

4-38 Application of Clear Polymethylmethacrylate (PMMA) Dosimeter Radix W to A Few MeV Electrons

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Introduction

As radiation processing using high-energy electron beams grows, accurate evaluation methods of electron dose become vital to the purpose of quality assurance of irradiation products. Clear PMMA dosimeter (Radix W), which is manufactured based on chemical crosslinkage, was developed to cover wider dynamic gamma ray dose for the dose range of 2 to 150 kGy, employing different readout wavelength of 280 nm and 320 nm for doses up to 10 kGy and higher, respectively¹⁾. Characteristics of Radix W were studied on electron dose response, its stability after irradiation and consistency with gamma-ray dose.

Experimental

Irradiation of 2-MeV electron beams was conducted using an accelerator with the 120 cm wide scanning beams at a current of 2 mA with the conveyor speed of 1.73m/min, ultimately at a dose rate of 10 kGy/pass. Two kinds of dosimeters, Radix W and CTA were used in this study. The physical and chemical characteristics of Radix W (Radia Industry, Co. Ltd) are described in elsewhere²). Dose comparison was performed for 2-MeV electrons employing CTA and Radix W dosimeters which were independently calibrated for 2-MeV electrons and ⁶⁰Co gamma rays using the calorimeter and the ionizing chamber, respectively³).

Results and discussion

Dose given by CTA and Radix dosimeters were compared under the same condition of 2-MeV electron irradiation. The doses estimated by these dosimeters are shown in Fig. 1 as a function of 'nominal dose', which means doses estimated from different irradiation pass numbers assuming at a dose rate of 10 kGy/pass. The ratio of dose given by CTA to dose given by Radix W was within 3.0% (1 σ) for dose range of 10 to 60 kGy. This result should enable us to achieve highly reliable dosimetry for the quality assurance in electron radiation processing on the basis of gamma-ray dose reference. The variation of absorbance within 24 h after irradiation is less than 7% as shown in Fig. 2. This variation of absorbance by 7% is approximately equivalent to absorbed dose value, 1 kGy. The error of 1 kGy can be ignored in real radiation processing, considering the requirement of quality assurance level.



Fig. 1 Comparison of absorbed dose in water estimated by CTA film dosimeters with those estimated by Radix W dosimeters.



Fig. 2 Stability of dose response of Radix W after the end of irradiation.

- 1) H. Seito et al., RADIOISOTOPES. 56(2007) 777.
- 2) M. Takehisa et al., Radiat.Phys.Chem.76(2007) 1619.
- 3) T. Kojima et al., IAEA-SM-365/56, (2003) 216.

4-39 Electroplating of Metal Micro-structures Using a Resist Micro-machined by Proton Beam Writing

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A proton beam writing $(PBW)^{1}$ is a direct writing technique for micro- or nano-meter sized machining using MeV focused proton beams with the size of 1 μ m or less in diameter. The MeV proton beams have long penetration depths of over 50 μ m in a photo resist. The straggling of the proton beams is smaller than that of electron beams. The technique,, thereby, has an advantage of micro-machining of thick resist²). We report the fabrication of Ni structures by electroplating a Ni layer on micro-machined PMMA, a typical resist material by the PBW.

A fabrication process of the Ni structure by electroplating on the resist micromachined using the PBW is shown in Fig. 1. This process involves the following steps. (i) Spin coating on a Si plate as a resist layer with 5 µm thick PMMA. (ii) Exposure by the PBW with 1.7 MeV focused proton beam at TIARA, JAEA. The beam size was estimated to be less than 1 µm in diameter from the secondary electron images of a commercially available Ni mesh. The PBW was carried out with a resolution of approximately 3500×3500 pixels in the area of $300 \times 300 \ \mu\text{m}^2$ by an electrostatic scanner. The fluence of protons was 100 nC/mm². (iii) Development of the PMMA with IPA-water (7:3) for 20 min and rinsed in deionized water. (iv) Deposition of a seed layer of 40 nm thick Au by magnetron sputtering for electroplating on the surface of the micro-structure. (v) Electroplating of the micro-structure using a typical nickel sulfamate bath solution. The bath temperature was kept at 35 °C. The deposition rate of Ni was controlled by the current density of the cathode, that is 40 A/m^2 for the first 50 µm thick Ni, 400 A/m^2 for the next 200 μ m, and 800 A/m² for the last 600 μ m. (vi) Removal of the PMMA structure was made by immersion in acetone and the Ni structure was rinsed in ethanol.

A SEM image obtained from the grid pattern of 5 μ m thick PMMA was shown in Fig. 2. The 0.8 μ m wide trenches with smooth and vertical sidewalls are observed from the image. The verticalness of side walls to the surface of the Si





plate are ascribed to the small lateral straggling of 1.7 MeV protons. The smooth sidewall was achieved by minimizing the effect of beam current fluctuation by the iterated exposure with the same position and with the same scanning pattern. Figure 2(b) shows the SEM image of a grid pattern structure of Ni obtained by electroplating on the structure of Fig. 2(a). The successful fabrication of the Ni structure with the vertical and smooth side walls indicates that the micro-machined PMMA by PBW has adequate quality as a template for the electroplating. Figure 3(a) is a SEM image of submicron sized character like Ni structures with a 300 nm height. Imprinting on a PMMA resist layer was carried out at 160 °C and 3.5 MPa using the Ni structure in Fig. 3(a). The imprinting result was shown in Fig. 3(b), where the submicron sized structure representing characters was successfully transferred on the PMMA surface.

The micro-structure of PMMA was successfully transcribed to the Ni structure as a reversal pattern. The potentiality of the PBW with the electroplating was shown by the fabrication of a high aspect ratio metal micro-structure such as metal stamp for the imprint lithography.

References

- S. V. Springham, T. Osipowicz, J. L. Sanchez, L. H. Gan, and F. Watt, Nucl. Instr. and Meth. in Phys. Res. B, 130 (1997) 155.
- N. Uchiya T. Harada, M. Murai, H. Nishikawa, J. Haga, T. Sato, Y. Ishii and T. Kamiya, Nucl. Instr. and Meth. B, 260 (2007) 405.



Fig. 2 (a) SEM image of 5 μm thick PMMA with grid trenches by PBW at 1.7 MeV (fluence: 100 nC/mm²).
(b) SEM image of Ni structure of 5 μm height walls by electroplating using grid patterned 5 μm thick PMMA.



Fig. 3 (a) SEM image of sub-micron-sized characters of a 300-nm-height structure of Ni. (b) SEM image of an imprinted pattern on PMMA using the Ni stamp.

4-40 Effects of Proton Beam Irradiation on the Formation of Si Nano-Crystals

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Si nano-crystals attract much interest because of their unique electrical and optical properties. The formation of Si nanocrystals and control of spatial distribution are important in industrial devices. In this work, Si nano-crystals are fabricated in Si-thermal oxide film by ion implantation and thermal annealing. Effects of proton beam irradiation were also investigated.

半導体ナノ結晶は量子サイズ効果による特異な電 気的特性や光学的特性を持つため、多くの注目を集め、 光・電子デバイスへの適用が検討されている。特に、 その発光性や電荷蓄積機能を利用したエレクトロルミ ネッセンス(EL)素子およびメモリ機能への適用は多く の注目を集めている。また、自然界に大量に存在する ため環境負荷が小さく、半導体や集積回路において広 く利用されているシリコンのナノ結晶(nc-Si)は産業用 途においても重要である。nc-Siの形成にSiイオン注入 法を用いる手法では、イオン注入量と加速エネルギー を変えることによって、nc-Siの析出する領域の深さ と分布を制御できるという利点がある。この際、nc-Siを析出するには、1000 ℃以上での熱処理が必須と される。しかし、近年の研究でRTA処理(Rapid Thermal Anneal)効果により1000 °C以下でのnc-Siの析 出が確認された。本研究ではMV程度の高エネルギー の集束プロトンビーム照射による、イオンの飛跡に 沿った局所的な加熱を利用し、nc-Siを空間的に析出 するための研究に取り組んでいる。

本研究の目的はSi⁺イオンを注入した熱酸化膜ヘプ ロトン照射し、その試料の発光特性や熱処理効果を評 価し、プロトン照射による局所的な照射効果によって nc-Siの形成促進を目指すことである。

Si熱酸化膜(膜厚600 nm)に対してシリコンイオン注入(Si⁺, エネルギー200 keV, 照射量:3-7×10¹⁶ ions/cm²) を行った。顕微フォトルミネッセンス(PL)による光学 特性評価(室温、488 nm励起)を行った。Si注入後の熱 酸化膜へのプロトンビーム照射は芝浦工大フレキシブ ル微細加工研究センターにて水素イオンビーム発生装 置を用いて加速電圧500 kVにて行った。

Fig. 1にプロトン照射後のPLスペクトルを示す。 650 nm付近に欠陥によるPLピークが観測された。また、Si注入のみの場合に比べ、プロトン照射によりその強度が増大した。その後、これらの試料に対して窒素雰囲気中1050 ℃にて1時間の熱処理を行った際のPL スペクトルの一例をFig. 2に示す。熱処理により、PL のピーク波長は720 nm付近にシフトし、これはnc-Si の形成によると考えられる。プロトン照射によりこの ピーク値は50%程度増大した。これより熱処理前のプ ロトン照射は、1050 ℃での熱処理時のnc-Siの析出を 促進する効果をもたらしたと考えられる。今後は、よ り低温での熱処理によるnc-Si析出する条件を探索し、 プロトンビーム描画の析出を空間的に制御する手法と するための検討を行う。



Fig. 1 Comparison of the PL spectra of Si implanted thermal oxide (600 nm, 200 keV, 7×10^{16} ions/cm²) of different depending on the proton beam (PB) fluence (500 keV).



Fig. 2 PL spectra after proton beam (PB) irradiation (500 keV, 3.0×10^{11} ions/cm²) and subsequent anneal in N₂ for 1 hour at 1050 °C obtained for Si-implanted thermal oxide (600 nm, 200 keV, 7×10^{16} ions/cm²).

4-41 Energy Dependence for 2-Dimentional Nuclear Reaction Distribution of Boron Doped Steel

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The microanalysis of trace amount of boron has been carried out using 1 μ m proton microbeam from 3MV single ended electrostatic accelerator of TIARA facility. In 2007 steel specimens containing 100 ppm boron have been bombarded by 1.5, 2.0 and 2.5 MeV proton beam for gamma ray detection. The 2-dimensional boron distributions dependent on proton energy have been measured by detecting 428 keV gamma ray from the ¹⁰B (p, $\alpha'\gamma$) ⁷Be nuclear reaction. As a result of imaging, segregation of several microns sized boron precipitates has been observed, but the correlation between energy dependent distributions could not be clearly seen.

鉄鋼材料に微量のホウ素を添加することにより強度 が増大することは経験的に知られているが、鉄鋼中で のホウ素の振舞いについてはまだ解明されていない。 これは鉄鋼中に限らず、ホウ素の局所的かつ定量的な 微量分析が、SIMS、EDXA、EELS などの分析方法でも 難しいことによる。ホウ素の局所的(三次元的)な定 量分析ができるようになれば、ホウ素添加鉄鋼材料の 強度特性とホウ素の分布の関係を明らかにでき、熱処 理等のプロセスにおいてホウ素分布の最適化を図るこ とができる。これは、ホウ素の効率的な処理によって、 クロム、モリブデンなどの鋼中合金元素を必要とする ことなく所要強度の実現を可能にするものであり、生 産コストの低減化、省資源化、そして環境に優しいリ サイクルマテリアルとなる。

材料中に含まれる微量のホウ素分布を空間的、定量 的に精度良く調べるために、陽子マイクロビームとホ ウ素との核反応を利用した分析法を採用し、新たな分 析手法の確立を目指すのが本研究テーマの目的である。

2007 年度は TIARA の大気マイクロ PIXE のポートを 用いて、陽子とホウ素の核反応で放出される y 線の二 次元分布の測定を行った。特に y 線の二次元分布の陽 子エネルギー依存性の測定を試みたので報告する。

ホウ素の定量的な分布を求めるために、これまで ¹⁰B (p, $\alpha' \gamma$)⁷Beの核反応によって放出される 428 keV の γ 線を、HPGe 半導体検出器を用いて測定を行ってきた¹⁾。 分解能は 1.33 MeV において 1.7 keV である。真空チャン バー側には Si(Li)の X 線測定用の検出器が設置されてお り、 γ 線と同時に X 線も測定する(PIXE)ことができるように なっている。

今回用いた試料は100 ppmのホウ素を含んだ鉄の板 (10×10×1 mm) で、Bの他、C, Si, Mn, P, S, N, Cr, W, Co,Vを含む。

Fig. 1に核反応の全断面積を示す²⁾。この核反応では 1.5 MeV付近に小さな共鳴があり、700 keV位から非共 鳴の断面積がなだらかに上昇する。3 MeV位になると 色々な核反応が現れてこの反応の測定が困難になるの で、今回は1.5、2.0 及び 2.5 MeVの入射エネルギーで ホウ素の二次元分布がどの様に変わるか、測定を試み た。それぞれのイオンが核反応を起こす最大の進入深 さは~8 (1.5)、~14 (2.0)、~22 μm (2.5 MeV) であり、 6 から 8 μmの進入深さの差があることから、ホウ素の 深さ分布の情報が得られるのではないかと考えられる。 さらに、これまでの二次元分布測定により鉄鋼中にあ るホウ素は直径10ミクロン位の粒状になって分布して いるのが観察されているので、深さ方向にも同様な分 布を持つと考えられ、3点のエネルギーではあるが、 互いに何らかの相関を持つ分布図が得られるのではな いかと考え、実験を行った。



Fig. 1 Total cross-section of ${}^{10}B(p,\alpha' \gamma)^7Be$ reaction.

Fig.2 に 1.5、2.0 及び 2.5 MeV の入射エネルギーで 得られた、ほぼ同一と思われる場所のホウ素の二次元 分布を示す。現在の試料セッティングでは完全に同一 場所への照射は難しく、この点は今後の課題である。 図中の黒い部分がホウ素であるが、入射エネルギーが 大きくなるにつれて黒い部分が多くなっているようで はあるが、これらの間に相関は良くわからない。



Fig. 2 A typical γ-ray images of 100 ppm boron contained steel specimen bombarded by 1.5 (left), 2.0 (middle) and 2.5 MeV proton microbeam (right).

- H. Shibata et al., Nucl. Instrum. Meth. Phys. Res. B 260 (2007) 321-324.
- R. Mateuset al., Nucl. Instrum. Meth. Phys. Res. B 219-220 (2004) 519-523.

4-42 Three-Dimensional Measurement of Elemental Distribution in Minute Samples by In-Air Micro-PIXE

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1. Introduction

An in-air micro-PIXE (Particle Induced X-ray Emission) analysis system was developed by Japan Atomic Energy Research Institute in 2000^{11} . In this system, two-dimensional distributions of trace elements in a single biological cell were successfully measured with spatial resolution of 1 µm. The system has been used as a powerful tool of two-dimensional elemental analysis for biological samples up to date, but it is not effective enough to measure the depth profile in the samples having lamellar structure. The purpose of this study is to measure the three-dimensional distribution of trace elements in a minute biological sample. Such a distribution is acquired in the same way as X-ray CT (Computed Tomography) by the reconstruction of the many two-dimensional images of micro-PIXE taken from various directions. Furthermore, the three-dimensional structure of the target must be measured accurately using STIM (Scanning Transmission Ion Microscopy) - $CT^{2)}$.

2. Experiment

The 3MV single-ended accelerator and the micro-PIXE system at the TIARA were used in this study. The 3 MeV proton microbeams were scanned over the target which was glued on a metal needle and was rotated by 180 degrees at intervals of 15 degrees. The tomograms were reconstructed from the twelve projections using the back projection method with the Shepp and Logan filter.

3. Result

Figure 1 is a photograph of the test sample: the metal adsorbent with radiation-induced graft polymerization developed by Environmental Polymer Group, JAEA³⁾, and Figure 2 shows its PIXE spectrum of the sample. There are many peaks of the characteristic X-rays in the spectrum because the sample recovered some elements from seawater. Twelve projections per a sample were measured. Using all tomograms and the back projection method with the Shepp & Logan filter, three-dimensional distribution of some trace elements were reconstructed as shown in Fig. 3. Since the fiber of about 10 μ m diameter was successfully visualized, three-dimensional maps of the trace elements have the spatial resolution of 10-20 μ m.

- T. Sakai et al., Int. J. PIXE Vol. 10 Nos. 3 & 4 (2000) 91.
- 2) J. C. Overley et al., Nucl. Instr. Meth. B 30 (1988) 337.
- 3) N. Seko et al., Nucl. Technol. 144 (2003) 274.



Fig. 1 Optical microscope image of the metal adsorbent fiber which glued on the metal needle.



Fig. 2 PIXE spectrum of the metal adsorbent which recovered some elements from seawater.



Fig. 3 Reconstructed three-dimensional distribution of **calcium** and **vanadium** which were recovered from seawater.

4-43 Measurement of Sensitivity of Micro Beam PIXE System in TIARA for Zinc and Platinum

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Standard Reference Material (SRM) for determination of platinum and zinc was made by anion-exchange resin. Macro porous anion-exchange resin, Macro-Prep 25Q (BioRad) was suspended in 3 M HCl and added the known amount of Zn and Pt. The concentration of Zn and Pt in the resin was 0-100 ppmv and 0-200 ppmv, respectively. Individual particle of the resin was subjected to 3 MeV proton bombardments by micro beam system of TIARA. It was found the macro porous nature of the resin allows $PtCl_6^{2-}$ and $ZnCl_3^{-}$ to react with the exchange sites located throughout the matrix. PIXEana program was used to analyze peak count for Zn-K α and Pt-L β . Sensitivity for Zn and Pt was calculated by elemental abundance in the resin and does of proton. Count for characteristic X-ray per pg (element) × nC (dose) was 203±15 (Zn, n=8) and 45.2±6.5 (Pt, n=8). Detection limit for typical biological analysis (10 × 10 × 10 μ m³ sample, 50 nC dose) was 0.18 fg (Zn) and 0.8 fg (Pt). And also minimum elemental concentration for image analysis to Zn and Pt was 0.18 ppmv and 0.80 ppmv, respectively.

白金(Pt)の錯体は、抗ガン剤としてその研究と利用 が進められ、マイクロPIXEシステムを用いたPtの細 胞内分布が調べられている¹⁾。PtのPIXE スペクトル 解析では、亜鉛 (Zn)のZn Kβ と Pt Lα 線との重なる ことと、Znが生体必須元素あることから、定量感度 を求めることが急務とされている。これまで、粒径約 25 μmの球状イオン交換樹脂に、微量元素を吸着させた 標準試料 (SRM)を試作し、有効性を確かめてきた²⁻⁴⁾。 本研究では、SRMに正確量のPtとZnを添加し、マイ クロビーム照射による特性X線強度と元素添加量とか ら、分析感度を求めたので報告する。

実験 Zn、Pt 含有SRMの調製	マクロ	コポーラ	ス型
陰イオン交換樹脂(Macro Prep	25Q:	ポアサ	イズ
725Å、BIORAD)を既報のと	Table 1	Concenti	ation
おり、水洗後、3M 塩酸に懸濁	of Zr	n and Pt in	SRM
した ²⁾ 。樹脂を0.358 mL含む懸	(ppm	ıv).	
濁液1 mLをガラスバイアルにと	No.	Zn	Pt
り Z nお上びPtの標準溶液を加	1	100	0
	2	75	50
え 旅どうした。 樹脂 () 傾の にり	3	50	100
のZnおよびPt濃度(ppmv)を	4	25	150
段階的に変化させた6種類の	5	0	200
SRMを調製した(Table 1)	Bl	0	0

<u>マイクロPIXE分析</u> 3 MeV pビームをサブミクロンに 絞って照射した。スキャンエリアを30×30 µm とし、 球状の樹脂1粒に対し、大気中75 - 140 nCの照射を 行った。検出器にはPure Geを用いた。

結果と考察 <u>SRMのマイクロPIXE像</u> マクロポー ラス型の樹脂内に均一に分布したイオン交換基に PtCl₆²⁻ および ZnCl₃⁻ が均一に分布し、その交換基と 金属イオンが反応することが確かめられた。Znおよ びPtの特性X線のエネルギー領域にROIをかけること で、それぞれの元素の樹脂内の元素分布を画像化でき た。解析プログラムのPIXEana により、照射した SRMの粒径を見積もるとともに、SRMからのX線スペ クトルを抽出し解析した。照射した10粒のSRMはほ ぼ球形であり、一粒の半径は、7.4 - 10.7 μm、体積は、 1700 - 5100 μm³であった。

<u>Zn、Ptの定量感度</u> SRM No.3 からのX線スペクトル をFig. 1に示す。Zn Kα、Zn Kβ + Pt Lα、Pt LβのX線領 域のピーク面積と樹脂中の元素量および照射電荷量か ら定量感度を求めた(Table 2)。例えば10×10×10 μ m³の試料に50 nCを照射すると、Pt-Lβ領域から、BG が135カウント発生することになる。検出下限が3 \sqrt{BG} とすると、Pt 0.8 fg相当となり、超高感度分析が可能 であることがわかる(Zn で8.9 fg)。同じ照射で、画像 解析に必要最低限のカウント(1ピクセルあたり1カウ ントとして1800カウント)をPt-Lβから得るためには、 絶対量として0.80 pg、面積密度として0.80 μ g/cm²のPt が必要になる。厚みが10 μ mとすると、濃度は 0.80 ppm(Znで0.18 ppm)と計算できる。



Table 2 Sensitivity Calculated from Spectrum Analysis and Elemental Concentration.

- Background Count and Sensitivity for Zn and Pt Zn -

	Zn-Kα	Pt-Lβ
Peak Count for Characteristic X-	203±15	45.2±6.5
ray (Count / pg nC)	(n=8)	(n=8)
Background Count Calculated from Blank Sample (Count / nC)	18	2.7

- 木村仁、桑野博行他、第1回高崎量子応用研究シンポジウム要旨集 p173.
- 2) Y. Iwata et al., TIARA annual report 2004 (2005) 320.
- 3) Y. Iwata et al., JAEA Takasaki annual report 2005 (2006) 171.
- 4) Y. Iwata et al., JAEA Takasaki annual report 2006 (2007) 181.

4-44 Simultaneous Measurement of Charge States and Emission Angle of C₂⁺ Fragments Emerging from a Thin Carbon Foil

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In regard to the interaction of a swift cluster ion with a solid, the energy-loss process of fragment ions of the projectile in the solid becomes more complicated than that of a single-ion projectile because the mutual interference of the vicinal ions participates in the process. The collision reaction mechanism of a cluster ion for a solid has been discussed by a lot of theories. Especially, the theoretical calculations of the collision reaction for a cluster or a molecular of light-ions at least qualitatively consist with the experimental data by adding effects of both a Coulomb explosion and a polarization weak to the theory for a projectile of a single ion. However, the theoretical calculations of the collision reaction for the heavy cluster ion of which the influence of the bound electrons of the projectile cannot be ignored in the case not has been verified because there are not enough the experimental data of it.

The relationship between the spatial arrangements of a cluster ion colliding with a foil and the charge states of fragment ions emerging from the foil is investigating, in order to understand the reaction mechanism of the heavy cluster ion collision with the solids. It was demonstrated both experimentally and theoretically that the charge state distribution of fragment ions emerging from the foil depends on the structure of the incident cluster in fiscal 2006 using 3-MeV, C_3^+ ion¹⁾. In the fiscal 2007, we improved the measurement system in order to observe the dependence of the average charge states of the fragment ions at the exit surface of the foil on the orientation (φ , see Fig. 1) of a C₂⁺ ion to the beam direction. In this report, the preliminary results of simultaneous measurement of the charge states and the emission angle of the fragment ions emerging from a thin carbon foil is presented with those of the improved measurement system which enhanced the resolution of the emission angle.

The experimental setup is schematically shown in Fig. 1. Carbon cluster ions (C_2^+) generated in a cesium sputter ion source were accelerated to 6-MeV (3-MeV/atom) by the tandem accelerator at JAEA/Takasaki. The intensity of incident C_2^+ ions was reduced at the rate of approximately



Fig. 1 Experimental setup.

1-2 clusters per second both by a beam attenuator and switching deflection plates, which were placed at the low energy beam line. A carbon foil of 1.0-µg/cm² thickness was used as the target. The fragment ions emerging from the foil were deflected depending on their charge states by an electric field applied to the deflection plates placed between the foil and a micro-channel plate (MCP). The deflected angles depend on their charge state. Two-dimensional pattern of the deflected ions was observed as luminance points on a fluorescent screen equipped with the MCP. To measure the emission angle of the fragment ions emerging from the foil in high accuracy, we used the projectile of three times higher energy and extended the distance between the target and the MCP by over seven times than the previous setting. The pattern of luminance points on the screen is recorded with a CCD camera as a digital image.

The emission-angle distribution of the fragment ions is shown in Fig. 2. The emission angle and charge state are estimated from the distance between two luminance points on the MCP in consideration of the deviated angle from the original trajectory by the electric field of the deflection plates. The emission-angle distribution having an independent peak was expanded to the wide angle side in proportion to a product of the charge state of fragment ions. We think that a very few differences in the Coulomb repulsion force depending on the charge combination between the fragments of C_2^+ ion at the exit surface of the foil was measured accurately by suppressing the effect of multiple scattering in the foil and enhancing resolution of the emission angle. Hereafter, we will analyze the observed emission-angle distribution using a trajectory calculation, and clarify the relationship between the orientation of the incident cluster ion and the charge states of the fragment ions emerging from the foil.



Fig. 2 The emission distributions -angle of the fragment ions emerging from the foil (1.0 μ g/cm²) with 6-MeV, C_2^+ . The distributions of each charge combination are plotted for $(q_1,q_2)=(2,3)$: (O), $(q_1,q_2)=(3,3)$: (•) and $(q_1,q_2)=(3,4)$: (�).

Reference

1) A. Chiba, Y. Saitoh, K. Narumi, M. Adachi and T. Kaneko, Phys. Rev. A 76 (2007) 06321.

4-45 Carbon-build-up Analysis of Si Surfaces Bombarded with 10-to-100-keV C₆₀ Ions

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One of the most important radiation effects of 10-to-100-keV C₆₀ ions is the strong sputtering localized around the surface layer, which is different from the case by single-ion bombardment. Intensive application studies of the sputtering have been made recently based on this effect for possible applications to the surface-sensitive analyses and secondary-ion-mass analyses of high-polymer materials and/or biomaterials. However, there is not enough understanding of the sputtering phenomenon induced by bombardment with C₆₀ ions in this energy range. The energy dependence of the sputtering yield and of the deposition, or implantation of carbon atoms could be the key measures to understand the relevant phenomena. We will present in this report the analysis of carbon concentration built-up on Si surfaces bombarded with 10-, 50- and 400-keV C₆₀ ions.

Pieces of Si(100) wafer were cleaned with a wet chemical treatment to reduce carbon contaminants on the surface. Then, the samples were irradiated with C_{60} ions from the 400-kV ion implanter of TIARA. Carbon concentration in the surface layer was evaluated with a nuclear reaction ${}^{12}C(d, p){}^{13}C$ using 1.2-MeV D⁺ from the single-ended accelerator after the C_{60} bombardment.

Figure 1 shows the fluence dependence of the carbon concentration in the surface layer of C_{60} -bombarded Si. The number of ¹²C nuclei per cm² was evaluated with the use of the cross section measured by Kashy *et al.*¹⁾ A solid line in the figure shows carbon areal density estimated from



Fig. 1 Fluence dependence of the carbon concentration at Si surfaces bombarded with 10-, 50- and 400-keV C_{60} ions. Solid line shows carbon areal density which is 60 times as much as C_{60} ion fluence. Broken lines show the result of the fitting by the rate equation (1).

Table 1	Sputtering	cross	section	for	carbon	and	number
of the	e implanted	carbo	n atom o	obta	ined as	the v	variables
in fitt	ing with the	rate e	equation	(1).			

Energy (keV)	Sputtering cross section for carbon (cm ²)	Number of the implanted carbon atom
10	1.2×10 ⁻¹⁵	130
50	1.8×10^{-14}	320
400	5.5×10 ⁻¹⁵	200

the C_{60} ion fluence, assuming that all of the carbon atoms due to an incident C_{60} ion remain at the surface. The experimental results are inconsistent with this line: At lower fluence, measured carbon concentration is higher than the estimated value, while at higher fluence the increase rate of carbon concentration decreases and the concentration is lower than the estimation.

In order to explain the results, we have assumed a rate equation describing the carbon concentration $N(/\text{cm}^2)$ at the surface as a function of the C₆₀ ion fluence $\phi(/\text{cm}^2)$:

$$\frac{dN}{d\phi} = 60 - 60B(E) - \frac{N}{N_c}Y(E) + \frac{N_a}{N_c}A(E),$$
(1)

where B(E) is the probability of backscattering of a carbon atom of an incident C_{60} ion, N_c (/cm²) the concentration of solid carbon, Y(E) the sputtering yield for carbon per C₆₀ ion, $N_{\rm a}$ (/cm²) the concentration of carbon of the surface contaminants, A(E) the probability of recoil implantation of a carbon atom of the surface contaminants, E the energy of an incident C_{60} ion. In eq. (1), the carbon concentration is determined by the competition between the sputtering and the carbon deposition or implantation which includes the contribution of C60-bombardment-induced recoil of the surface contaminants; and the sputtering cross section for carbon Y/N_c and the number of the carbon atom implanted by C₆₀-ion bombardment $60(1 - B) + N_a A/N_c$ are variables. Broken lines in Fig. 1 show results calculated from eq. (1). The results reproduce the observed fluence dependence well, which supports our assumption.

The obtained sputtering cross section and number of the implanted carbon atom are shown in Table 1. Both of them seem to have peaks around 50 keV. What such energy dependence is attributed to is the next issue under consideration.

Reference

1) E. Kashy et al. Phys. Rev. 117 (1960) 1289.

4-46 Effect of Energetic 10 MeV Iodine Irradiation on the Magnetic Properties of FeRh Thin Films

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Recently we have found that energetic heavy ion irradiation induces the ferromagnetic state in Fe-50%Rh alloy at low temperatures^{1,2)}. To discuss the ion irradiation effects more quantitatively, we have to perform the irradiation by using thin films, thickness of which is much smaller than sample thickness. Under such an experimental condition, irradiating ions pass completely through the sample and the loss of ion energy in the sample is very small. We have therefore prepared thin FeRh films for the energetic ion irradiation experiments. This is our first report concerning the effect of energetic ion irradiation on the magnetic properties of FeRh thin films.

Fe₄₆Rh₅₄ thin films about 200 nm thick were deposited on amorphous SiO₂ substrates by using an ion beam sputtering. The thin samples were irradiated with 10 MeV iodine ions at JAEA-Takasaki tandem accelerator. The fluences of ions were 1×10^{12} , 2×10^{12} , 5×10^{12} , 1×10^{13} , 2×10^{13} , 5×10^{13} and 1×10^{14} /cm². After the irradiation, the magnetization was measured for each sample as a function of temperature by means of SQUID magnetometer. The results are shown in Fig. 1 for low ion fluence (below 5×10^{12} /cm²) and in Fig. 2 for high ion fluence (5×10^{12} /cm² - 1×10^{14} /cm²). As can be seen in Fig. 1, the magnetization below the ferromagnetic(FM) – antiferromagnetic (AF) transition temperature increases and the FM-AF transition temperature shifts to a lower temperature side with increasing the ion-fluence. The magnetization of the film exhibits the maximum value for the ion-fluence of 5×10^{12} /cm², where the FM-AF transition completely disappears. The tendency of the increase in Ms of FeRh thin films by the ion irradiation is the same as that of bulk samples. On the other hand, above the transition temperature, the magnetization of FeRh thin films does not change even when irradiated up to the fluence of 5×10^{12} /cm². For the high ion-fluence, Figure 2 shows that the magnetization of the film is decreased by the irradiation. An X-ray diffraction measurement indicates that the B2 structure is changed into the nonmagnetic A1 structure by irradiation, leading to the strong decrease in Ms.

Through the present experiment, we could observe the 10 MeV iodine irradiation effects of FeRh thin films. As a next step, we plan to discuss quantitatively the dependence of magnetic property change on some irradiation parameters (ion energy, electronic and nuclear stopping powers, ion velocity and so on) by conducting a variety of irradiation experiments.

- A. Iwase, M. Fukuzumi, Y. Zushi, M. Suzuki, M. Takagaki, N. Kawamura, Y. Chimi, N. Ishikawa, J. Mizuki, and F. Ono, Nucl. Instr. and Meth. B 256 (2007) 429.
- Y. Zushi, M. Fukuzumi, Y. Chimi, N. Ishikawa, F. Ono, and A. Iwase, Nucl. Instr. and Meth. B 256 (2007) 434.



Fig. 1 M-T curves of the FeRh thin films for the low ion-fluence ($\leq 5 \times 10^{12}$ ion/cm²). M-T curve for unirradiated sample is also shown.



Fig. 2 M-T curves of the FeRh thin films for the high ion-fluence ($\ge 5 \times 10^{12}$ ion/cm²).

4-47 Emission Yields of Positive and Negative Secondary Ions Originating from Organic Contaminants on a Silicon Wafer upon Cluster Ion Impacts

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Organic contaminants on the surface of a semiconductor wafer causes serious degradation on electrical performance of integrated circuit devices fabricated on the wafer. In order to optimize the manufacturing processes for high performance devices, analytical tools for the surface contaminations are required. Time-of-flight (TOF) secondary ion mass spectrometry is one of the most powerful tools to characterize the surface contaminants with high sensitivity. Mass analysis of secondary-ions is based on the phenomenon that secondary ions are emitted from the surface when the primary ions bombard the target. To TOF secondary ion mass spectroscopy, we have applied pulsed cluster ion beams, which gives different secondary ion emission yields from those for monoatomic ions because of their peculiar irradiation effect^{1,2}. In this paper, we compare the emission yields of positive and negative secondary ions (P- and N-SIs) originating from organic contaminants on a silicon wafer between impacts of cluster and monoatomic ions with the same velocity $^{3)}$.

A Si(100) wafer was chemically cleaned by repeated NH₄OH/H₂O₂/H₂O and HF/H₂O treatments, followed by rinsing in deionized water. The wafer was kept in a polymer container before being set on a grounded holder. A 3MV tandem accelerator at the Japan Atomic Energy Agency (JAEA)/Takasaki¹⁻³⁾ was used for the experiments. A fresh point of the samples was bombarded for each measurement. P- and N-SI TOF measurements were performed with a linear type TOF mass analyzer, detailed in previous paper²⁾, using pulsed monoatomic and cluster ion beams. For the N-SI TOF measurements, several improvements were made over the TOF system.

For quantitative comparison of mass spectra by TOF measurements, peak intensities should be scaled under the condition of the same number of incident primary atoms. In our previous paper,²⁾ the scaling of P-SI emission yield per incident atom was performed based on I_P/nI_o , where I_o , I_P , and n are the direct current (DC) of the incident beam, P-SI current under DC beam irradiation, and cluster number, respectively. In addition to the I_P/nI_o measurements, the relative N-SI emission yield per incident atom I_N/nI_o was obtained by combining I_P/nI_o with the total counts C_i of detected SI (i=P or N : P and N are for positive and negative TOF measurements, respectively) during the TOF measurements.

Figure 1 shows the P- and N-SI TOF mass spectra of the organically contaminated Si for the impacts of carbon monoatomic and cluster ions with the same velocity (0.5 MeV/atom- C_1^+ and C_8^+). The relative intensity on the

vertical axis is proportional to the SI emission yield per incident atom. The major peaks in the figure were assigned to atomic and cluster hydrogen, and carbon and hydrogenated carbon clusters $[C_pH_q^-, C_pH_q^+, (p\geq 1, q\geq 0)]$. C atom(s) of $C_pH_q^-$ and $C_pH_q^+$ should be from the surface organic contaminants as an average implantation depth of incident C atom for 0.5 MeV-C in silicon is as high as ~1 μ m. We see in the N-SI spectra that the intensities of N-C_pH_q SIs with even p numbers are higher than those with odd p numbers for both the C_1^+ and C_8^+ impacts.

The other feature in the N-SI yields is that $C_pH_q^-$ for the cluster ion impact exhibits the highest emission yield per incident atom among $C_pH_q^+$ with the same p number. The highest relative $C_pH_q^-$ emission yield for the cluster ion impact reaches ~20 and ~60 times higher thamthose of $C_pH_q^-$ and $C_pH_q^+$ with the same p number for the impact of the monoatomic ion with the same velocity, respectively. This result shows that combination of negative secondary ion TOF measurements with cluster impact ionization is a promising tool for highly sensitive detection of organic-contaminants on silicon wafers.



Fig. 1 P- and N-SI TOF mass spectra of organically contaminated Si for 0.5-MeV/atom C_1^{+} and C_8^{+} . The relative intensity is proportional to the SI emission yield per incident atom.

- 1) K. Hirata, Y. Saitoh, A. Chiba, K. Narumi, Y. Kobayashi, and K. Arakawa, Appl. Phys. Lett., **83**, (2003) 4872.
- K. Hirata, Y. Saitoh, A. Chiba, K. Narumi, Y. Kobayashi, and Y. Ohara, Appl. Phys. Lett., 86, (2005) 044105.
- K. Hirata, Y. Saitoh, A.Chiba, and K. Narumi, Appl. Phys. Express, 1 (2008) 047002.

4-48 Visualization of a Single Cluster Particle Track in Polymeric Materials

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Charged particles accelerated up to MeV-order high energy have often cause severe interaction with the organic materials releasing their energy densely along their trajectories, giving cylinder-like nano-space of particle tracks. The cross-linking reaction in the nano-space of polymer materials used as the target materials gives 1-D polymer nanowires, and the energy distribution within the nano-space has been successfully visualized by the dissolution of un-crosslinked polymer materials after the charged particle irradiation.¹⁻⁵⁾ Theoretical formulation of the energy distribution reveals to give a good correlation with the size of the nano-space.1) Thus we can estimate precisely the size of the 1-D nanowire, hence, the energy distribution can be "measured" with nm-resolution if we use the polymer materials of which cross-linking efficiency has been known.^{1,4)} In the present report, we have successfully formed the nanostructure of well-known polymeric materials upon irradiation to a variety of cluster ion particles, and the remarkable correlation between the size and the linear energy transfer (LET) of the particle has been observed.

Commercially available high sensitive crosslinking-type polymer materials $SU-8^{6,7)}$ was used as the target polymer material in the present study. The polymer was over coated onto Si substrate after an adequate surface treatment, and irradiated by a series of C or Al cluster ion beams from



Fig. 1 AFM micrographs of nanostructures based on SU-8 produced by cluster ion particle irradiation. Images (a) – (d) were observed in the polymer films of SU-8 at ~0.3 μ m thick after irradiation of 1.5 MeV/atom Al₁ – Al₄ particles, respectively at the fluence of 0.50 ~ 2.0 ×10⁹ particles cm⁻².

Tandem accelerator at Takasaki Advanced Radiation Research Institute, JAEA. The irradiated polymer films were immersed into diacetone alcohol to isolate the nanostructures on the substrate, followed subsequently by rinsing in isopropylalcohol. The size of the nanostructures was directly measured by atomic force microscope (AFM).

Figure 1 shows the observed AFM micrographs of nanostructures formed by various Al cluster particles. The nanostructures represent wire-like 1-D features, and the cross-linked polymer nanowires are observed clearly upon irradiation to all the clusters. Although the number density of the nanowires corresponds precisely to the number of incident Al ions in the case of Al₁, the number density is apparently larger than the number of the incident clusters of $Al_2 - Al_4$, suggesting that fragmentation of the cluster particles occurs and each fragment gives corresponding nanowire in these films. The nanowires produced by Al₃ and Al₄ (Fig. 1-c,d) show considerable non-homogeneity in the thickness of the nanowires in contrast to the uniform thickness observed in the case of Al₁. It should be noted that the thickness of the "thin" nanowires in Fig. 1-d is consistent with that in Fig. 1-a, which is the case giving the "thin" nanowires produced by the Al₁ fragments from the cluster particles. The cross-linking reactions in SU-8 upon irradiation have been well investigated as the chemically amplified processes promoted by proton catalyzed reactions. The G value of cross-linking (G(x)), therefore, has been reported to be very high as $G(x) \sim 3-5 (100 \text{ eV})^{-1.6,7)}$ The theoretical formulation based on the value of G(x) and LET of the cluster particles (calculated as the sum of atomic LET) gives the estimates of the cross-sectional radii of the nanostructures as 9.0 nm (Al₁) \sim 18 nm (Al₄). However the observed sizes of the nanostructures are 10, 13, 18, and 25 nm, respectively for Al₁ - Al₄ cluster particles. The observed sizes show slightly higher values than the values predicted by our previous formulation based on the single particle (atom, ion) interaction with the polymeric materials, and increases almost linearly with the number of atoms contributing a cluster particles. This is suggestive that the estimate of the LET of a cluster particle should be higher than the sum of the atomic LET.

- 1) S. Seki, et al., Phys. Rev. B, 70 (2004) 144203.
- 2) S. Seki, et al., Adv. Mater., **13** (2001) 1663.
- 3) S. Tsukuda, et al., J. Phys. Chem. B, 110 (2006) 19319.
- 4) S. Tsukuda, et al, Appl. Phys. Lett., 87 (2005) 233119.
- S. Seki, et al., Surf. Coat. Technol., (2007), 201, 8486; ibid., (2007), 201, 8526.
- 6) D. Stumbo, et al, J. Vac. Sci. Technol. B, 11 (2003) 2432.
- 7) H. Lorenz, et al., J. Micromech. Microeng, 7 (1997) 121.

4-49 Projectile Energy Dependence of Secondary Ion Emission from HOPG Target Bombarded by Swift Cluster Ion Beams

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The interaction of fast (MeV) cluster ions with solids has attracted attention in basic physics and application fields. Swift cluster ion bombardment causes nonlinear effect or synergetic effect, which is unusual for single atom collision with solid targets. A time of flight (TOF) mass spectrometer combined with pulsed swift cluster ion beams which were produced by the TIARA tandem accelerator was used for a secondary ion measurement. In this report we present some results of projectile energy dependence of positive and negative secondary ion emission from a highly oriented pyrolytic graphite (HOPG) target bombarded with carbon cluster ions.

高速(高エネルギー)クラスターイオンと物質との 相互作用では単原子イオンの数の集合体として線形的 に振る舞うのではなく、個数以上、あるいは個数以下 の非線形的な効果を与える。二次荷電粒子放出の収量 や二次イオン放出スペクトルの測定を通して、高速ク ラスターイオン照射の特徴を調べることが本研究の目 的である。H19年度は直線飛行時間型(TOF)質量分 析器を用いて炭素とHOPG(Highly Oriented Pyrolytic Graphite:高配向性熱分解炭素)標的との衝突で放出さ れる正負の二次イオンの入射イオンエネルギー依存性 の測定を行ったので報告する。

標的試料には HOPG を用いた。HOPG 試料は層状に 剥がすことができ新しい表面を測定に使えるので他の 試料に比べて不純物の吸着が少ないと考えられる。そ れでも相当不純物(例えば、水、ナトリウムなど)が 観測されるので、ECR イオン源からの Ar ビーム照射に より表面のクリーニングを行った。TOF スペクトルの 計測はイオンビームをパルス化し、TAC (時間波高変 換)を用いる方法により行われた¹⁾。

Fig. 1に0.5, 1.0及び1.5 MeV/atom の炭素クラスター イオンを HOPG に照射した際に放出された正負二次イ オンに対する水素を除いた正イオンの全収量の C_1^+ イ オン照射における正イオン全収量に対する比を示す (C_1^+ 照射における正イオン放出の全収量をそれぞれ 1 とした)。各々のエネルギーにおける正負二次イオン スペクトルに相違はほとんど見られなかった。 0.5 MeV/atom の場合以外は2点のみの測定であった が大まかな傾向は推測できよう。

Fig.1からわかるように3種の入射エネルギーについて比較してみると、エネルギーによらず正負イオンの収量は正の非線形性を示している。1.0 及び

1.5 MeV/atom の場合は2点のデータしかないのではっ きりしたことはわからないが、エネルギーが大きくな るにつれ収量もクラスターサイズに対しほぼ指数的に 増加していると思われる。すべての入射エネルギーに ついて、負イオンの収量は正イオンの数倍から数10倍 になっていて、この傾向はエネルギーに関わらず同様 である。この理由は現時点では明らかではない。

二次イオン放出現象においては、イオンの速度が Bohr 速度以下の低速領域では、核的衝突が支配的でク ラスターイオンでは負の非線形性が、また Bohr 速度以 上の高速領域では電子的衝突が支配的で正の非線形性 が表れることが理論的に知られている²⁾。今回の実験で は核子当たり約42 keV (0.5 MeV/atom)の場合から 126 keV (1.5 MeV/atom)の場合と Bohr 速度より大き く、阻止能で考えると、エネルギーが大きくなるにつ れ阻止能も大きくなり126 keV 付近では最大値付近に なっていると考えられる。以上のように定性的には実 験結果を説明できる。



Fig. 1 The ratio of yields of positive (left) and negative (right) secondary ions emitted from HOPG target bombarded with 0.5, 1.0 and 1.5 MeV/atom carbon incident ions. The yield of positive secondary ions bombarded by C_1^+ ion is unity.

- 1) Shibata et al., TIARA Annual Report 2005 (2007) 177.
- 2) P.Sigmund et al., Nucl. Instrum. Meth. B112 (1996) 1.

4-50 Fast Single-ion Hit System for Heavy-ion Microbeam at TIARA Cyclotron

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The focusing microbeam system ^{1,2)} is being developed at the HX course of the TIARA cyclotron. The minimum beam size of 0.6 μ m in diameter was obtained using the flat-top acceleration (FT)³⁾. The microbeams were supplied to research experiments⁴⁾ on radiation biology and single event upset of semiconductors in space environment, often with a moderate size of 1.2~2.0 μ m without FT.

Those experiments employ single-ion hit technique, sequential hits of one ion at one targeted point. The single-ions are delivered reducing beam intensity by some orders of magnitude after forming a microbeam. Therefore the spatial targeting accuracy is the same as the beam spot size, which is sufficiently small already.

On the other hand, the required hit rate of higher than 10/s is a subject to be realized. The magnetic scanner installed in the HX course was replaced with a fast electrostatic scanner for this purpose. The measurement result of scanner performance and other improvement made in fiscal 2007 are described below.

The scanner consisting of two pairs of parallel plate electrodes, one for X and the other for Y, is placed upstream the focusing lens. The electrodes are 70 mm long and 30 mm wide, and their gap is 10 cm. The maximum voltage of ± 10 kV can be applied. The coordinates of the targeting points are preset in a control computer for the high-voltage supply. The measurement was carried out using about 2 μ m size beam of 260-MeV ²⁰Ne⁷⁺ accelerated without FT. A film detector of CR-39 on the focal plane of the focusing lens was hit with single-ions targeting an array of 2401 points on a square grid spaced 20 μ m apart. The target coordinate was swiftly switched to the adjacent point successively by triggers of signals from a pin-photo diode beneath the film to detect ions passing through it.



Fig. 1 A part of a photomicrograph of 2401 etched pits on CR-39 film. The full hit area is 1000 μ m \times 1000 μ m.

As a result of the measurement, the following performances has been verified: 1) The targeting area is larger than 1000 μ m × 1000 μ m. 2) The targeting accuracy is maintained at the highest speed of targeting shift when the targeting travels 1000 μ m from one edge to the other. 3) The average hit rate over 2401 hits is higher than 10/s. Figure 1 shows an example of etched pits of hit points and Fig. 2 demonstrates that any preset pattern can be plotted.

Microbeams are extracted out into the air through a film separating vacuum from the air when a living cell is irradiated as a biological target. The beam is scattered and enlarged by the film. In order to reduce scattering and beam size growth comparable to the beam diameter, a film of 7.5 μ m thick polyimide was replaced with 0.2 μ m thick silicon nitride.

A quick change of microbeam ion species was tested using the cocktail beam acceleration without FT. After forming 1.8 μ m 260-MeV ²⁰Ne⁷⁺ microbeam, the beam was changed to 520-MeV ⁴⁰Ar¹⁴⁺ with the same mass to charge ratio using the cocktail beam acceleration⁵⁾. The Ar beam was focused to 1.4 μ m with minor adjustment of beam transport and final focusing. All changing process took about 20 minutes while it takes several hours from cyclotron startup to completion of microbeam formation.

- 1) M. Oikawa et al., Nucl. Instr. Meth. B 260 (2007) 85.
- 2) T. Kamiya et al., JAEA-Review 2007-60 (2007) 190.
- 3) S. Kurashima et al., Nucl. Instr. Meth. B 260 (2007) 65.
- 4) S. Onoda et al. and T. Sakashita et. al., in this annual report.
- 5) M. Fukuda et. al., JAERI-Review 98-16 (1998) 233.



Fig. 2 A logo of JAEA made with ion pits. Upper left is the original logo from which the targeting coordinates were created.

4-51 Uniform Beam Formation Using Multipole Magnets

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The Gaussian transverse distribution of a charged-particle beam can be transformed into a uniform distribution by introducing an odd-order nonlinear focusing field in a beam transport system¹⁾. Based on this idea, we have been developing the <u>multipole</u> magnet beam profile <u>uniformization system</u> (MuPUS) for advanced ion-beam applications at the JAEA AVF cyclotron facility. This irradiation technique is superior to currently available uniform irradiation methods (the beam scanning method using time-varying dipole magnetic fields and the beam expansion method using scatterers) in that we can perform high-uniformity irradiation at a constant particle fluence rate over the whole area of a large target.

First, we have made a theoretical study on beam uniformization by means of the nonlinear focusing method for construction of the uniform irradiation system²). To study the effect of the nonlinear magnetic fields on the beam profile, we have advanced the previous theoretical studies on beam uniformization of a Gaussian beam using odd-order fields. We have demonstrated the feasibility of



Fig. 1 Schematic view of the LB line and a typical beam envelope for uniform beam formation. The doublet quadrupole magnet (TDQLB2) has been relocated 1.0 m upstream from its original position and its magnetic polarity has been also reversed. The beam duct around TDQLB2 has been replaced by a square one (□100 mm) to reduce the beam loss. Two pairs of sextupole and octupole magnets have been installed around the doublet. For reduction of undesirable betatron-coupling induced by multipole magnets, the beam cross-section at the multipole magnets is set flat. For confirmation of the flat beam profile, a three-wire profile monitor (PM) has been introduced around the multipole magnets.

Table 1 Main parameters of multipole magnets.

	Sextupole	Octupole
	magnet	magnet
Axial length (mech.)	0.30 m	0.30 m
(effective)	0.33 m	0.33 m
Bore radius	57.5 mm	57.5 mm
Maximum field gradient	300 T/m^2	13000 T/m ³
Maximum coil current	350 A	320 A

uniformization of not only a Gaussian beam using the even-order fields instead of the odd-order ones, but also an asymmetric beam, e.g., a misaligned beam, by making a combination of the even and odd-order fields, especially with the sextupole and octupole ones. According to these formulas, the LB beam course of the cyclotron facility has been improved as shown in Fig. 1. Main parameters of the fabricated multipole magnets are summarized in Table 1.

After installation of multipole magnets, we successfully obtained the first experimental results. To produce a beam with a Gaussian distribution, required for precise uniformization, a thin aluminum foil (t 0.8 µm) was set onto the beam line behind the cyclotron exit³⁾. The beam is extracted through a thin titanium foil (t30 µm, \$100 mm) into air at the end of the beam line. The transverse profile at the target was tuned, monitoring the light emission from a fluorescent screen (AF995R, Desmarquest). Adjusting the strength of the multipole magnets, a 2D uniform beam was achieved. GAFCHROMIC films (HD-810, International Specialty Products) were used for more precise measurement of 2D distribution⁴⁾. Figure 2 shows the relative intensity distribution of the beam on the target film. A uniform area of 30 mm × 50 mm was formed. The rms uniformity of the central part of the uniform area is about 2 %, calculated from the optical density of the irradiated film.

We have achieved uniformization of a beam extracted from the cyclotron for the first time. A research and development study is now in progress for public use of MuPUS.

References

- 1) P. F. Meads, Jr., IEEE Trans. Nucl. Sci. **30** (1983) 2838.
- Y. Yuri et al., Phys. Rev. ST Accel. Beams 10 (2007) 104001.
- 3) Y. Yuri et al., in Proceedings of the 4th Annual Meeting of Particle Accelerator Society of Japan (2007) 607.
- 4) T. Agematsu et al., Radioisotopes, 57 (2008) 87.



Fig. 2 2D and 1D beam intensity distributions at the target measured using a GAFCHROMIC film, which was irradiated by a 10-MeV proton beam. The beam travels for 130 mm in air from the extraction foil to the film.

4-52 Development of Scintillation Detector for Beam Phase Measurement inside the Cyclotron

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In the JAEA AVF cyclotron, less than 1 µm sized heavy ion micro-beam formation with energies of hundreds MeV was achieved by a flat-top (FT) acceleration system, in which the beam phase width was restricted to 13.6 rf degrees, so that the energy spread of the beam was reduced to $0.02\%^{1)}$. The beam phase was controlled width with two pairs of beam-phase-defining slits in the cyclotron central region, and measured with the plastic scintillator at the first diagnostic station (CS0) after extraction from the cyclotron. The beam phase width of 1.2, 1.0 and 5.2 rf degrees in full width at half maximum (FWHM), was obtained for acceleration harmonics 1, 2, and 3, respectively²⁾. However, the relation between the beam phase width and the positions of the beam-phase-defining slits has not been determined exactly, since the beam phase width inside the cyclotron has not been measured precisely. Therefore we have developed a scintillation detector, embedded in a new main probe, for accurate beam phase measurement inside the cyclotron.

The original main probe consisted of the integral head of a carbon block for total beam current in the cyclotron and the differential head of three metal fingers for a beam current distribution along the radius. In the new main probe, the differential head has been replaced with a plastic scintillation detector for measurement of the beam phase width. As shown in Fig. 1, the detector is composed of a plastic scintillator, acrylic light guides and optical cables. Optical cables have been connected to a photomultiplier set at the end of in-air part of the main probe to keep it away from a strong magnetic field and rf noises inside the cyclotron.

During the measurement of beam current at the carbon block, the plastic scintillation detector is moved to the position behind the back of the integral head to avoid incident of a beam. If the carbon block is irradiated with beam power of 2 kW, its temperature will be more than 2000 °C. The radiant heat from the carbon block can melt the plastic scintillator because of its low melting point of about 70 °C. According to thermal analysis, we found that



Fig. 1 Picture of the new main probe head.

such a high beam power can be tolerated by inserting a cooling aluminum plate of a thickness of 1 mm between the integral head and the plastic scintillation detector.

The phase distribution of 45-MeV H^+ beam were measured for different positions of beam-phase-defining slits with both of the plastic scintillation detectors at the main probe and at CS0, respectively as shown in Fig. 2. The peak of the phase distribution at the main probe shifts in conjunction with the position of the slits. By contrast, no clear correlation between the peak location and the slit position is indicated at CS0. When the position of the slits is 12.6 mm, the beam phase widths at the main probe and that at CS0 are 9.8 rf degrees and 2.3 rf degrees respectively. The beam is partly lost at the extraction devices, probably due to extension of a radial beam spread.

Since precise control of the beam phase distribution during acceleration in the cyclotron is required for the FT acceleration system, the phase measurement inside the cyclotron is more important. The new plastic scintillation detector can contribute reduction of the beam energy spread for heavy ion micro-beam formation.

- 1) S. Kurashima et al., Nucl. Instr. Meth. B 260 (2007) 65.
- N. Miyawaki et al., in Proceedings of the 3rd Annual Meeting of Particle Accelerator Society of Japan and the 31st Linear Accelerator Meeting in Japan (2006) 907.



Fig. 2 Beam phase distribution measured at the main probe (a) and CS0 (b). Figure legend indicates the position of one of phase-defining-slits at pillar intervals of 0.8 mm.

4-53 Advanced Techniques in the Cyclotron for Microbeam Production

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Magnetic field stabilization

For production of a focused heavy-ion microbeam¹⁾ in the JAEA AVF cyclotron, the beam energy spread has to be reduced to $\Delta E/E = 2 \times 10^{-4}$ to avoid the chromatic aberration on beam-focusing. The magnetic field stability of the cyclotron is an indispensable prerequisite for the energy spread reduction achieved by fine tuning of the operating parameters including flat-top (FT) acceleration²⁾. While the magnetic field variation is known for many cyclotrons, the magnetic field of the JAEA cyclotron has been stabilized by controlling the iron temperature³⁾. Imperfection of the magnetic field stability, caused by temperature fluctuations, such as change of the outside air temperature, was observed.

We, therefore, tried to correct the magnetic field with both an additional coil and a control system with a proportional-integral-derivative (PID) feedback. The additional coil was winded around the upper main coil of the cyclotron. By adjusting the exciting current, fed from a small power supply (20A-20V), the magnetic field variation of the order of 10^{-5} can be corrected. The magnetic field measured with an NMR probe inside the cyclotron was used as a process variable for the PID control. The difference between the process variable and a set point is used to determine the exciting current of the additional coil. The PID control algorithm was executed on a Windows PC with a LabVIEW program.

A preliminary experiment on the magnetic field stabilization with the PID control was carried out for the acceleration of a 100 MeV $^{16}O^{4+}$ beam. Figures 1 and 2 show the magnetic field variations without and with the PID control, respectively. The results indicate the good performance of the PID control.



Fig. 1 Magnetic field variation is less than $\pm 5 \times 10^{-6}$ without the PID control.

Quick change of ion species of microbeam by cocktail beam acceleration technique

We can provide a 260 MeV ²⁰Ne⁷⁺ microbeam with a spot size less than 1 µm by using an FT acceleration system²⁾. Users of the microbeam need a wide range of linear energy transfer (LET). In order to vary LET widely, we have to change the ion species and kinetic energy. However, it usually takes about 6 hours or more to tune the cyclotron step by step for a new ion species. A cocktail beam acceleration technique⁴⁾ is, therefore, applied to microbeam formation for reduction of the tuning time. We can change the ion species to be accelerated by slightly shifting the acceleration rf frequency. Beam development for quick change of ion species from 260 MeV $^{20}\mbox{Ne}^{7+}$ (M/Q \approx 2.85) to 520 MeV 40 Ar¹⁴⁺ has been carried out for microbeam formation. As a result, we have succeeded to confirm formation of a 520 MeV 40Ar14+ microbeam using a secondary electron microscope image of a copper mesh within 30 minutes after the 260 MeV ²⁰Ne⁷⁺ microbeam.

- M. Fukuda, *et al.*, Nucl. Instrum. Methods Phys. Res. B 210, 33 (2003).
- S. Kurashima, *et al.*, Nucl. Instrum. Methods Phys. Res. B 260, 65 (2007).
- S. Okumura, *et al.*, Rev. Sci. Instrum. 76, 033301 (2005).
- M. Fukuda, *et al.*, in: Proc. 11th Symp. on Accelerator Science and Technology, Harima Science Garden City, 139 (1997).



Fig. 2 Magnetic field is stabilized within $\pm 1 \times 10^{-6}$ with the PID control. The magnetic field is corrected by adjusting the additional coil current.

4-54 Improvement of Beam Attenuator for Reliable Control of Beam Intensity

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Beam intensity control over a very wide range is required for experiments and beam tuning in the JAEA cyclotron. To attenuate the beam intensity quickly, an attenuator composed of metal meshes with many regularly-arrayed holes, was installed at each diagnostic station of IS2 and IS5 in the injection line of the cyclotron. We expect that, by inserting the meshes into the beam line, the beam intensity can be controlled with the beam size and emittance almost maintained. The attenuators of IS2 and IS5 can insert seven and two meshes in maximum, respectively. The beam intensity can be decreased with the attenuation ratio from 10^{-1} to 10^{-12} by combining meshes with the opening ratios η of 10^{-3} , 10^{-2} and 10^{-1} .

From previous experiments¹⁾, it was cleared that the $\eta = 10^{-3}$ mesh at IS5, where the beam size is small due to the waist point in the beam transport, the attenuation ratio did not agree with the designed opening ratio, and that the beam intensity distribution was also changed. Furthermore, it was cleared that, when one or more meshes with $\eta = 10^{-3}$ or 10^{-2} at IS2 were inserted, similar imperfection in beam attenuation to the $\eta = 10^{-3}$ mesh at IS5 was observed in spite of the large beam size. Then, we stopped using the $\eta = 10^{-3}$ mesh at IS5 and remodeled the IS2 station for a longer space between meshes. Additionally, new meshes with a smaller hole spacing were examined.

To attenuate the beam uniformly, the diameter and the hole spacing of the new meshes were 1/10 smaller than the original ones, as compared in Table 1. The attenuated beam intensity was measured using Faraday cups at the injection line and at the transport line after extraction from the cyclotron. The 2D distribution of beam intensity was measured at the target point of the LB course using GAFCHROMIC films²). As shown in Fig. 1, the profile of the beam attenuated by the new $\eta = 10^{-3}$ mesh is similar to that by the $\eta = 10^{-1}$ mesh, while the old $\eta = 10^{-3}$ mesh distorted the profile.

The IS2 station was replaced with a new one lengthened in the direction of the beam line. The steering magnet IST2 has been moved downstream by 1167 mm for providing the installation space of additional attenuators at IS2, while the old IS2 station had only one port. A three-sheet type attenuator (sheet interval: 55 mm) has been installed in addition to the previous seven-sheet type one (sheet interval: 20 mm). The largest mesh distance has been expanded from 120 mm to about 1100 mm by combining meshes at the seven-sheet type and the three-sheet one. The arrangement of the meshes is shown in Fig. 2. A view port has been also equipped at the new station for observation of the beam profile using a fluorescent screen. We will investigate further for the performance of new meshes and the effect of a long mesh distance at IS2.

Table 1 Main specifications of the old and new meshes.

	Old mesh		New mesh		
Material	SUS 304		SUS 304 Coj		oper
Thickness	0.076 mm		0.03 mm		
Diameter	0.1 mm		0.01 mm		
Spacing	2.8 mm	0.89 mm	0.28 mm	0.089 mm	
Opening ratio η	10-3	10 ⁻²	10-3	10-2	







(20ms irradiation)

Fig. 1 2D intensity distribution of the 260-MeV $^{20}Ne^{+7}$ beam measured using GAFCHROMIC films at the LB2 port. The beam was attenuated by the meshes of the opening ratios η of (a) 10^{-1} , (b) 10^{-3} (new), and (c) 10^{-3} (old).



Fig. 2 Block diagram of injection line with meshes of IS2 and IS5 after the IS2 remodeling. White block: meshes for users; yellow block: new meshes for examination; pink block, BV: beam viewer (fluorescent screen) for monitoring the beam profile; #: mesh number on the control box for users.

- N. Miyawaki, S. Okumura, S. Kurashima, Y. Yuri T. Yuyama, T, Ishizaka, K. Yoshida, I. Ishibori, T. Agematsu, T. Nara, and W. Yokota, JAEA Takasaki Annual Report 2006 (2007) 192.
- T. Agematsu, H. Hanaya, and T. Kojima, RADIOISOTOPES, 57 (2008) 87.

4-55 Development of Beam Generation and Irradiation Technology for Electrostatic Accelerator

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1. Influence of terminal vibration of the tandem accelerator on beam-energy stability

The beam-energy stability is an important factor for the micro-beam technology of TIARA tandem accelerator. The dependence of beam stability on a pressure of a charge exchange gas was investigated using the resonant nuclear reaction¹⁾ in FY 2006. The acceleration voltage is controlled by a feedback signal from the generating volt meter (GVM), so the fluctuation of the signal by the mechanical vibration of the GVM would spoil the acceleration voltage stability. And a main source of the vibration is rotation of pellet chains. In FY 2007, we examined the relationship between the rotation of the chains and the beam energy stability. We obtained the best stability when the rotating speed of the chain motor 1 and the motor 2 were 1,500 rpm and 2,100 rpm, respectively, and the energy stabilities of those were 7.1×10⁻⁴ and 8.9×10^{-4} . The beam-energy stability depends on the speed of the pellet chains going around. It is indicated that the control of the mechanical vibration is more effective to improve the beam-energy stability than that of the pressure of a charge-exchange gas.

2. Transmission measurement of cluster ions through a tandem accelerator using a helium gas

One of the crucial points to accelerate cluster ions at a tandem accelerator is the charge exchange from negative to positive by collisions with a target gas at a high voltage terminal. A small amount of cluster ions remains intact and is stripped to positive charged ions. These ratios depend mainly on target gas density and gas species. To obtain intense cluster beam, we compare helium and nitrogen for target gas. we measure transmissions of cluster ions through the tandem accelerator as a function of gas density. Figure 1 shows the transmission of C_8 ions in the terminal voltage of 2.5 MV. The results revealed that helium performs 20 percent better than N₂ as a target gas for a C_8 cluster ion acceleration.

3. Measurement of MeV proton beam emittance

The improvement of the ion beam brightness originated from an emittance of the RF ion source in TIARA 3MV Single-ended Accelerator is an essential factor to form the beam size of less than several hundred nano-meters in diameter in MeV Microbeam System. An emittance monitor with high angular resolution within 1 mrad is required to evaluate the brightness. The emittance monitor²⁾ using luminescence was placed at the SC beam line. As a result, the emittance of 1.7 MeV proton beam was less than 6π mm mrad (MeV)^{1/2}. The emittance of the RF ion source is aiming to be improved in less than 1π mm mrad (MeV)^{1/2}. We are planning to evaluate the beam brightness based on enhancing beam emittance after improving the emittance monitor in the high angular resolution within 0.2 mrad.

4. The prototype of the ion beam intensity distribution monitor for low energy heavy ion beam

The luminescence by ion beam stimulation is a powerful tool to monitor a beam profile of a high energy ion beam, while it is not applied to monitor that of a low energy one because of the low intensity of luminescence. We are developing the ion beam intensity distribution monitor using multi Faraday cup (FC). High aspect ratio FC is necessary to measure a beam current accurately. The prototype monitor consists of twenty one high aspect ratio FCs ($\varphi 2 \times 14 \text{ mm}$) which are covered in 30×30 mm irradiation area with a ceramic support as shown in Fig. 2. The beam profile of 100 keV C_{60}^+ generated with the 400 kV ion implanter was measured, and the profile is plotted as shown in Fig. 3. We will develop high spatial resolution type multi FC and high speed measurement system.







Fig. 2 The prototype Multi FC.

Fig. 3 Beam profile of C_{60}^{+} .

- Y. Ishii, et al., Nucl. Inst. Meth. Phys. Res. B 260 (2007) 15.
- 2) A. Chiba, et al., TIARA annual report 2002 (2003) 327.

4-56 Development of Irradiation Position Control Techniques for Ion Microsurgery Using an Ion Beam Induced Fluorescent Analysis

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The heavy particles have unique characteristics of high energy transfer localized in the vicinity of almost straightforward tracks. The heavy particles are suitable for the treatment of small tumors with the size of several millimeters or less. This research aims at developing an innovative microsurgery treatment system, using the carbon-ion pencil beam, for age-related macular degeneration (AMD).

群馬大学21世紀COEプログラム「加速器テクノロ ジーによる医学・生物学研究」の主要なテーマのひと つとして、原子力機構・高崎研と連携して、高精度炭 素イオンマイクロサージェリー技術の開発を行ってい る。従来のX線やγ線照射などよりも生物への照射効 果の大きな重粒子線を用い、ブラッグピークを利用し て狭小領域の患部に集中的にエネルギー付与し、選択 的かつ集中的に治療を行うことを目指している。

本研究では、難病にも指定されている眼底疾患のひ とつである加齢黄斑変性症を治療想定症例として、高 精度炭素イオンマイクロサージェリー技術の検討を進 めている。黄斑部は網膜の中で最も視神経が集中する 非常に重要な部位であり、この部位周辺に網膜下の狭 小領域に新生血管が発生し網膜を歪めてしまうのが加 齢黄斑変性症(以下AMD)である。症状として、非可逆 的かつ高度な視力低下や視野の中心が歪む変視症の症 状が見られ、最悪の場合、視野の中心が完全に失われ てしまう。推定の患者数は約40万人で、その予備群も 含めると約600万人にもなり、今後、超高齢化社会を 迎える我が国において、無視できない患者数である。

これまで、ビーム径0.1~5 mmφのペンシルビーム を形成するための光学計算、構成機器およびその最適 配置に関する検討を行ってきた。今回、照射位置決め 法として、眼底蛍光造影剤を用いた重イオンビーム誘 起蛍光法を考案し、重イオンビーム誘起蛍光計測シス テムを用いて実験を行うとともに、照射照準位置制御 技術の検討を行った。¹⁾

現在、眼底の狭小・微細領域にある患部を、正確 に3次元精密照射するために必要な照射照準制御技術 の開発が、最大の課題となっている。その方法として、 今年度は眼底患部に注入した眼底蛍光造影剤(インド シアニングリーン;ICG)に、重粒子線を照射し、飛 程をレンジシフタで微調整しながら、ICGの発光を専 用特殊眼底カメラ等を用いて観測し、その発光画像を 解析して照射位置の決定を行うことを検討した。

眼底蛍光造影剤への照射実験は、原子力機構・高崎研 TIARA 施設の AVF サイクロトロン・HA コースおよび HY コースを用いて行った。空気飽和した眼底蛍

光造影剤水溶液(ICG およびフルオレセイン; Fl)に、 炭素イオンビーム($^{12}C^{5+}-220$ MeV)を照射し、眼底 蛍光造影剤水溶液からの発光スペクトルおよび発光 画像を撮影した。サンプルの濃度は標準的な人体に 投与した際の最高血中濃度(1.0×10^{-2} mg/mL, 1.3×10^{-5} M)に調整した。また、サンプル前方にフィルムミ ラーを設置し、ビームをフィルミラーに透過させた 後、フィルミラーに反射した発光画像の撮影を行っ た。

その結果、重粒子線照射による眼底蛍光造影剤からの発光画像および発光スペクトルが取得できた (Fig. 1)。光励起に比べ、発光波長はほぼ同じであるが、 Flの蛍光量子収率(0.98)に対して発光強度が非常に 微弱であるため、発光色素や検出系の再検討が必要で あることが分かった。さらには、サンプル濃度の影響 により、発光の自己吸収と考えられるスペクトル波形 を示した。また、眼底蛍光撮像に向けた発光画像撮影 においても、治療および位置決定用減弱ビーム強度に 対して、撮像が非常に困難であった。今後、さらにカ メラの高感度化と共に、眼底血管を模擬したサンプル を作成し、撮像を行う予定である。



Fig. 1 The fluorescence spectrum of fluorescein in water at 293 K obtained to C⁵⁺ beam irradiation.

Reference

 H. Shimada, *et al.*, JAEA Takasaki Ann. Rep. 2006, JAEA-Review 2007-060 (2008) 191.

5. Status of Irradiation Facilities 2007

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5-01 Safety Measures, Utilization Status and Spread of **Research Results at TIARA Facility**

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Periodical inspections every month and for three months were performed and safety of the facility was also maintained certainly¹⁾. Several safety measures in the TIARA facility, such as the installation of indication light at the entrance to first-class radiation controlled area, the repair of the water sealing at the pipes penetrated into the building, the renewal and disposal of radiation protection materials, and so on were



carried out as shown in Photo. 1.

A central

Renewal of

part of personal access control system, which

Renewal of "RI shoes" (left) and instal-Photo 1 lation of indication light at entrance (right).

is one of the important interlock safety devices, was renewed because of long-term utilization and serious deterioration.

An average count of personal sensing event for one month amounted to 37,000-41,000 for recent a few years. Several PC's and principal software including the OS(LINUX) and control programs were replaced with new ones completely. A pair of PC and LCD is shown in Photo. 2.

The shift of the number of experimental subjects is shown in Fig. 1. The number of common use Photo. 2 for latest two years was multiplied main PC and LCD for about two times. The common use personal access control. is divided into the facility common and the common innovation use which was newly introduced in 2007.

In FY2007, the constitution ratios for internal, common and effective use occupying whole utilization frame had been designed as 70:20:10 comprehensively for 4 accelerators. However, since in this condition the allotment rates for the cyclotron, tandem accelerator and implanter were predicted to decrease largely to less than 50%, these ratios of utilization frame in FY2008 were changed as shown in Table 1.



Fig. 1 Shift of the number of experimental subjects in each research field since 1991.

Table	1	Comparison	of	designed	utilization	frame
be	tween	FY2007 and	FY2	2008.		

\geq	Percentages of designed utilization frame (total 100 %)			
Utilization	JAEA	Common		Effective
frame	internal	Facility common	Common innovation	use
FY2008	> 75	17 - 19		8 - 6
FY2007	> 70	< 20		< 10

The shift of the application and allotment time, their ratio in JAEA internal use for the cyclotron since 1999 is shown in Fig. 2. As a result, the allotment rate through FY2008 could be recovered up to 0.484 and 0.495 for cyclotron and three electrostatic accelerators, respectively.



Fig. 2 Shift of application and allotment time, their ratio in JAEA internal use for the cyclotron since 1999.

The works for utilization and usher's window were smoothly performed similarly to past years. The average number of persons for one month was 47.6 (49.3 for last year) for staying at the visitor's house for outside users, 38.8 (51.5) for the loan of glass budge and ID card, 159 (179) for the temporary visitors in TIARA facility, and so on. The number of carrying-out articles for a month was reduced to 170 from 327 rapidly, because the procedure for article carrying-out was altered strictly. On the other hand, the number of carrying-out for low-level radioactive samples was 26.8 as almost the same as 29.6 in last year.

The second Takasaki Advanced Radiation Research Symposium was held successfully on June 21-22 at Takasaki city gallery. The number of presentations for oral and poster sessions was 22 and 154, respectively. Participants to this symposium were 355, which decreased 57 persons compared to the first symposium.

In addition, the JAEA Takasaki annual report 2006 summarizing research results and activities at TARRI was also issued. This report compiled 187 papers consists of 243 pages including 22 coloured sheets, and was printed as 700 books.

Reference

1) Y. Nakamura et al., JAEA Takasaki annual report 2006 (2007) 203.

5-02 Operation of the AVF Cyclotron

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Operation

The AVF cyclotron was steadily operated throughout fiscal 2007 and supplied various ion beams for research experiments. The cumulative operation time was 53,224 hours and the total number of experiments was 6,710 from the first beam extraction in 1991 to March 2007.

Table 1 shows the statistics of the cyclotron operation of fiscal 2007. The total operation time amounted to 3318.3 hours, and monthly operation times are shown in Fig. 1. The percentages of operation time of the year used for regular experiments, joint-use, the innovation program, beam tuning, and beam development are 72.1%, 4.4%, 1.2%, 21.1%, and 1.3%, respectively. The number of machine troubles was 279. The only trouble that caused a cancellation of experiments was a breakdown of a motor bearing of the cooling system for temperature stabilization of the cyclotron magnet.

Table 1. Statistics for cyclotron operation in	fiscal 2007.
Beam service time	2576.5 hr
Machine tuning	699.2 hr
Beam development	42.6 hr
Total operation time	3318.3 hr
Change of particle and/or energy	270 times
Change of beam course	343 times
Change of harmonic number	57 times
The number of experiments	689
Experiment cancelled due to machine trouble	3 (24hr)

A multi-cusp ion source is used to produce H^+ and D^+ ions. For production of ions heavier than Helium, two ECR ion sources are used alternatively. Table 2 describes the operation time of each ion source. Popularity of major ions used for experiments is shown in Fig. 2. The tendencies of the statistics are similar to those of the past years.

Table 2	Operation	times of ion	sources.

Ion source	Operation time (h)	Ratio(%)
Multi-cusp	1190.6	32.7
ECR (OCTOPUS)	1082.8	29.8
ECR (HYPERNANOGAN)	1366.5	37.5

Maintenance

The regular yearly overhaul and maintenance was carried out. The principal items were as follows: 1) Correction of the original positions of some beam probes, two phase slits, the deflector, etc. 2) Cleaning of the deflector electrodes. 3) Inspection of the RF system and evaluation of its characteristics. 4) Change of lubricating oil for about 50 rotary pumps. 5) Chemical cleaning of the heat



exchanger of the main cooling system. 6) Routine maintenance of the power supplies. 7) Modification of the beam diagnostic station (IS-2) to improve accuracy of reduction ratio of the beam attenuator.

Technical development

The new beam acceleration tests were carried out for 12 MeV H^+ , 18 MeV H^+ and 75 MeV $^{28}Si^{5+}$, and they are ready for use in experiments.

A multipole magnet system for formation of a 2D uniform beam^{1,2)} was installed in the LB beam course and named MuPUS (<u>Multipole magnet beam Profile</u> <u>Uniformization System</u>). Test operation to investigate and improve its performance is under way.

The all-permanent-magnet type ECR ion source $(LECR)^{3}$ was installed to an injection system of the cyclotron and practical operation will start in fiscal 2008.



Fig. 2 Major ion species used for experiments in fiscal 2007.

- Y. Yuri et al., Phys. Rev. ST Accel. Beams 10 (2007) 104001.
- Y. Yuri et al., "Uniform beam formation using multipole magnets", in this annual report (2008).
- 3) K. Yoshida et al., JAEA-Review 2007-060 (2006) 193.

5-03 Operation of the Electrostatic Accelerators

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1. Operation and Status

Three electrostatic accelerators were operated smoothly in FY 2007, and all the planned experiments were carried out except those canceled by users. The yearly operation time of the tandem accelerator, the single-ended accelerator and the ion implanter amounted to 1949, 2437 and 1900 hours respectively, similarly to past years. The total operation time of each accelerator since operation started is 28313, 32022 and 25127 hours respectively. The monthly operation time is shown in Fig. 1. The time is shorter in April, July, August and December than in the other months because of the maintenance. Ion species used for experiments in FY 2007 are shown in Fig. 2.



Fig. 1 Monthly operation time of each accelerator in FY 2007.

2. Maintenance

The number of troubles was smaller than that for past years. A couple of troubles took place in the interface circuits of the safety interlock systems; one was in the circuit between the electro-static accelerators and the radiation control system, the other between the accelerators and the personal access control system. We made it sure that safety was kept under the fail-safe design during the troubles.

The component of high voltage generator of the single-ended accelerator was contaminated by spouted grease from the bearing of the motor-generator drive rod. This was caused due to deterioration of grease containing fluorine with low radiation resistance. The grease worked less than 1000 hours in this case.

The high-pressure residual gas of SF_6 is leaking from the SF_6 gas handling system of the tandem accelerator. The actual spots of leak are not identified. The most amount of the leak comes out when the accelerator tank pressure is high. Therefore, in order to reduce the leak by lowering the residual gas pressure, a pipe was installed to carry the gas in the high-pressure part of the system for the accelerator to the low-pressure storage tank. The sealing rubbers in the system were renewed for leak tight.

3. New Beam Development

As to the ion implanter, Pb ion, which was requested by users, was successfully generated using oven method and accelerated with intensity of 1.5 μ A. We tried to generate Se ion for the tandem accelerator. But the inside of the ion source seriously contaminated by metallic Se, and obtained beam current was not enough to provide to experiments.



Fig. 2 Statistics of ion species used for experiments in FY 2007.

5-04 Operation of the Electron Accelerators and Gamma-ray Irradiation Facilities

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1. Operation

1.1 Electron accelerator

The electron accelerator was on service without serious trouble in $9:00 \sim 17:30$ on Monday and Friday, and in $8:30 \sim 23:00$ on Tuesday to Thursday, to satisfy the demand of operation time for users.

The annual operation time of the electron accelerator, as shown in Fig. 1, is 706.3 h (552.5 h for vertical beams, 153.8 h for horizontal beams), and that of the accelerator No.2 operated until 2004 is also shown in Fig. 1. Total operation time of the accelerator No.1 in FY2007 increased by about 18% compared with that in FY2006, and analysis of the number of research subject indicated the increase of longer-time irradiation for nuclear facilities subject.

The accelerator served mainly for graft-polymerization for new material development, radiation effect study on semiconductors and various experiments of visiting users.



Fig. 1 Shift of annual operation time of electron accelerators.

1.2 Gamma-ray irradiation facilities

The Co-60 gamma-ray irradiation facilities consist of three buildings (8 rooms) and cover a wide dose-rate range from 0.04 Gy/h to 20 kGy/h with eight irradiation cells. The annual operation time for the first, second cobalt irradiation facilities and food irradiation facility is 23,433 h, 9,010 h and 10,631 h, respectively, as shown in Fig. 2.

The first irradiation facility served mainly for radiation-resistance testing of cables used in nuclear power plants and various materials used in J-PARC facility with long irradiation period. The second irradiation facility served mainly for development of new functional materials and other research subject of visiting users, involving irradiation room No.6 operated as hourly scheduled. The food irradiation facility served mainly for development of detection method for irradiated foods and radiation resistance testing at lower dose rates.



Fig. 2 Shift of annual operation time of Co-60 gamma-ray irradiation facilities.

2. Maintenance

2.1 Electron accelerator

The operation of the accelerator was interrupted for 10 days in October, 2007 and 4 days in January 2008 because of periodical maintenance check on accelerator body and incidental equipment, e.g. conveyor and doors of irradiation room and repair of vacuum system at the vertical beam line.

2.2 Gamma-ray irradiation facilities

The periodical maintenance check mainly on mechanical system for radiation source transportation has been performed every three year among three gamma-ray irradiation facilities one by one. The periodical check mainly on interlock system has been performed two times a year for all the facilities. The maintenance check of the food irradiation facility was done in July 2007, with 11 days interruption.

Co-60 sources were purchased and loaded to the irradiation room No.2 in the first irradiation facility to maintain total activities of the room. The old waste sources of about 30 pieces were disposed through The Japan Radioisotope Association (JRIA).

5-05 Utilization of the Electron Accelerator and Gamma-ray **Irradiation Facilities**

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Present status of the utilization of electron accelerator and gamma-ray irradiation facilities

An electron accelerator and three gamma-ray irradiation facilities were operated for various research subjects as operation plan in FY 2007 without serious trouble. Distribution of research subjects and number of experiment subjects are shown in Fig. 1 and Fig. 2, respectively. Table 1 shows irradiation time and the number of research subjects for each facility.

The number of experiment subject at the electron accelerator increased in the research fields of environment and nuclear facilities, and decreased in the field of heat resist. Also, the number at gamma-ray irradiation facilities increased in the field of resource & bio-technology, and decreased in the field of nuclear facilities. The irradiation time in FY2007 of each facility increased in comparison with that in FY2006.

Electron Accelerator

Table 1 Irradiation time and number of experiment subjects in each research field at each facility in FY2007.

Facility	Elect Accele	ron erator	Gamma-ray Irradiation facilities		
Research fields	Irradiation time(hr)	number	Irradiation time(hr)	number	
Material processing	145.5	281	12933	411	
Heat-resist	24.5	8	174	3	
Material for space	140.8	38	7559	57	
Nuclear facilities	145.0	37	53076	113	
Environment	149.1	48	550	74	
Basic technology	70.9	41	5180	45	
Resource & Bio- technology	0	0	2621	276	
Joint use	140.1	52	19859	195	
Total	815.8	505	101951	1174	



Distribution of research subjects (FY 2007). Fig. 1



Gamma-Ray Irradiation Facilities

Gamma-ray Irradiation Facilities

5-06 COMMON USE PROGRAM in Takasaki Advanced Radiation Research Institute

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1. Introduction

The facilities of JAEA are widely opened to users from universities, public institutes, and the industries, etc. COMMON USE PROGRAM, the system of facility use for the user's purpose on fee-charging basis, started since 2006 taking over the former system. There are 3 OPEN USE facilities in Takasaki; Co-60 Gamma-ray Irradiation Facilities, Electron Accelerator and TIARA. In addition to these, some of the off-line devices can be used.

We round up the Research Proposals dividing one year into two terms, and the execution possibility and the validity of the experimental plan are checked by the special committee. The facility usage fee will be the total of the admission fee, irradiation fee and additional charge. In case of Non-proprietary research, the users must report the irradiation result to JAEA, and could be exempt from irradiation fee. JAEA will open the reports to the public. It is also possible for universities to apply from the facility use program of The University of Tokyo. Those applications are accepted as priority case. Table 1 shows the outline of the program.

Table 1	Classification	of COMMON	USE PROGRAM.

Purpose	Research a	Except R&D		
Classifi- cation	Genera	1	Priority case	Commercial
Result	Non- proprietary	Proprietary		
Referee	Yes	No		
Charge*	A		В	С

*A= admission fee

B= admission fee + irradiation fee

C= admission fee + irradiation fee + depreciation

2. Public relations

The information like an outline of this system, guidelines for applicants, format download, etc can be seen on JAEA website (<u>http://www3.tokai-sc.jaea.go.jp/sangaku/</u><u>3-facility/index.html</u>). In addition to this, we go out for publicity to search current needs of the outside of JAEA. In 2007, we participated in International Nanotechnology Exhibition & Conference 2008 (Tokyo), Seminar for advancement of RI/radiation use (Nagoya), Radiation use event applied to daily life (Osaka), and so on.

3. Use in FY2007

There were 35 Research Proposals in FY2007 at

Takasaki Institute, and 22 of all hoped to use with open result. Including the users from priority case, we accepted 300 applications from 52 users and half of the users were new. We can find that the percentage of users from universities exceeds the others except Co-60 Facility from Table 2. However we expect there should be latent needs in the industries. Therefore we are trying to exhume them by the "Public relations" and improve convenience of the usage procedure by introducing new system to increase the industries user.

Figure 1 shows classification of all 300 applications. On TIARA, Non-proprietary use represents more than 50%, while Co-60 Facility and Electron Accelerator have a few. This tendency may come from the user's affiliation.

Table 2 User's affiliation of each facility.

Fac	User eility	Univer- sity	Public Institute	Others	Total
	AVF Cyclotron	7	3	5	15
X	3MV Tandem Accelerator	1	0	0	1
TIA	3MV Single-ended Accelerator	2	0	0	2
	400kV Ion Implanter	5	0	1	6
Co- Irra	-60 Gamma-ray Idiation Facility	7	1	10	18
Ele	ctron Accelerator	6	1	3	10
Tot	al	28	5	19	52



Fig. 1 Percentage of each classification. The percentage is calculated based on the number of applications.

5-07 Radiation Control in TIARA

Safety Section

Department of Administrative Services, TARRI, JAEA

1 Individual monitoring

(1) Individual monitoring for the radiation workers

Table 1 shows a distribution of effective dose of the radiation workers in FY 2007. The effective dose values of almost all radiation workers were below the detection limit (0.1 mSv).

The maximum dose was 0.6 mSv/y due to the overhaul of the TIARA AVF cyclotron.

Table 1 Distributions of the effective dose of the radiation workers in FY 2007.

	porcond	Number	h periods			
Items	persons	l st quarter	2nd quarter	3rd quarter	4th quarter	Total
	$\rm HE < 0.1$	523	539	551	572	693
of effective dose	$0.1 \leq \text{HE} \leq 1.0$	1	7	0	0	8
	$1.0 < \text{HE} \leqq 5.0$	0	0	0	0	0
HE:Effective dose *1	$5.0 < \text{HE} \leqq 15.0$	0	0	0	0	0
(mSv)	15.0 < HE	0	0	0	0	0
Number of persons under radiation control (A)		524	546	551	572	693
Exposure	Number of persons (B)	0	0	0	0	0
above 1mSv	(B)/(A)×100(%)	0	0	0	0	0
Mass effective dose (Person • mSv)		0.1	2.0	0.0	0.0	2.1
Mean dose (mSv)	0.00	0.00	0.00	0.00	0.00	
Maximum dose (ms	Sv)	0.1	0.6	0.0	0.0	0.6

*1 Not detected according to internal exposure.

(2) Individual monitoring for the visitors and others

Table 2 shows number of persons who temporally entered the radiation controlled areas. The effective dose of all persons was less than 0.1 mSv.

Table 2 The number of persons who temporary entered the radiation controlled areas in FY 2007.

Periods	l st quarter	2nd quarter	3rd quarter	4th quarter	Total
Number of persons	504	535	457	409	1905

2 Monitoring of radioactive gas and dust

Table 3 shows the maximum radioactive concentrations and total activities for radioactive gases released from the stack of TIARA, during each quarter of FY 2007.

Small amounts of ⁴¹Ar, ¹¹C and ¹³N were detected for some time during operation of the cyclotron or experiment, but the pulverized substances (⁶⁵Zn, etc.) were not detected.

Table 3 Monitoring results of released radioactive gases and dust in FY 2007.

Nuclide	Items	Periods	1st quarter	2nd quarter	3rd quarter	4th quarter	Total
⁴¹ A r	Maximum cone (Bq/cm ³	centration	<1.5×10 ⁻⁴	<1.5×10 ⁻⁴	2.4×10 ⁻⁴	1.5×10 ⁻⁴	2.4×10 ⁻⁴
л	Activity ((Bq)	5.6×10 ⁸	2.0×10 ⁸	1.2×10 ⁹	9.5×10 ⁸	2.9×10 ⁹
¹¹ C	Maximum con (Bq/cm ³	centration	<1.5×10 ⁻⁴				
C	Activity ((Bq)	1.2×10 ⁸	6.0×10 ⁷	2.9×10 ⁸	1.9×10 ⁸	6.6×10 ⁸
¹³ N	Maximum con (Bq/cm ³	centration	<1.5×10 ⁻⁴				
.,	Activity ((Bq)	7.0×10 ⁸	6.2×10 ⁷	1.7×10 ⁸	8.8×10 ⁶	9.4×10 ⁸
⁶⁵ Zn	Maximum cone (Bq/cm ³	centration	<6.0×10 ⁻¹⁰	<6.2×10 ⁻¹⁰	<6.7×10 ⁻¹⁰	<6.6×10 ⁻¹⁰	<6.7×10 ⁻¹⁰
2.11	Activity ((Bq)	0	0	0	0	0

3 Monitoring for external radiation and surface contamination

External radiation monitoring was routinely carried out in/around the radiation controlled areas and surface contamination monitoring was also carried out. Neither unusual value of dose equivalent rate nor surface contamination was detected.

Figure 1 shows a typical example of distribution of the dose equivalent rate at the radiation controlled area in the cyclotron building.



Fig. 1 Dose equivalent rate distribution at the radiation controlled area in the cyclotron building. Measurement date: 26th March 2008,

Measuring position: Indicated with $\times 1$ m above floor, Unit: μ Sv/h. (The values are not indicated at the places where the dose equivalent rates are less than 0.2 μ Sv/h).

5-08 Radioactive Waste Management in TIARA

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1. The radioactive wastes management

The radioactive wastes generated in TIARA are managed by Utilities and Maintenance Section. The main radioactive wastes are the solid wastes generated by the research experiment and the maintenance of the cyclotron. Other radioactive wastes are the liquid wastes such as inorganic waste fluids generated from the research experiment and the air-conditioning machines in controlled area. These wastes are managed according to properties.

2. Solid radioactive waste

Table 1 shows the amounts of solid wastes at various properties and kinds generated in each quarter of FY 2007. The main solid waste is generated by the research experiment and the maintenance of the cyclotron.

Combustible wastes are rubber gloves, paper, and clothes, etc. Incombustible wastes are metal pieces, the glasses, and contaminated parts.

3. Liquid radioactive waste

Table 2 shows the amounts of liquid wastes generated in each quarter of FY 2007. Liquid waste was almost inorganic waste water generated with chemical experiments and operation of air conditioning units installed in each room of the first class radiation controlled area. Larger quantity of waste water in summer season (2nd quarter) is mainly due to condensed water, which is treated by evaporation, and inorganic water is reused in the controlled area. Only small amounts of concentrated liquid are generated by the evaporation.

Table 1 Radioactive solid wastes generated in FY 2007.

	Amounts	Amo	Amounts of generation in each periods (m ³)					
Items		1 st quarter	2nd quarter	3rd quarter	4th quarter	Total	package /drum	
Category A*	¢	0	0.50	0.28	0.38	2.34		
1) Combus	tible	0	0.40	0.20	0.56	1.16	7**	
2) Incombustible		0	0.10	0.10	0.98	1.18	0	
Compress	sible	0	0.10	0.10	0.24	0.44	3**	
Filters		0	0	0	0.74	0.74	0	
Incompre	ssible	0	0	0	0	0	0	
Ion excha	nge resin	0	0	0	0	0	0	
Category B*	<	0	0	0	0	0	0	
* 1.0								

* defined by dose at the outer surface of container : (A) $\leq 2 \text{ mSv/h} \leq$ (B).

** 200-liter drum

Table 2 Radioactive liquid waste generated in FY 2007.

Category A*	10.88	20.44	5.60	3.38	40.30	
1) Inorganic	10.88	20.44	5.60	3.18	40.10	treatment
2) Organic	0	0	0	0	0	0
Organic	0	0	0	0	0	0
Oil	0	0	0	0	0	0
3) Sludge	0	0	0	0	0	0
4) Evaporation residue	0	0	0	0.20	0.20	2
Category B*	0	0	0	0	0	0

* defined by concentrations in Bq/cm³ (β , γ): (A) < 3.7×10 \leq (B) < 3.7×10⁴.

Appendix

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Appendix 1. List of Publication

A 1.1 Publications in Journal

07J001 1-02, 1-03 C, I

Y. Takahashi, T. Ohki, T. Nagasawa,

Y. Nakajima, R. Kawanabe, K. Ohnishi, T. Hirao,S. Onoda, K. Mishima, K. Kawano and H. Itoh,

"Heavy-ion induced current through an oxide layer"

Nucl. Instrum. Meth. Phys. Res., Vol.B-260, No.1 (2007) 309.

07J002 1-07

H. Okada, Y. Nakanishi, A. Wakahara,
A. Yoshida and T. Ohshima,
"380 keV proton irradiation effects on photoluminescence of Eu-doped GaN"
Nucl. Instrum. Meth. Phys. Res., B 266 (2008) 853–856.

I

07J003 1-07

A. Wakahara, K. Takemoto, F. Oikawa,
H. Okada, T. Ohshima, and H. Itoh,
"Investigation of Tb related green emission in group III nitrides by time resolved photoluminescence measurements"
Phys. Status Solidi, (a) 205(1) (2008) 56–59.

I

07J004 1-08 E

G. Pensl, F. Schmid, S.A. Reshanov, H.B. Weber,
M. Bockstedte, A. Mattausch, O. Pankratov,
T. Ohshima and H. Itoh,
"(Nitrogen-Vacancy)-Complex Formation in SiC: Experiment and Theory"
Mater. Sci. Forum, 556-557 (2007) 307-312.

07J005 1-08 I

K. K. Lee, M. Laube, T. Ohshima,
H. Itoh and G. Pensl,
"Hall Effect and Admittance Measurements of n-channel 6H-SiC MOSFETs"
Mater. Sci. Forum, 556-557 (2007) 791-794.

07J006 1-08 T, I

T. Ohshima, T. Satoh, M. Oikawa, S. Onoda,
S. Hishiki, T. Hirao, T. Kamiya, T. Yokoyama,
A. Sakamoto, R. Tanaka, I Nakano,
G. Wagner and H. Itoh,
"Degradation of Charge Collection Efficiency Obtained for 6H-SiC n+p Diodes Irradiated with Gold Ions"
Mater. Sci. Forum, 556-557 (2007) 913-916.

07J007 1-08 T, I

S. Onoda, T. Ohshima, T. Hirao, K. Mishima,
S. Hishiki, N. Iwamoto and K. Kawano,
"Impact of auger recombination on charge collection of a 6H-SiC diode by heavy ions"
IEEE Trans. Nucl. Sci., 54 (2007) 2706-2713.

07J008 1-08 T, I

- S. Onoda, T. Ohshima, T. Hirao, K. Mishima,
- S. Hishiki, N. Iwamoto,

K. Kojima and K. Kawano,

"Decrease of charge collection due to displacement damage by gamma rays in a 6H-SiC diode"

IEEE Trans. Nucl. Sci., 54 (2007) 1953-1960.

07J009 1-08 E

L. Storasta, H. Tsuchida,
T. Miyazawa and T. Ohshima,
"Enhanced annealing of the Z1/2 defect in 4H–SiC epilayers"
J. Appl. Phys., 103 (2008) 013705-1 – 7.

07J010 1-10 E

H. Matsuura, N. Minohara, Y. Inagawa,

M. Takahashi, T. Ohshima and H. Itoh,

"Mechanisms of Decrease in Hole Concentration in Al-doped 4H-SiC by Irradiation of 200 keV Electrons"

Mater. Sci. Forum, 566-557 (2007) 379-382.

07J011 1-11 T, E, G

J. Isoya, T. Umeda, N. Mizuochi, N.T. Son,
E. Janzén and T. Ohshima,
"EPR identification of intrinsic defects in SiC"
Phys. Status Solidi, b 245, 1298-1314 (2008)

07J012 1-11 E

T. Umeda, J. Isoya, T. Ohshima, N. Morishita,H. Itoh and A. Gali,

"Identification of positively charged carbon vacancy antisite-vacancy pairs in *4H*-SiC" Phys. Rev., B 75, 245202 (1-5) (2007)

07J013 1-11 E

N. T. Son, I. G. Ivanov, A. Kuznetsov,
B. G. Svensson, Q. X. Zhao, M. Willander,
N. Morishita, T. Ohshima, H. Itoh, J. Isoya,
E. Janzén and R. Yakimova,
"Recombination centers in as-grown and electron-irradiated ZnO substrates"
J. Appl. Phys., 102, 093504(1-5) (2007)

07J014 1-13 G

武田信和、角舘聡、中平昌隆、柴沼清、 "核融合炉機器の遠隔保守に関する開発の 現状" プラズマ・核融合学会誌 Vol. 84, No. 2 (2008) 100-107.

07J015 1-25 T

Y. Miwa, Y. Kaji, N. Okubo,
K. Kondo and T. Tsukada,
"New Evaluation Method of Material Degradation Considering Synergistic Effects of Radiation Damage"
J. Solid Mech. Mater. Eng., vol.2 (2008) 145

07J016 1-25 T

N. Okubo, Y. Miwa, K. Kondo,
Y. Kaji and T. Tsukada,
"Effect of residual stress on radiation hardening in stainless steels"

J. Nucl. Mater., to be published.

07J017 1-29 G

J. Chen, M. Asano, Y. Maekawa, T. Sakamura,
H. Kubota and M. Yoshida,
"Preparation of ETFE-based fuel cell membranes by UV-induced photografting and electron beam-induced crosslinking techniques"
ESC Trans., 3(1) (2006) 103-113.

07J018 1-29 G

J. Chen, M. Asano, Y. Maekawa and M. Yoshida, "Polymer electrolyte hybrid membranes prepared by radiation grafting of styryltrimethoxysilane into ETFE films"

J. Membr. Sci., 296 (1-2) (2007) 77-82.
07J019 1-29 G

M. Asano, J. Chen, Y. Maekawa, T. Sakamura,
H. Kubota and M. Yoshida,
"Novel UV-induced photografting process for preparing PTFE-based proton-conducting membranes"
J. Polym. Sci. A:Polym Chem., 45 (2007)

2624-2637.

07J020 1-30 G

J. Chen, M. Asano, Y. Maekawa and M. Yoshida, "Double crosslinked polyetheretherketone-based polymer electrolyte membranes prepared by radiation and thermal crosslinking techniques" Polymer, 48(20) (2007) 6002-6009.

07J021 1-31 C

T. Yamaki Y. Kozone, A. Hiroki, K. Hosoi,
M. Asano, H. Kubota and M. Yoshida,
"Proton exchange membranes for fuel cell applications prepared by ion track technology" Electrochemistry, 75 (2007) 175.

07J022 1-31 C

八巻徹也、R. Rohani, 越川博、高橋周一、
長谷川伸、浅野雅春、K.-O. Voss、
R. Neumann、前川康成、
"イオンビーム照射ポリフッ化ビニリデン薄膜の エッチング挙動 ーエッチング前処理・照射 イオン効果の検討"
高分子論文集, 65 (2008) 273.

07J023 2-04 E
N. Ito, A. Mase, Y. Kogi, Z. Shen,
N. C. Luhmann, Jr. N. Seko,
M. Tamada and E. Sakata,
"Advanced Fabrication Method of Planar

Components for Plasma Diagnostics" J. Plasma Fusion Res., Vol. 2, (2007) S1042_1-S1042_4.

07J024 2-04 E

N. Ito, A. Mase, Y. Kogi, N. Seko,
M. Tamada and E. Sakata,
"Development of Millimeter-Wave Planar Devices Using Low-Loss Substrates"
Kyushu Univ. Sourikou-Rep. (in press)

07J025 2-04 E

N. Ito, A. Mase, Y. Kogi, N. Seko,
M. Tamada and E. Sakata,
"New Advanced Fabrication Technique for the Millimeter-wave Planar Components based on Fluororesin Substrates using Graft Polymerization"
Jpn. J. Appl. Phys., Vol. 47, No.6 (in press)

07J026 2-04 E

N. Ito, A. Mase, Y. Kogi, N. Seko,
M. Tamada and E. Sakata,
"Development of Planar Components Using Advanced Fabrication"
Dig. IEEE Joint 32nd Int. Conf. Infrared
Millimeter Waves, 15th Int. Conf. Terahertz
Electron. 2007, Vol. 2, (2007) 791-792

07J027 2-06 E, G

M. Takigami, H. Amada, N. Nagasawa,
A. Hiroki, N. Kasai, F. Yoshii and M. Tamada,
"Absorption of Phosphate Ion in Swine Urine Using CMC Gel"
Trans. Mater. Res. Soc. Jpn., in press

07J028 2-07 E, G

T. Kasahara, M. Takigami, N. Nagasawa,
P. Prawitwong and S. Takigami,
"Preparation and Characterization of CMC-Konjac Mannan Mixture gel Trans Mater"
Trans. Mater. Res. Soc. Jpn., in press

07J029 2-10 E

T. Hakoda, K. Matsumoto, A. Shimada, T. Narita, T. Kojima and K. Hirota,

"Application of ozone decomposition catalysts to electron-beam irradiated xylene/air mixtures for enhancing carbon dioxide production" Radiat. Phys. Chem., 77 (2008) 585

07J030 2-11 G

A. Kimura, M. Taguchi, Y. Ohtani, Y. Shimada,H. Hiratsuka and T. Kojima,"Treatment of wastewater having estrogen activity by ionizing radiation"

Radiat. Phys. Chem., 76 (2007) 699-706.

07J031 2-12 G

Hoang Hoa Mai, H. M. Solomon,
M. Taguchi and T. Kojima,
"Polybutyral films containing leuco-malachite green as low dose dosimers"
Radiat. Phys. Chem., 77 (2008) 457-462.

07J032 3-02 G

S. Takahashi, A.N. Sakamoto,
A. Tanaka and K. Shimizu,
"AtREV1, a Y-family DNA polymerase in Arabidopsis, has deoxynucleotidyl transferase activity in vitro"
Plant Physiol., 145 (2007) 1052.

07J033 3-26 C

A. Urushibara, N. Shikazono, P. O'Neill, K. Fujii,
S. Wada and A. Yokoya,
"LET dependence of the yield of single-, double-strand breaks and base lesions in fully hydrated plasmid DNA films by ⁴He⁽²⁺⁾ ion irradiation"

Int. J. Radiat. Biol., 84 (2008) 23-33.

07J034 3-31 G

鵜飼光子、"総説 食品の放射線照射の意義と課題"電気評論、6 (2007) 7-12.

07J035 3-31 G

Y. Shimoyama, M. Ukai and H. Nakamura,"Advanced protocol of Detection for Irradiated Food by ESR"Radiat. Phys. Chem., 76 (2007) 1837-1839.

07J036 3-31 G
亀谷宏美、鵜飼光子、
"酸素フリー雰囲気での ESR によるγ線照射で 衛生化したアガリクスの分析"
RADIOISOTOPES, 56 (2007) 437-441.

07J037 3-31 G 松浦昌彰、小川聡子、大和田滋、鵜飼光子、 "X-band ESR を用いた殺菌胡椒のラジカルの 評価" 食品照射、42 (2007) 9-13.

07J038 3-31 G 亀谷宏美、加恵田庸子、中村秀夫、鵜飼光子、 "二種のグルコースポリマーの照射処理により 新規に誘導されるラジカルの解析" 食品照射、42 (2007) 4-8.

07J039 3-31 G

鷲野郁海、尾崎文彦、鵜飼光子、 "若者のくらしと放射線に関する意識調査" 食品照射、42 (2007) 24-33.

07J040 3-32 G

M. Kikuchi, C. R. Gunawardane, Md. K. Alam,
A. Z. Mohd. Dzomir, R. C. Pitipanaarachchi,
T. Funayama, N. Hamada, T. Sakashita, S. Wada,
K. Satoh, I. Narumi and Y. Kobayashi,
"Chemiluminescence ELISA for the Detection of Oxidative DNA Base Damage Using Anti-8-hydroxy-2'-deoxyguanosine Antibody: Application to the Detection of Irradiated Foods" RADIOISOTOPES, 56 (2007) 509-517.

07J041 3-33 C, G

横田裕一郎、井上雅好、鳴海一成、舟山知夫、 小林泰彦、田中淳、 "高等植物の放射線耐性メカニズム ータバコ BY-2 培養細胞における DNA 損傷と その修復から見えてくることー" 放射線生物研究, 42 (2007) 402-415.

07J042 3-34 G

T. Sakashita, N. Hamada, D.D. Ikeda, S. Yanase,M. Suzuki, N. Ishii and Y. Kobayashi,

"Modulatory effect of ionizing radiation on food-NaCl associative learning: the role of gamma subunit of G protein in Caenorhabditis elegans"

FASEB J. 22 (2008) 713.

07J043 3-34 G

T. Sakashita, N. Hamada, M. Suzuki, D.D. Ikeda,S. Yanase, N. Ishii and Y. Kobayashi,"Effects of γ-ray irradiation on olfactory

adaptation to benzaldehyde in Caenorhabditis elegans" Biol. Sci. in Space 21 (2007) 117.

07J044 3-35 C

T. Funayama, S. Wada, Y. Yokota, K. Fukamoto,

T. Sakashita, M. Taguchi, T. Kakizaki,

N. Hamada, M. Suzuki, Y. Furusawa,

H. Watanabe, K. Kiguchi and Y. Kobayashi,"Heavy-ion microbeam system in JAEA for microbeam biology"J. Radiat. Res., 49 (2008) 71.

07J045 3-35 C

T. Funayama, N. Hamada,
T. Sakashita and Y. Kobayashi,
"Heavy-ion microbeams - development and applications in biological studies"
IEEE Trans. Plasma Sci., 36 (2008) in press.

07J046 3-36 C

N. Hamada, H. Matsumoto,
T. Hara and Y. Kobayashi,
"Intercellular and intracellular signaling pathways mediating ionizing radiation-induced bystander effects"
L. Badiat, Bas., 48 (2007) 87.

J. Radiat. Res., 48 (2007) 87.

07J047 3-36 C

N. Hamada, T. Hara, T. Funayama,
T. Sakashita and Y. Kobayashi,
"Energetic heavy ions accelerate differentiation in the descendants of irradiated normal human diploid fibroblasts"
Mutat. Res., 637 (2008) 190.

07J048 3-36 C

N. Hamada, M. Ni, T. Funayama,

T. Sakashita and Y. Kobayashi,

"Temporally distinct response of irradiated normal human fibroblasts and their bystander cells to energetic heavy ions"

Mutat. Res., 639 (2008) 35.

07J049 3-36 C

N. Hamada, T. Hara, M. Omura-Minamisawa,

T. Funayama, T. Sakashita, S. Sora, Y. Yokota,

T. Nakano and Y. Kobayashi,

"Energetic heavy ions overcome tumor radioresistance caused by overexpression of Bcl-2"

Radiother. Oncol. (2008) in press (doi: 10.1016/j.radonc.2008.02.013).

07J050 3-36 C

N. Hamada, T. Hara, M. Omura-Minamisawa,
T. Funayama, T. Sakashita, S. Sora,
T. Nakano and Y. Kobayashi,
"The survival of heavy ion-irradiated Bcl-2 overexpressing radioresistant tumor cells and their progeny"
Cancer Lett. (2008) in press
(doi: 10.1016/j.canlet.2008.03.029).

07J051 3-36 C

M. Iwakawa, N. Hamada, K. Imadome,
T. Funayama, T. Sakashita,
T. Imai and Y. Kobayashi,
"Expression profiles are different in carbon ion-irradiated normal human fibroblasts and their bystander cells"
Mutat. Res. (2008) in press (doi: 10.1016/j.mrfmmm.2008.04.007).

07J052 3-36 C

T. Funayama, N. Hamada,
T. Sakashita and Y. Kobayashi,
"Heavy-ion microbeams – Development and applications in biological studies"
IEEE Trans. Plasma Sci., 36 (2008) in press.

07J053 3-39 C

T. Oishi, A. Sasaki, N. Hamada, S. Ishiuchi,

- T. Funayama, T. Sakashita, Y. Kobayashi,
- T. Nakano and Y. Nakazato,
- "Proliferation and cell death of human glioblastoma cells after carbon-ion beam exposure: morphologic and morphometric analyses" Neuropathology (in press)

C, T

07J054 3-41

M. Hino, S. Wada, Y. Tajika, Y. Morimura,
N. Hamada, T. Funayama, T. Sakashita,
T. Kakizaki, Y. Kobayashi and H. Yorifuji,
"Heavy ion microbeam irradiation induces ultrastructural changes in isolated single fibers of skeletal muscle"
Cell Struct. Funct., 32 (2007) 51-56.

07J055 3-42 O

Al-Jahdari WS, Y. Suzuki, Y. Yoshida, et al."Growth cone collapse and neurite retractions: An approach to examine X-irradiation affects on neuron cells"

J. Radiat. Res. (2008); in press

07J056 3-43 C

C. Shao, Y. Furusawa, Y. Matsumoto, Y. Pan,P. Xu and H. Chen,"Effect of gap junctional intercellular

communication on radiation responses in neoplastic human cells" J. Radiat. Res., 167 (2007) 283-288

07J057 3-43 C

T. Konishi, K. Amemiya, T. Natsume,A. Takeyasu, N. Yasuda,Y. Furusawa and k. Hieda,"A New Method for the Simultaneous Detection of Mammalian Cells and Ion Tracks on a Surface of CR-39."

J. Radiat. Res., 48(3) (2007) 255-261.

07J058 3-43, 3-48 C

T. Funayama, S. Wada, Y. Yokota, K. Fukamoto, T. Sakashita, M. Taguchi, T. Kakizaki,

N. Hamada, M. Suzuki, Y. Furusawa,

H. Watanabe, K. Kiguchi and Y. Kobayashi,

"Heavy-ion microbeam system at JAEA-Takasaki for microbeam biology"

J. Radiat. Res., 49(1) (2008) 71-82.

07J059 3-44 C

A. Takahashi, H. Matsumoto and T. Ohnishi,
"Hdm2 and nitric oxide radicals contribute to the p53-dependent radioadaptive response"
Int. J. Radiat. Oncol. Biol. Phys., in press.

07J060 3-44 C

H. Matsumoto,

"Revisiting sensitization mechanisms in cancer thermochemotherapy:does the production of radicals hold the key to sensitization?" Therm. Med., 24 (2008) 13.

07J061 3-44 C

H. Matsumoto, A. Takahashi and T. Ohnishi,

"Nitric oxide radicals choreograph a radioadaptive response" Cancer Res., 67 (2007) 8574.

07J062 3-44 C

H. Matsumoto, H. Hamada, A. Takahashi,
Y. Kobayashi and T. Ohnishi,
"Vanguards of paradigm shift in radiation biology: Radiation-induced adaptive and bystander responses"
J. Radiat. Res., 48 (2007) 97.

07J063 3-44 C

N. Hamada, H. Matsumoto,
T. Hara and Y. Kobayashi,
"Intercellular and intracellular signaling pathways mediating ionizing radiation-induced bystander effects"

J. Radiat. Res., 48 (2007) 87.

07J064 3-48 C

K. Fukamoto, K. Shirai, T. Sakata, T. Sakashita,
T. Funayama, N. Hamada, S. Wada, T. Kakizaki,
S. Shimura, Y. Kobayashi and K. Kiguchi,
"Development of the irradiation method for the first instar silkworm larvae using locally targeted heavy-ion microbeam"
J. Radiat. Res., 48 (2007) 247-253.

07J065 3-49 C

Y. Miyazawa, T. Sakashita, T. Funayama,

- N. Hamada, H. Negishi, A. Kobayashi,
- T. Kaneyasu, A. Ooba, K. Morohashi,
- T. Kakizaki, S. Wada, Y. Kobayashi,
- N. Fujii and H. Takahashi,
- "Effects of Locally Targeted Heavy-ion and Laser Microbeam on Root Hydrotropism in

Arabidopsis thaliana" J. Radiat. Res. in press doi: 10.1269/jrr.07131

07J066 3-50 O

T. Kimura, K. Takahashi, Y. Suzuki, Y. Konishi,
Y. Ota, C. Mori, T. Ikenaga, T. Takanami,
R. Saito, E. Ichiishi, S. Awaji,
K. Watanabe and A. Higashitani,
"The Effect of High Strength Static Magnetic Fields and Ionizing Radiation on Gene Expression and DNA Damage in Caenorhabditis

elegans"

Bioelectromagnetics (2008) in press.

07J067 3-51 C

N. Kawachi,

"Functional Diagnosis of Photosynthesis in a Leaf -Quantitative Kinetic Analysis of Photosynthetic Functions Using Positron Emitting Tracer Imaging System (PETIS)-"

JAEA R&D Review 2007, vol. 2 (March 2008) 48 (未来を拓く原子力 第2号 頁48 2007年)

07J068 3-51 C

K. Kikuchi, S. Ishii, S. Fujimaki, N. Suzui,

S. Matsuhashi, I. Honda,

Y. Shishido and N. Kawachi,

"Real Time Analysis of Photoassimilate Translocation in Intact Eggplant Fruit using ¹¹CO₂ and a Positron-Emitting Tracer Imaging System"

J. Jpn. Soc. Hort. Sci., vol. 77, no. 2 (2008) 199-205.

07J069 3-51, 3-52 C

N. Kawachi, S. Fujimaki, K. Sakamoto,N. S. Ishioka, S. Matsuhashi and H. Sekimoto,

"Analysis of NO₃ interception of the parasitic angiosperm Orobanche spp.by using a Positron-Emitting Tracer Imaging System and ¹³NO₃: A new method for visualization and quantitative analysis of the NO₃ interception ratio"

Soil Sci. Plant Nutr., vol. 54 (2008) 408-416.

07J070 3-51 C

河地有木、
"植物分子イメージングによる光合成機能の
可視化"
光合成研究 第17巻 第3号 頁74-78 2007年
12月

07J071 3-53 C

R. Suwa, S. Fujimaki, N Suzui, N Kawachi,
S Ishii, K Sakamoto, N T Nguyen, H Saneoka,
P K Mohapatra, R E. Moghaieb,
S Matsuhashi and K Fujita,
"Use of positron emitting tracer imaging system for measuring the effect of salinity on temporal and spatial distribution of ¹¹C tracer and coupling between source and sink organs"
Plant Sci. (2008) in press

07J072 3-54 C

K. Kikuchi, S. Ishii, S. Fujimaki, N. Suzui, S. Matsuhashi, I. Honda,

Y. Shishido and N. Kawachi,

"Real-Time Analysis of Photoassimilate

Translocation in Intact Eggplant Fruit using

¹¹CO₂ and a Positron-Emitting Tracer Imaging System"

J. Jpn. Soc. Hort. Sci., 77 (2008) 199-205.

07J073 3-56 C

中村進一、茅野充男、藤巻秀、鈴井伸郎、 石岡典子、河地有木、松橋信平、 "土壌中からのカドミウムの除去技術(ファイトレメ ディエーション)の実用化を目指した研究におけ るポジトロンイメージング技術の貢献" 放射線と産業, 117 (2008) 9-14.

07J074 3-59 O

S. Watanabe, T. Katabuchi, N. S. Ishioka,
S. Matsuhashi and H. Muramatsu,
"Production of Endohedral ¹³³Xe-Higher Fullerenes by Ion Implantation"
J. Radioanal. Nucl. Chem., 272 (2007) 467–469.

07J075 3-59 O

S. Watanabe, T. Katabuchi,
N. S. Ishioka and S. Matsuhashi,
"Synthesis of Endohedral ¹³³Xe-Fullerenol by Using Higher Fullerene"
J. Nucl. Radiochem. Sci., 8 (2007) 105-107.

07J076 3-61 S

S. Harada, S. Ehara, K. Ishii, H. Yamazaki,
S. Matsuyama, T. Kamiya, T. Sakai, T. Sato,
S. Oikawa, K. Sera and J. Ito,
"Optimizing the radiosensitive liquid-core microcapsules for the targeting of chemotherapeuric agents"
Nucl. Instrum. Meth. Phys. Res., B 260 (1) (2007) 164

07J077 3-61 S

S. Harada, S. Ehara, K. Ishii, H. Yamazaki,S. Matsuyama, T. Kamiya, T. Sakai, T. Sato,S. Oikawa, K. Sera and J. Ito,"Improvement of radiosensitive liquid-core"

microcapsules by Yttrium polymerization" Int. J. PIXE, Vol 17, Nos. 1&2 (2007) 33

07J078 3-63 S

T. Nagamine, H. Takada, T. Kusakabe,
K. Nakazato, T. Sakai, M. Oikawa, T. Kamiya,
K. Arakawa and M. Iha,
"Intracellular Changes of Metal Elements by Fucoidan Extracted from Brown Seaweed (Cladosiphon okamuranus)"
Biol. Trace Elem. Res. (2008) May 13.

07J079 3-63 S

T. Nagamine, K. Nakazato,K. Suzuki,
T. Kusakabe, T. Sakai, M. Oikawa, T. Satoh,
T. Kamiya and K. Arakawa,
"Analysis of tissue cadmium distribution in chronic cadmium-exposed mice using in-air micro-PIXE"
Biol. Trace Elem. Res. (2007) Summer;117(1-3)
115-26.

07J080 3-63 S

T. Kusakabe, K. Nakajima, K. Suzuki,

- K. Nakazato, H. Takada, T. Satoh , M. Oikawa,
- K. Kobayashi, H. Koyama,
- K. Arakawa and T. Nagamine,

"The changes of heavy metal and metallothionein distribution in testis induced by cadmium exposure"

Biometals. (2008) Feb;21(1) 71-81.

07J081 3-63 S

K. Nakazato, T. Nagamine, K. Suzuki,T. Kusakabe, HD. Moon, M. Oikawa,T. Sakai and K. Arakawa,"Subcellular changes of essential metal shown by

in-air micro-PIXE in oral cadmium-exposed mice" Biometals. (2008) Feb;21(1) 83-91.

07J082 3-66

K. Ishii, H. Yamazaki, Y. Kikuchi, K. Inomata,
Y. Watanabe, A. Ishizaki, R. Oyama,
Y. Kawamura, T. Yamaguchi, G. Momose,
M. Nagakura and M. Takahashi,
"PROGRESS AND APPLICATION OF THE TOHOKU MICROBEAM SYSTEM"
Nucl. Instrum. Meth. Phys. Res., B,260 (2007)
55-64.

07J083 3-66

K. Ishii, Y. Kikuch, S. Matsuyama, Y. Kanai,

K. Kotani, T. Ito, H.Yamazaki, Y. Funaki,

R. Iwata, M. Itoh, K.Yanai, J.Hatazawa, N.Itoh,

N. Tanizaki, D. Amano,

M. Yamada and T. Yamaguchi,

"First achievement of less than 1 mm FWHM resolution in practical semiconductor animal PET scanner"

Nucl. Instrum. Meth. Phys. Res., A,576 (2007) 435-440.

07J084 3-66

K. Ishii, S. Matsuyama, Y. Watanabe,
Y. Kawamura, T. Yamaguchi, R.Oyama,
G. Momose, A.Ishizaki,
H. Yamazaki and Y. Kikuchi,
"3D-Imaging using micro-PIXE"
Nucl. Instrum. Meth. Phys. Res., A,571 (2007)
64-68.

07J085 3-66

H. Dai, K. Kaneko, H. Kato, S. Fujii, Y. Jing,

E. Sakurai, M. Kato, N. Okamura,
A. Kuramasu and K. Yanai,
"Selective cognitive dysfunction in mice lacking histamine H1 and H2 receptors"
Neurosci. Res. (2007) 57(2):306-13

07J086 4-01 S

A. Inouye, S. Yamamoto, S. Nagata,
M. Yoshikawa and T. Shikama
"Hydrogen Behavior in Gasochromic Tungsten Oxide Films Investigated by Elastic Recoil Detection Analysis"
Nucl. Instrum. Meth. Phys. Res., B 266 (2008) 301-307.

07J087 4-01 S

S. Yamamoto, A. Inouye and M. Yoshikawa
"Structural and Gasochromic Properties of Epitaxial WO3 Films Prepared by Pulsed Laser Deposition"
Nucl. Instrum. Meth. Phys. Res., B 266 (2008)
802-806.

07J088 4-05 C

M. Sugimoto, M. Yoshikawa,
S. Tukuda and S. Seki,
"Synthesis of Ceramic Nano fiber from Precursor Polymers by Ion Beam Irradiation"
Trans. Mater. Res. Soc. Jpn. (submitted)

07J089 4-06 C

S. Seki, S. Tsukuda, M. Sugimoto and S. Tagawa,
"Cross-Linked Silicon Based Polymer Nanowire Formation by High Energy Charged Particles"
Surf. Coat. Technol., 201 (2007) 8486-8489.

07J090 4-06 C

S. Seki, S. Tsukuda, M. Sugimoto and S. Tagawa,
"Cross-Linked Silicon Based Polymer Nanowire Formation by High Energy Charged Particles"
Surf. Coat. Technol., 201 (2007) 8486-8489.

07J091 4-06 C

S. Tsukuda, S. Seki, M. Sugimoto and S. Tagawa,
"Effect of Ion Beam Energy and Polymer Weight on the Thickness of Nanowires Produced by Ion Bombardment of Polystyrene Thin Films"
Surf. Coat. Technol., 201 (2007) 8526-8530.

07J092 4-06 C

S. Tagawa, S. Tsukuda, S. Seki, M. Sugimoto,
T. Sato, M. Oikawa and T. Sakai,
"Ion Beam Induced Nano-Space Reactions and Nanowire Formation in Polymers by High Energy Sub- μ m Heavy Ion Beams"

Surf. Coat. Technol., 201 (2007) 8495-8498.

07J093 4-06 C

Y. Koizumi, S. Seki, A. Saeki and S. Tagawa,"Electronic Properties of the Charge Carriers on Oligofluorene Backbone"Radiat. Phys. Chem., 76 (2007) 1337-1341.

07J094 4-08 C

Y. Suzuki, T. Yamaki, H. Koshikawa. M. Asano,
K.-O. Voss, R. Neumann and M. Yoshida,
"Preparation of ion-track membranes of poly(p-phenylene terephthalamide): Control of pore shape by irradiation with different ion beams"
Nucl. Instrum. Meth. Phys. Res., B 260 (2007)

693.

07J095 4-08 C

T. Yamaki, M. Asano, Y. Maekawa, Y. Suzuki,
M. Yoshida, R. Neumann and K.-O. Voss,
"Preparation of Ion-Track Membranes of Poly(p-phenylene terephthalamide)"
GSI Sci. Rep., 2006 (2007) 322.

07J096 4-08 C, I

H. Koshikawa, Y. Maekawa and H. Usui,
"Formation of composite films of ion-track membranes embedded with oblique Cu nanowires for anisotropic infrared absorption"
Radiat. Phys. Chem., 77 (2008) 453.

07J097 4-09 I

Y. Kasukabe, S. Nishida, S. Yamamoto,
M. Yoshikawa and Y. Fujino,
"Atomistic Nitriding Processes of Titanium Thin Films due to Nitrogen-Implantation"
Appl. Surf. Sci., (2008) in press.

07J098 4-12 I

K. Takahiro, K. Kawatsura, S. Nagata,B. Tsuchiya, S. Yamamoto and H. Naramoto,"Improvement in crystal quality of epitaxial Ag and Cu films induced by self-ion irradiation"Surf. Coat. Technol., 201 (2007) 8273.

07J099 4-13, 4-19 S

Y. Maeda, T. Jonishi, K. Narumi, Y. Ando,
K. Ueda, M. Kumano, T. Sadoh and M. Miyao,
"Axial orientation of molecular-beam-epitaxy grown Fe₃Si/Ge hybrid structures and its degradation"
Appl. Phys. Lett., 91 (2007) 171910.

07J100 4-13, 4-19 S

Y. Ando, K. Ueda, M. Kumano, T. Sadoh,
K. Narumi, Y. Maeda and M. Miyao,
"Low Temperature Hetero-Epitaxy of Ferromagnetic Silicide on Ge Substrates for Spin-Transistor Application"
電子情報通信学会 信学技報 ED2007-101, 6 月号 221-224.

07J101 4-16 S

A. Iwase, M. Fukuzumi, Y. Zushi,
N. Ishikawa and F. Ono,
"Study on irradiation-induced magnetic transition in FeRh alloy by means of K-edge XMCD spectroscopy"
Nucl. Instrum. Meth. Phys. Res., B256 (2007) 429-433.

07J102 4-18 I

三浦健太、種村豪、花泉修、山本春也、 高野勝昌、杉本雅樹、吉川正人、 "シリコンイオンを注入した石英板からの 青色発光帯の観測" 電子情報通信学会論文誌 C J90-C (2007) 618.

07J103 4-18 I
K. Miura, T. Tanemura, O. Hanaizumi,
S. Yamamoto, K. Takano,
M. Sugimoto and M. Yoshikawa,
"Fabrication of blue-light emission fused-silica substrates by using Si ion implantation and high-temperature annealing"
Nucl. Instrum. Meth. Phys. Res., B 263 (2007) 532.

07J104 4-19 S T. Jonishi, Y. Ando, A. Imai and Y. Maeda, "Carrier control of β-FeSi₂ by 1.2MeV-Au++ ion irradiation" Thin Solid Films, 515 (2007) 8175-8178.

07J105 4-19 S

K. Ueda, R. Kizuka, H. Takeuchi, A. Kenjo,
T. Sadoh and M. Miyao,
"Influence of Substrate Orientation on Low-Temperature Epitaxial Growth of Ferromagnetic Silicide Fe₃Si on Si"
Thin Solid Films, 515 (2007) 8250-8253.

07J106 4-19 S

T. Sadoh, K. Ueda, Y. Ando, M. Kumano,
K. Narumi, Y. Maeda and M. Miyao,
"Low-Temperature Epitaxial Growth of [Fe3Si/SiGe]n (n=1-2) Multi-Layered Structures for Spintronics Application"
ECS Trans., 11 (2007) 473-480.

07J107 4-19 S

M. Kumano, Y. Ando, K. Ueda, T. Sadoh,
K. Narumi, Y. Maeda and M. Miyao,
"Effect of Fe/Si Ratio on Epitaxial Growth of Fe₃Si on Ge Substrate"
ECS Trans., 11 (2007) 481-486.

07J108 4-19 S

K. Ueda, Y. Ando, M. Kumano, T. Sadoh,
Y. Maeda and M.Miyao,
"Formation of Fe₃Si/Ge/Fe₃Si Multi-Layer by Double Heteroepitaxy on High Quality Fe₃Si/Ge Substrate for Spintronics Application"
ECS Trans., 11 (2007) 487-492.

07J109 4-20 C

T. Okumura, H. Nishido, S. Toyoda, T. Kaneko,

S. Kosugi, Y. Sawada and K. Komuro,

"Evaluation of Radiation-damaged Halo in Quartz by Cathodoluminescence as a geochronological tool"

Quaternary Geochronol., 2008 (in press).

07J110 4-21 S, I

K. Takahiro, K. Kawatsura, S. Nagata,
B. Tsuchiya, S. Yamamoto and H. Naramoto,
"Improvement in Crystal Quality of Epitaxial Ag and Cu Films Induced by Sel-ion Irradiation"
Surf. Coat. Technol., 201 (2007) 8273-8277.

07J111 4-21 O

Pavel Avramov, H. Naramoto, S. Sakai,
K. Narumi, Vasily Lavrentiev and Y. Maeda,
"Quantum Chemical Study of Atomic Structure Evolution of Co_x/C₆₀ (x= 2.8) Composites"
J. Phys. Chem. A111 (2007) 2299-2306

07J112 4-21 O

S. Sakai, H. Naramoto, Pavel V. Avramov,
T. Yaita, Vasily Lavrentiev, K. Narumi,
Y. Baba and Y. Maeda,
"Comparative study of structures and electrical properties in cobalt–fullerene mixtures by systematic change of cobalt content"

Thin Solid Films, 515 (2007) 7758–7764.

07J113 4-21 O

X. D. Zhu, K. Narumi and H. Naramoto,"Buckling Instability in Amorphous Carbon Films"J. Phys., Condens. Matter, 19 (2007)

236227-236232.

07J114 4-21 O

Z. Q. Chen, S. J. Wang, M. Maekawa,

A. Kawasuso, H. Naramoto, X.

L. Yuan and T. Sekiguchi,

"Thermal Evolution of Defects in As-grown and Electron-irradiated ZnO Studied by Positron Annihilation"

Phys. Rev., B75 (2007) 245206.1-245206.9.

07J115 4-21 O

S. Sakai, I. Sugai, S. Mitani, K. Takanashi,
Y. Matsumoto, H. Naramoto, Pavel Avramov,
S. Okayasu and Y. Maeda,
"Giant Tunel Magnetoresistance in Co-deposited Fullerene-Cobalt Films in the Low Bias-voltage Regime"
Appl. Phys. Lett., 91 (2007) 242104.1-242104.3

ippi: 1 iijo: Dott., 91 (2007) 2 i210 iii 2 i210

07J116 4-21 O

Pavel Avramov, S. Sakai, H. Naramoto,
K. Narumi, Y. Matsumoto and Y. Maeda,
"Theoretical DFT Study of Atomic Structure and Spin States of the Co_x(C₆₀)n (x=3-8, n=1,2) Complex Nanoclusters"
J. Phys. Chem., (2008) Accepted for publication

07J117 4-24 O

Y. Fukaya, M. Hashimoto,
A. Kawasuso and A. Ichimiya,
"Structural analysis of Si(111)-√21×√ 21-(Ag,Au) surface by using reflection high-energy positron diffraction"
Surf. Sci., 601 (2007) 5187.

07J118 4-24,4-25 O

M. Hashimoto, Y. Fukaya,A. Kawasuso and A. Ichimiya,

"Surface structure of In/Si(111) studied by reflection high-energy positron diffraction" Surf. Sci., 601 (2007) 5192.

07J119 4-24 O

Y. Fukaya, A. Kawasuso and A. Ichimiya,
"Reflection high-energy positron diffraction study on Si(111)-√3×√3-Ag surface"
Phys. Rev., B 75 (2007) 115424.

07J120 4-25 O

M. Hashimoto, Y. Fukaya,
A. Kawasuso and A. Ichimiya,
"Quasi-one-dimensional In atomic chains on Si(111) at low temperature studied by reflection high-energy positron diffraction and scanning tunneling microscopy"
Appl. Surf. Sci. (2008) in press

07J121 4-25 O

M. Hashimoto, Y. Fukaya,
A. Kawasuso and A. Ichimiya,
"Phase transition of In/Si(111) surface studied by reflection high-energy positron diffraction"
e-J. Surf. Sci. Nanotechnol. (2008) in press

07J122 4-26 G

S. Nakagawa,
"Reaction mechanism of hydroxymaleimide γ-irradiated in alcohol solution"
Radiat. Phys. Chem., accepted.

07J123 4-26 C

S. Nakagawa, M. Taguchi, N. Ohta and K. Hirota,"Degradation of Hydroxymaleimide in 2-Propanol by Irradiation of Energetic Heavy Ions (I)-Air-saturated system-" Radiat. Phys. Chem., accepted.

07J124 4-30 E, G

R. Yamada, R. Nagaishi,
Y. Hatano and Z. Yoshida,
"Hydrogen production in the γ-radiolysis of aqueous sulfuric acid solutions containing Al₂O₃, SiO₂, TiO₂ or ZrO₂ fine particles"
Int. J. Hydrogen Energy, 33(3) (2008) 929-936.

07J125 4-30 E, G

R. Motoki, H. Aoki, S. Uchida,
R. Nagaishi and R. Yamada,
"Development of production methods of the Sr-90/Y-90 source for hydrogen production experiments"
JAEA-Technol. 2008-014 (2008).

07J126 4-36 C

Y. Shikaze, Y. Tanimura, J. Saegusa,
M. Tsutsumi, Y. Yamaguchi and Y. Uchita,
"INVESTIGATION OF PROPERTIES OF THE TIARA NEUTRON BEAM FACILITY OF IMPORTANCE FOR CALIBRATION APPLICATIONS"
Radiat. Prot. Dosim., 126 (2007) 163-167.

07J127 4-38 G

H. Seito, T. Ichikawa,
H. Watanabe and T. Kojima,
"Application of Clear Polymethylmethacrylate Dosimeter, Radix RN-15 to Several kGy Range"
RADIOISOTOPES, 56 (2007) 777.

07J128 4-41 S

H. Shibata, Y. Kohno, K. Shibata, T. Sato, M. Oikawa, J. Haga and T. Sakai, "Nuclear Reaction Microanalysis of Boron Doped Steels"

Nucl. Instrum. Methods Phys. Res., B 260 (2007) 321-324.

07J129 4-44 T

A. Chiba, Y. Saitoh, K. Narumi,
M. Adachi and T. Kaneko,
"Average charge and its structure dependence of fragment ions under irradiation of a thin carbon foil with a 1-MeV/atom C₃⁺ cluster ion"
Phys. Rev., A 76 (2007) 063201.

07J130 4-45 O

P.V. Avramov, D.G. Fedorov, P.B. Sorokin,L. A. Chernozatonskii and M.S. Gordon,"Atomic and electronic structure of new hollow-based symmetric families of silicon nanoclusters"

J. Phys. Chem. C 111 (2007) 18824-18830.

07J131 4-45 O

P.V. Avramov, A.A. Kuzubov, A.S. Fedorov,
P.B. Sorokin, F.N. Tomilin and Y. Maeda,
"Density-functional theory study of the electronic structure of thin Si/SiO₂ quantum nanodots and nanowires"

Phys. Rev., B 75, 205427 (2007).

07J132 4-45 O

P.V. Avramov,

L.A. Chernozatonskii and P.B. Sorokin,

"Multiterminal Nanowire Junctions of Silicon: A Theoretical Prediction of Atomic Structure and Electronic Properties"

Nano. Lett., 7, 00973-3 (2007).

07J133 4-47 I

K. Hirata, Y. Saitoh, A.Chiba, M. Adachi,
K. Yamada, and K. Narumi,
"Development of secondary ion mass spectroscopy using medium energy C₆₀ ion impacts"
Nucl. Instrum. Methods Phys. Res., B 266 (2008) 2450.

07J134 4-47 T

K. Hirata, Y. Saitoh, A.Chiba, and K. Narumi,
"Organic Contaminant Detection of Silicon Wafers Using Negative Secondary Ions Induced by Cluster Ion Impacts"
Appl. Phys. Express, 1 (2008) 047002

07J135 4-48 T,I

K. Maehashi, H. Ozaki, Y. Ohno, K. Inoue,
K. Matsumoto, S. Seki, and S. Tagawa,
"Formation of Single Quantum Dot in Single-Walled Carbon Nanotube Channel Using Focused-Ion-Beam Technique"
Appl. Phys. Lett., 90 (2007) 023103-1 - 023103-3.

07J136 4-48 T

S. Seki, S. Tsukuda, S. Tagawa, M. Sugimoto,A. Watanabe and T. Miyashita,"Formation of Isolated Ultrafine Optical Nanofibers by Single Particle Nanofabrication Technique"

J. Photopolym. Sci. Technol., 20 (2007) 125-128.

07J137 4-48 T

S. Seki, S. Watanabe, M. Sugimoto,

S. Tagawa and S. Tsukuda,

"Multi-Functional Polymer Nanowires with Ultra-high Aspect Ratio Produced by Single Particle Nano-fabrication Technique" J. Photopolym. Sci. Technol., (2008) in press.

07J138 4-50 C

S. Kurashima, N. Miyawaki, S. Okumura,
M. Oikawa, K. Yoshida, T. Kamiya, M. Fukuda,
T. Satoh, T. Nara, T. Agematsu, I. Ishibori,
W. Yokota and Y. Nakamura,
"Improvement in beam quality of the JAEA AVF cyclotron for focusing heavy-ion beams with energies of hundreds of MeV"
Nucl. Instrum. Meth. Phys. Res., B 260 (2007) 65.

07J139 4-50 C

M. Oikawa, T. Satoh, T. Sakai, N. Miyawaki,
H. Kashiwagi, S. Kurashima, S. Okumura,
M. Fukuda, W. Yokota and T. Kamiya
"Focusing high-energy heavy ion microbeam system at the JAEA AVF cyclotron"
Nucl. Instrum. Meth. Phys. Res., B 260 (2007) 85.

07J140 4-51 C

Y. Yuri, N. Miyawaki, T. Kamiya, W. Yokota,K. Arakawa and M. Fukuda,

"Reply to "Comment on 'Uniformization of the transverse beam profile by means of nonlinear focusing method' "

Phys. Rev., ST Accel. Beams, 11 (2008) 039002

07J141 4-51, 5-02 C

Y. Yuri, N. Miyawaki, T. Kamiya, W. Yokota,
K. Arakawa and M. Fukuda,
"Uniformization of the transverse beam profile by means of nonlinear focusing method"
Phys. Rev., ST Accel. Beams, 10 (2007) 104001

A 1.2 Publication in Proceedings

07C001 1-01 C

H. Shindou, S. Kuboyama, N. Ikeda, Y. Satoh,
T. Tamura and T. Hirao,
"Evaluation of the Proton Induced Bulk Damage in SDRAM Utilizing 90nm Process Technology"
IEEE Nucl. Space Radiat. Eff. Conf. (NSREC 2007).

07C002 1-01 C

S. Kuboyama, C. Kamezawa, N. Ikeda, Y. Satoh,
H. Oyama and T. Hirao,
"Single-Event Burnout of Silicon Carbide Schottky
Barrier Diodes Caused by High Energy Protons"
IEEE Nucl. Space Radiat. Eff. Conf. (NSREC 2007).

07C003 1-01 C

A. Makihara, H. Asai, Y. Tsuchiya, Y. Amano,
M. Midorikawa, H. Shindou, S. Kuboyama,
Y. Nakajima, Y. Takahashi, K. Ohnishi,
S.Onoda and T. Hirao,
"Optimization for SEU/SET Immunity on 0.15 μm
Fully Depleted CMOS/SOI Digital Logic
Devices"
IEEE Nucl. Space Radiat. Eff. Conf. (NSREC 2006).

07C004 1-02, 1-03 C, T

府金賢、高橋芳浩、中嶋康人、長澤賢治、 今川良、野本敬介、大西一功、平尾敏雄、 小野田忍、大島武、 "MOSFET における重イオン照射誘起ゲート電流" 応用物理学会学術講演会 5p-ZF-13 (2007). **07C005 1-02, 1-03 C, G, T** 高橋芳浩、 "MOS デバイスのハードエラーおよびその抑制" 応用物理学会学術講演会 6p-ZE-9 (2007).

07C006 1-02, 1-03 C, T 府金賢、今川良,大澤一仁、大脇章弘、 平尾敏雄、大西一功、高橋芳浩、 "MOSFET における重イオン照射誘起ゲート電流" 日大理工学術講演会 M-25 (2007).

07C007 1-02, 1-03 C, G, T 小野田忍、府金賢、平尾敏雄、大島武、 高橋芳浩、今川良、大西一功、 "重イオンがγ線照射前後の MOS キャパシタに誘 起する過渡電流" 応用物理学関係連合講演会 28p-ZR-5 (2008).

07C008 1-02 T 平尾敏雄、高橋芳浩、府金賢、小野田忍、 大島武、 "TIBIC を用いた酸化膜中に誘起した過渡電流の 解明に関する研究" JAEA-Conf 2008-005

```
07C009 1-02 T
府金賢、高橋芳浩、平尾敏雄、小野田忍、
中嶋康人、大西一功、
"酸化膜を介したイオン照射誘起電荷の収集機構
解明に向けた研究"
JAEA-Conf 2008-005
```

07C010 1-05 I, C, T, E M. Saito, M. Imaizumi, T. Ohshima and Y. Ito, "EFFECTS OF IRRADIATION BEAM

CONDITIONS ON RADIATION

DEGRADATION OF SOLAR CELLS" Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf., December 2007, (Fukuoka), pp.963-964.

07C011 1-05 I, C, E

H. Miyamoto, S. Sato, T. Ohshima, C. Morioka,
M. Imaizumi and K. Kawano,
"EFFECTS OF IRRADIATION TEMPERATURE
ON DEGRADATION OF ELECTRICAL
CHARACTERISTICS OF InGaP SOLAR
CELLS"
Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf.,
December 2007, (Fukuoka), pp.961-962.

07C012 1-05 I, C, E

M. Imaizumi, H. Toyota, H. Ogawa and M. Tajima, "RESULTS OF CELL DURABILITY TESTS FOR JAXA'S HIHT PROJECTS, VENUS AND MERCURY EXPLORATION MISSIONS" Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf., December 2007, (Fukuoka), pp.959-960.

07C013 1-05 I, C, E

M. Yamaguchi, N.J. Ekins-Daukes, H-S. Lee,M. Imaizumi, T. Takamoto and T. Ohshima,"ANALYSIS FOR RADIATION-RESISTANCEOF INGAP SUB-CELLS"Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf.,

December 2007, (Fukuoka), pp.957-958.

07C014 1-05 E

T. Ohshima, S. Sato, H. Miyamoto, M. Imaizumi, H. Hanaya and K. Kawano, "RECONSIDERATION OF ELECTRON IRRADIATION TEST METHODS FOR THE EVALUATION OF SPACE SOLAR CELLS" Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf., December 2007, (Fukuoka), pp.955-956.

07C015 1-05 I, C, T, E

C. Morioka, M. Imaizumi, H. Sugimoto, S. Sato,
T. Ohshima and M. Tajima,
"STRUCTURAL STUDY ON (Al)InGaP
SINGLE-JUNCTION SOLAR CELL FOR
PERFORMANCE IMPROVEMENT OF
TRIPLE-JUNCTION SOLAR CELLS"
Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf.,
December 2007, (Fukuoka), pp.504-505.

07C016 1-05 I, C, T, E

S. Sato, H. Miyamoto M. Imaizumi, K. Shimazaki, C. Morioka, K. Kawano and T. Ohshima, "DEGRADATION MODELING OF InGaP/GaAs/Ge TRIPLE JUNCTION SOLAR CELLS IRRADIATED WITH VARIOUS ENERGY PROTONS" Tech. Dig. 17th Int. Photovolt. Sci. Eng. Conf., December 2007, (Fukuoka), pp.502-503.

07C017 1-07 I

A. Wakahara, T. Shimojo, H. Kawai, H. Okada,
T. Ohshima and S. Sato,
"Optical Gain of Eu³⁺ Ion Implanted AlGaN and Its Al Compositional Dependence"
7th Int. Conf. Nitride Semicond., (Sept. 16-21, 2007, Las Vegas) WP57

07C018 1-07 I

A. Wakahara,"Monolithic Optoelectronic Integrated Circuits"4th Int. Workshop Nanoscale Semicond. Devices

07C019 1-07 I

J. H. Park, Y. Furukawa,
H. Okada and A. Wakahara,
"Luminescence Properties of Tb-Implanted
Al_xGa_{1-x}N Epitaxial Layer"
8th Korea-Japan Joint Workshop Adv. Semicond.
Processes Equip., (Oct. 4-6, 2007, Busan, Korea)
Proc., pp.126-128

07C020 1-07 I

A. Wakahara, H. Okada, J. -H. Park, K. Takemoto,
T. Shimojyo and A. Yoshida,
"Development of Eu-doped AlGaN for
Optoelectronic Integrated System-Optical Gain and Loss Measurement"
8th Korea-Japan Joint Workshop Adv. Semicond.
Processes Equip., (Oct. 4-6, 2007, Busan, Korea)
Proc., pp.89-92

07C021 1-09 O

A. Miyashita, T. Ohnuma, M. Iwasawa,
H. Tsuchida and M. Yoshikawa,
"Generation of amorphous SiO₂/SiC interface structure by the first-principles molecular dynamics simulation"
Mater. Sci. Forum, 556-557, (2007) 521.

07C022 1-09 O

T. Ohnuma, A. Miyashita, M. Iwasawa,
M. Yoshikawa and H. Tsuchida,
"Dynamical simulation of SiO₂/4H-SiC(0001)
interface oxidation process: from first-principles"
Mater. Sci. Forum, 556-557 (2007) 615.

07C023 1-09 O

T. Ohnuma, A. Miyashita, M. Iwasawa,M. Yoshikawa and H. Tsuchida,

"Dynamical simulation of SiO₂/4H-SiC interface on C-face oxidation process: from first principles" Proc. Int. Conf. Silicon Carbide Related Mater. 2007 (to be published)

07C024 1-11 E

N. T. Son, I. G. Ivanov, A. Kuznetsov,
B. G. Svensson, Q. X. Zhao, M. Willander,
N. Morishita, T. Ohshima, H. Itoh, J. Isoya,
E. Janzén and R. Yakimova,
"Magnetic resonance studies of defects in electron-irradiated ZnO substrates"
Proc. 24th Int. Conf. Defects Semicond.
(Albuquerque, USA, 22-27 July 2007)
Physica B 401-407, 507-510 (2007)

07C025 1-11 T, E, G

J. Isoya, T. Umeda, N. Mizuochi, N.T. Son,
E. Janzén and T. Ohshima,
"EPR Identification of defects and impurities in SiC: To be decisive"
Proc. Int. conf. Silicon Carbide Related Mater.,
(ICSCRM2007) (Otsu, Japan, 14-19 Oct. 2007),
Mater. Sci. Forum (in press)

07C026 1-11 E

T. Umeda, N. Morishita, T. Ohshima,
H. Itoh and J. Isoya,
"Photo-EPR sudy of vacancy-type defects in irradiated n-type 4H-SiC"
Proc. Int. conf. Silicon Carbide Related Mater.,
(ICSCRM2007) (Otsu, Japan, 14-19 Oct. 2007),
Mater. Sci. Forum (in press)

07C027 2-06 O M. Takigami, H. Amada, N. Nagasawa, A. Hiroki,

N. Kasai, F. Yoshii and M. Tamada,

"Absorption of Phosphate Ion in Swine Urine Using CMC Gel" Trans. Jpn. Mater. Res. Soc., in press

07C028 2-10 E

島田明彦、箱田照幸、広田耕一、 "改正大気汚染防止法に向けた VOC 対策技術と しての電子ビーム利用" 第12回放射線プロセスシンポジウム 講演要旨・ ポスター発表要旨集 p.113

07C029 3-03 C

吉原亮平、安田愛、佐藤良平、長谷純宏、 鳴海一成、滝本晃一、 "rpsL 遺伝子を用いた植物における突然変異解 析" 変異機構研究会・第20回夏の学校 講演要旨集, 11-12 (2007)

07C030 3-03 C

吉原亮平、長谷純宏、滝本晃一、鳴海一成、 "シロイヌナズナにおけるイオンビームおよびガン マ線誘発突然変異の解析" 第49回 日本植物生理学会年会要旨集,346 (2008)

07C031 3-04 C

Y. Matuo, S.Nishijima, Y. Hase, A. Sakamoto,
I. Narumi and K. Shimizu,
"Molecular analysis of carbon ion induced mutations in the budding yeast Saccharomyces cerevisiae"
Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc.
(2007) p83.

07C032 3-04 C 松尾陽一郎、泉佳伸、西嶋茂宏、清水喜久雄、 "重粒子線照射による変異誘発と二本鎖切断修復 の効果" 日本原子力学会「2007 年秋の年会」F40 (2007)

07C033 3-06 C

M. Okamura, "Flower Breeding by Quantum Beam Technology and its Commercialization" GAMMA FIELD SYMPOSIA No.45, pp 77-89, Inst. Radiat. Breeding, NIAS (2006).

07C034 3-09 C

飯塚正英、木村康夫、長谷純宏、田中淳、 "イオンビーム照射によるオステオスペルマム新品 種の作出" 第4回イオンビーム育種研究会大会講演要旨集 (2007)

07C035 3-14 C 坂本咲子、小林伸雄、中務明、長谷純宏、 鳴海一成、 "イオンビーム照射が各種ツツジ属種子の発芽に 及ぼす影響" 園芸学会中四国支部要旨 46:39, (2007).

07C036 3-14 C 加納さやか、小林伸雄、中務明、長谷純宏、 鳴海一成、 "イオンビーム照射がアスクレピアスの生育および 開花に及ぼす影響" 園芸学会中四国支部要旨 46:40, (2007).

07C037 3-18 C 松尾洋一、長谷純宏、吉原亮平、鳴海一成、 大藪榮興、 "2007 重イオンビーム照射によるユズのトゲ消失変 異個体の誘発"

園芸学研究 第6巻 別冊1 40.

07C038 3-23 C

近藤恵美子、長谷純宏、田中淳、石坂宏、 "イオンビーム照射によるシクラメン種間雑種 (Cyclamen persicum × C. purpurascens)の突然変 異体作出" 育種学研究9(別2), 2007, 247.

07C039 3-40 C

A. Oue, N. Shimizu, A. Tanaka, T. Ohtsuki,
M. Shinagawa, T. Mori, M. N Saha, A. N. Hoque,
S. Islam, T. Nakamura, S. Wada, N. Hamada,
T. Funayama, Y. Kobayashi and H. Hoshino,
"Stress-induced premature senescence in a human glioma-derived cell line after irradiation with heavy ion beam"
Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc., Vol.

2007, p341 (2007)

07C040 3-40 C

H. Hoshino, N. Shimizu, A. Oue, A. Tanaka,M. N. Saha, M. Shinagawa, T. Ohtsuki, T. Mori,A. Hoque, S. Islam, N. Hamada, Y. Kobayashi,S. Wada, T. Funayama and I. Narumi,

"Analyses of changes of cell properties specifically induced by heavy ion irradiation using viruses as analyzing tools"

Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc., Vol 2007, p346 (2007)

07C041 3-40 C

T. Ohtsuki, A. Oue, A. Tanaka, S. Islam,N. Shimizu, S. Wada, N. Hamada, T. Funayama,Y. Kobayashi and H. Hoshino,"Analyses on mechanisms of HIV-1 infection using irradiation of heavy ion beam"

Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc., Vol 2007, p347 (2007)

07C042 3-41 C, T

M. Hino, S. Wada, Y. Tajika, Y. Morimura,
N. Hamada, T. Funayama, T. Sakashita,
T. Kakizaki, Y. Kobayashi and H. Yorifuji,
"Ultrastructural changes of isolated skeletal muscle fibers induced by heavy ion microbeam irradiation"
Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc.,
(2007)

07C043 3-42 O

Al-Jahdari Wael Salem、鈴木義行、吉田由香里、 浜田信行、岡本雅彦、野田真永、舟山知夫、 坂下哲哉、小林泰彦、中野隆史、 "RBE of carbon beams on cell neurons; Approach using growth cone collapse assay" 日本放射線腫瘍学会第 20 回学術大会 2007 年 12 月 13-15 日、福岡

07C044 3-42 O

W. Al-Jahdari, Y. Suzuki, Y. Yoshida, N. Hamada,
S. Noda, K. Shirai, et al,
"Relative Biological Effectiveness of
Carbon-beams on Cell Neurons: Approach Using
Grwth Cone Collapse Assay"
Radiol. Soc. North America (RSNA)
November25-30, 2007, Chicago

07C045 3-42 O

吉田由香里、鈴木義行、浜田信行、白井克幸、 S. Al-Jahdari Wael、舟山知夫、坂下哲哉、 小林泰彦、小澤瀞司、中野隆史、 "組織培養切片を用いた正常脳における炭素線の 生物学的効果の検討" 第 50 回日本放射線影響学会併催 COE シンポジ ウム 2007 年 11 月 16 日、千葉 【受賞:同 50 回大会優秀発表賞】

07C046 3-42 O

S. Al-Jahdari Wael、鈴木義行、吉田由香里、 浜田信行、野田真永、白井克幸、舟山知夫、 坂下哲哉、小林泰彦、中野隆史、 "Neuonal sensitivity to carbon beams; Approach using growth cone collapse assay" 第 50 回日本放射線影響学会併催 COE シンポジ ウム 2007 年 11 月 16 日、千葉

07C047 3-42 O

W.S. Al-Jahdari, Y.Suzuki, Y. Yoshida,
N. Hamada, S. Noda, K. Shirai, T. Funayama,
T. Sakashita, H. Sakurai,
Y. Kobayashi and T. Nakano,
"Growth cone collapse assay approach to examine sensitivity of cell neurons to carbon beams"
第 13 回癌治療増感研究会 2007 年 5 月 18-19 日、

前橋

07C048 3-42 O

白井克幸、鈴木義行、水井利幸、浜田信行、 野田真永、舟山知夫、吉田由香里、小林泰彦、 白尾智明、中野隆史、 "培養ラット未熟神経細胞における、アポトーシスを 指標とした炭素線の生物学的効果比に関する検 討" 第13回癌治療増感研究会2007年5月18-19日、 前橋

07C049 3-43 C

M. Suzuki, Y. Furusawa, C.T suruoka,K. Kobayashi, N. Usami, M. Maeda, T. Funayama,T. Sakashita, Y. Yokota, K. Fukamoto,

N. Hamada and Y. Kobayashi,

"Radiation-quality-dependence in bystander cellular effects on normal human cells induced by carbon-ion and X-ray microbeams" Proc. 24th Space Util. Symp., Space Util. Res., 24, (2007) 328-330.

07C050 3-48 C

K. Shirai, K. Kiguchi, T. Sakata. K. Fukamoto,
T. Sakashita, T. Funayama,
N. Hamada and Y. Kobayashi,
"Regeneration mechanism of hemopoietic organs in the silkworm, Bombyx mori, after heavy-ion irradiation: Analysis of transplantation of the irradiated organs using a transgenic silkworm strain"
Asia-Pacific Congr. Sericulture Insect Biotechnol.

2008 Nagoya, 91 (2008)

07C051 3-54 C

菊地郁、石井里美、藤巻秀、鈴井伸郎、 本多一郎、河地有木、 "ポジトロンイメージング装置(PETIS)によるナス果 実への光合成産物転流の可視化" 園芸学会平成19年度秋季大会(園芸学研究)第6 巻(2)383

07C052 3-58 C

Y. Iida, H. Hanaoka, S. Watanabe, S. N. Ishioka,
S. Watanabe, S. Matsuhashi, H. Yoshioka,
B. Paudyal, N. Oriuchi, T. Higuchi and K. Endo,
"A Direct Comparison between In Vivo Optical Imaging and PET Imaging in Mice"
Proc. AMI/SMI Joint Mol. Imaging Conf. 2007,
Providence, Rhode Island, USA (2006).

07C053 3-58 C

飯田靖彦、花岡宏史、吉岡弘樹、高柳恵美、

Paudyal Bishnuhari、Paudyal Pramila、渡辺 智、 石岡典子、渡辺茂樹、松橋信平、富永英之、 樋口徹也、織内 昇、遠藤啓吾、 "がん診断におけるPETの有用性と新しいPET用 放射性核種の利用" 群馬大学 生体調節研究所 シンポジウム 前橋 (2007).

07C054 3-63

草壁孝彦、中里享美、高田久嗣、久永悦子、 文 賢徳、中嶋克行、鈴木慶二、及川将一、 佐藤隆博、荒川和夫、長嶺竹明、 "Analysis of heavy metals in testis of cadmium-exposed rat using in air micro Particle Induced X-ray Emission(PIXE)" Proc. 50th Annu. Meet. Jpn. Radiat. Res. Soc., 21st Century COE Prog., 3rd Int. Symp. Biomed. Res. Using Accel. Technol., 21st Century COE Prog.

07C055 3-63

草壁孝彦、高田久嗣、中里享美、鈴木慶二、 中嶋克行、及川将一、佐藤隆博、神谷富祐、 長嶺竹明、 "肝線維化における微量元素の変動とメタロチオネ イン、熱ショック蛋白質の変化について" 第10回環境ホルモン学会研究発表会

07C056 3-63

草壁孝彦、高田久嗣、中里享美、文賢徳、 鈴木慶二、中嶋克行、及川将一、佐藤隆博、 荒川和夫、伊波匡彦、長嶺竹明、 "肝線維化ラットを用いたフコイダン(オキナワモズ ク)による肝線維化抑制メカニズムの解析" メタロチオネインおよびメタルバイオサイエンス研 究会 2007

07C057 3-63

草壁孝彦、高田久嗣、中里享美、佐藤隆博、 神谷富祐、伊波匡彦、長嶺竹明、 "ラット肝線維化モデルにおけるフコイダン(オキナ ワモズク)投与による肝臓組織の微量元素の変動" 第24 回 PIXE シンポジウム

07C058 3-63

草壁孝彦、中里享美、高田久嗣、文 賢徳、 中嶋克行、鈴木慶二、及川将一、佐藤隆博、 荒川和夫、長嶺竹明、 "カドミウムの曝露によるセルトリ細胞の金属元素分 布とメタロチオネイン、熱ショック蛋白質の変化に ついて" 第18回日本微量元素学会

07C059 3-66

K. Yanai, H. Dai, E. Sakurai and T. Watanabe, "The roles of histamine H(1) receptors on cognition" Inflamm. Res., 2008 Mar 14: Supple1:S01-S02.

07C060 4-13, 4-19 S, I

Y. Hiraiwa, T. Jonishi, Y. Ando, K. Narumi,
M. Kumano, K. Ueda, T. Sadoh,
M. Miyao and Y. Maeda,
"Diffusion behavior in Fe₃Si/Ge hybrid structures and related degradation of axial orientation"
Extended Abst. 5th Int. Symp. Cont. Semicond.
Interfaces, 97-98 (2007).

07C061 4-13, 4-19 S

Y. Hiraiwa, Y. Ando, M. Kumano, K. Ueda,
T. Sadoh, M. Miyao, K. Narumi and Y. Maeda,
"Axial orientation of epitaxially grown
Fe₃Si/Ge(111)"
Tech. Rep. IEICE., SDM2007-230, 35-38 (2007).

07C062 4-13 S

T. Jonishi, Y. Ando, Y. Hiraiwa, K. Narumi,
M. Kumano, K. Ueda, T. Sasoh,
M. Miyao and Y. Maeda,
"Atomic diffusion at Fe₃Si/Ge epitaxial interfaces"
JSAP Proc. 10th Summer Symp. Semicond.
Silicides Related Mater., 55-57 (2007).

07C063 4-18 I

K. Miura, T. Tanemura, O. Hanaizumi,
S. Yamamoto, K. Takano,
M. Sugimoto and M. Yoshikawa,
"Observation of blue light emission from Si ion implanted fused-silica substrates"
Conf. Dig. Eur. Conf. Lasers Electro-Optics, Int.
Quant. Electron. Conf. (CLEO/Europe-IQEC)
2007, CE6-1-WED (2007).

07C064 4-18 I

K. Miura, T. Tanemura, O. Hanaizumi,
S. Yamamoto, K. Takano,
M. Sugimoto and M. Yoshikawa,
"Blue-light emission from Si ion implanted
fused-silica substrates generated by high
temperature annealing"
Tech. Dig. 12th Optoelectron. Commun. Conf.,
16th Int. Conf. Integrated Optics, Opt. Fiber
Commun. (OECC/IOOC 2007), 754-755 (2007).

07C065 4-19 S

K. Ueda, Y. Ando, M. Kumano, T. Sadoh,
Y. Maeda and M. Miyao,
"Low temperature formation of multi-layered structures of ferromagnetic silicide Fe₃Si and Ge" Extended Abst. 5th Int. Symp. Cont. Semicond. Interfaces, 99-100 (2007).

07C066 4-19 S

M. Miyao, K. Ueda, M. Kumano, K. Narumi,
Y. Ando and Y. Maeda,
"Atomically controlled hetero-epitaxy of
Fe₃Si/SiGe for spintronics application"
Proc. 5th Int. Conf. Silicon Epitaxy Heterostruct.
(ICSI-5), 151-152 (2007).

07C067 4-19 S

T. Sadoh, K. Ueda, Y. Ando, M. Kumano,
K. Narumi, Y. Maeda and M. Miyao,
"Low-temperature epitaxial growth of [Fe₃Si/SiGe]n(n=1-2) multi-layered structures for spintronics application"
Proc. 212th Meet. Electrochem. Soc. (ECS), 1315 (2007).

07C068 4-19 S

T. Jonishi, Y. Ando, Y. Hiraiwa, M. Narumi,
M. Kumano, K. Ueda, T. Sadoh,
M. Miyao and Y. Maeda,
"Atomic diffusion at Fe₃Si/Ge epitaxial interface"
JSAP Proc.10th Summer Symp. Semicond.
Silicides Related Mater., 55-57 (2007).

07C069 4-19 S, I

Y. Ando, K. Ueda, M. Kumano, T. Sadoh,
K. Narumi, Y. Maeda and M. Miyao,
"Low Temperature Hetero-Epitaxy of Ferromagnetic Silicide on Ge Substrates for
Spin-Transistor Application"
Proc. 15th Asia-Pacific Workshop Adv. Semicond.
Devices 2007
Tech. Rep. IEICE. SDM, Vol.107, No.111,
221-224,(2007).

07C070 4-22

大原宏太、石川法人、境誠治、松本吉弘、 道上修、太田康之、 "CeO2の高エネルギーイオン照射による損傷" 日本物理学会「第63回年次大会」予稿集第2分 冊 p.157(2008)

07C071 4-26 C

S. Nakagawa, N. Ohta. M. Taguchi and K. Hirota, "LET effect on radiation of hydroxymaleimide in 2-propanol (2)"

Proc. 50th Annu. Meet. Jpn. Soc. Radiat. Chem., 119-120 (2007).

07C072 4-30 E, G

R. Nagaishi,

"Solution chemical approach to radiation-induced reactions in aqueous solutions containing oxide fine particles"

Conf. Abs. Book ASR2007 Symp., Int. Symp. "Charged Particle and Photon Interactions with Matter", 12 (2007).

07C073 4-31 G

Y. Sugo, Y. Sasaki, T. Kimura and T. Sekine, "Attempts to Improve Radiolytic Stability of Amidic Extractants"

Proc. Int. Conf. Adv. Nucl. Fuel Cycles Syst. (Global 2007) in CD-ROM, Idaho, USA, pp. 1870-73 (2007).

07C074 4-31 G

S. Suzuki, T. Yaita, Y. Sugo and T. Kimura,
"Study on Selective Separation of Uranium(VI) by New N,N-Dialkyl Carboxyamides"
Proc. Int. Conf. Adv. Nucl. Fuel Cycles Syst.
(Global 2007) in CD-ROM, Idaho, USA, pp. 1137-1141 (2007).

07C075 4-51 C

Y. Yuri, S. Okumura, T. Yuyama, T. Agematsu,
N. Miyawaki, I. Ishibori and T. Ishizaka,
"Study of Beam Profile Modification for Beam Uniformization Using Multipole Magnets"
Proc. 4th Annu. Meet. Jpn. Part. Accel. Soc., 32nd Linear Accel. Meet. (August 1-3, 2007, Wako Japan) pp. 607-609.

07C076 4-52 C

N. Miyawaki, S. Okumura, S. Kurashima,
H. Kashiwagi, K. Yoshida, Y. Yuri, T. Yuyama,
I. Ishibori, T. Agematsu, T. Ishizaka, T. Nara,
M. Fukuda, T. Ueda and T. Tachikawa,
"Development of the main probe for measuring a beam phase width in the JAEA AVF cyclotron"
Proc. 4th Annu. Meet. Jpn. Part. Accel. Soc., 32nd
Linear Accel. Meet. (August 1-3, 2007, Wako
Japan) pp.604-606

07C077 4-53 C

S. Kurashima, S. Okumura, N. Miyawaki,
I. Ishibori, K. YOoshida, T. Satoh, M. Oikawa,
H. Kashiwagi, Y. Yuri, T. Agematsu, T. Nara,
T. Kamiya, M. Fukuda and W. Yokota,
"Developments at JAEA AVF Cyclotron Facility for Heavy-ion Microbeam"
Proc. 18th Int. Conf. Cyclo. Their Appl., Giardini Naxos, Italy (2007).

Appendix 2

List of Related Patents

07PAT001 1-20

根本 義之(原子力機構・原基工)他、 住友金属テクノロジー、東北大との共同出願 「照射誘起応力腐食割れの予兆診断システム」 出願手続き中

07PAT002 1-21

井岡、木内、加藤、丸山、 神戸製鋼所、コベルコ科研との共同出願 「耐粒界腐食性および耐応力腐食割れに優れた オーステナイト系ステンレス鋼およびその製造方 法」 特願 2007-117981 号

07PAT003 1-31

吉田 勝、八巻 徹也、浅野 雅春(原子力機構・量 子ビーム)他トヨタ自動車㈱との共同出願 「機能性膜及びその製造方法、並びに燃料電池 用電解質膜及びその製造方法」 特願 2007-504840 号

07PAT004 1-31

八巻 徹也、浅野 雅春、吉田 勝、前川 康成(原 子力機構・量子ビーム)他トヨタ自動車(株)との共 同出願 「燃料電池用電解質膜及びその製造方法」 特願 2007-323426 号

07PAT005 1-31

八巻 徹也、浅野 雅春、吉田 勝、前川 康成(原 子力機構・量子ビーム)他トヨタ自動車(株)との共 同出願 「機能性膜及びその製造方法、及び燃料電池用 電解質膜の製造方法」 特願 2007-323242 号

07PAT006 2-03 麦林 裕弘、内田 敏仁(栗田工業㈱)、鈴木 義男 (オリエンタル技研工業㈱)、玉田 正男、 瀬古 典明(原子力機構・量子ビーム)の共同出願 「有害物質捕集材、その製造方法及び有害物質 処理方法」 特願 2007-254960 号

07PAT007 2-05

植木 悠二、玉田 正男(原子力機構・量子ビー ム)、角田 欣一、堀田 弘樹、佐伯 俊彦(群馬 大・院工)、小竹 玉緒(東京大・院工) 「生体高分子の高速分離を可能とする電気泳動用 低濃度ゲル及びその製造方法」 (出願中)

07PAT008 2-06

長澤 尚胤、八木 敏明(原子力機構・量子ビーム)他群馬県産業支援機構、前原康延との共同出 願 「磁気共鳴診断用組成物」 特願 2007-177746 号

07PAT009 2-06

長澤 尚胤、廣木 章博、玉田 正男(原子力機構・ 量子ビーム)他群馬県産業支援機構との共同出 願

「カルボキシメチルセルロースアルカリ塩のゲルの 製造方法及びそれによって得られた カルボキシ メチルセルロースアルカリ塩のゲル」 特願 2007-188845 号

07PAT010 2-06

長澤 尚胤、廣木 章博、笠井 昇、吉井 文男、 玉田 正男(原子力機構・量子ビーム)他群馬県産 業支援機構との共同出願 「リン酸イオン吸着用組成物とその製造方法ならび にリン酸イオン除去方法」 特願 2007-314615 号

07PAT011 2-08

廣木 章博、長澤 尚胤、玉田 正男(原子力機構・ 量子ビーム) 「高吸水性ゲルの生分解性制御方法及び その生分解性が制御された生分解性ゲル」 特願 2007-293982 号

07PAT012 3-28

豊島 快幸、赤川 巧(ヤマサ醤油㈱)、 鳴海 一成、佐藤 勝也、長谷 純宏(原子力機構・ 量子ビーム)

(日本原子力機構とヤマサ醤油㈱の共同出願) 「イオンビーム照射を用いた糸状菌の変異体作出 方法」 特願 2008-042989 号

07PAT013 3-30

半谷 吉識、小山 泰二、伊藤 考太郎、 武市 順也、大塚 照雄(キッコーマン(株))、 鳴海 一成、佐藤 勝也、小林 泰彦、坂下 哲哉、 長谷 純宏(原子力機構・量子ビーム) (日本原子力機構とキッコーマンの共同出願) 「黄麹菌変異株の育種方法」 特願 2007-269956 号

07PAT014 3-30

半谷 吉識、小山 泰二、伊藤 考太郎、 武市 順也、大塚 照雄(キッコーマン(株))、 鳴海 一成、佐藤 勝也、長谷 純宏、小林 泰彦、 坂下 哲哉(原子力機構・量子ビーム) (日本原子力機構とキッコーマンの共同出願) 「麹菌、その育種法及び醤油の製造法」 特願 2007-269954 号

07PAT015 3-51

河地 有木、藤巻 秀、松橋 信平(原子力機構・量 子ビーム) 「光合成機能イメージのための新規方法」 特願 2007-154225 号(出願中)

07PAT016 4-30

山田 禮司、永石 隆二(原子力機構・原基工) 「石英、アルミナなどの耐強酸性の酸化物を触媒と するアルコール添加硫酸水溶液を使用する放射 線誘起水素製造法」 特開 2007-326755 号

07PAT017 4-56

島田、及川、加藤、佐藤、堀内、遊佐、岸、中野 (群馬大・院医)、佐藤、酒井、上松、福田、荒川 (原子力機構・高崎研) 「荷電粒子線の照準位置決定装置,その使用方 法,及び照準位置決定装置を用いた治療装置」 PCT/JP2007/050347

Appendix 3

List of Related Press-Release and TV Programs

07NP001 3-09

平成19年5月15日、上毛新聞 他7紙に 掲載、 「花 イオンで新色に 新技術で品種改良」

07TV001 3-09

平成20年4月15日11時、NHK総合、 「こんにちはいっと6けん」 「オステオスペルマム、ヴィエントフラミンゴにつ いて」

07NP002 3-23

平成20年3月7日、日本農業新聞、 「シクラメン鮮やか、イオンビームで突然変異」

07NP003 4-50

平成19年9月5日、読売、日刊工業、上毛、電 気、化学工業、科学新聞、日経産業、に掲載、 「世界で初めて数百 MeV 級重イオンで1ミクロ ン以下のビーム形成に成功 ー耐放射線性半導体、バイオなどの最先端研 究の促進に期待-」

		I ype	of Re	search		Ir	radio	tion	Fac	ilita	*2			Type	of Re	search		Īr	radia	tion	Fac	ilita	*2
Paper		Col	labora	tion ^{*1}		- 11.	auto	uion	Fac	me	`	Paper		Col	labora	tion ^{*1}			aula		Fac	mes	3
Number	Joint	Entr.	Coop.	JAEA	Com.	С	т	s	I	F	G	Number	Joint	Entr.	Coop.	JAEA	Com.	С	т	s	Т	F	G
	Res.	Res.	Res.	Inter.	Use	C	1	5	1	Ľ	U		Res.	Res.	Res.	Inter.	Use	C	1	5		Г	0
1-01						0	0					3-05						0					
1-02						0	0					3-06						0					
1-03						0	0		0			3-07	•					0					0
1-04						0	0					3-08						0					
1-05						0	0		0	0		3-09						0					
1-06								0	0			3-10	•					0					
1-07									0	0		3-11						0					
1-08	-						0	6	0	0	0	3-12	•					0					
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1-10										6	0	3-14						6				<u> </u>	
1_10							6			6	6	3-15						6					
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1-14				•							0	3-18						0			<u> </u>		<u> </u>
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1-18				•							0	3-22	•					0			<u> </u>	\vdash	
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1-21							0	0	0			3-25						0					
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1-26							0					3-30						0					
1-27							0	0	0			3-31											0
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1-30	•									-	0	3-34				•		0					0
1-31						0					-	3-35				•		0					<u> </u>
						<u> </u>						3-36	•					0				-	
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2-02										6	-	3-38						6				<u> </u>	
2-03										•	6	3-39						6					
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3-03						0						3-52						0					
3-04						0						3-53						0				1	

Appendix 4 Type of Research Collaboration

JAEA-Review 2008-055

Type of Research				*2	Type of Research							*2											
Paper		Col	labora	tion ^{*1}	-	lr	radia	ation	Fac	ilites	5	Paper		Col	labora	tion ^{*1}		lr	radia	ation	Fac	ilites	5
Number	Joint Res.	Entr. Res.	Coop. Res.	JAEA Inter.	Com. Use	С	Т	S	Ι	Е	G	Number	Joint Res.	Entr. Res.	Coop. Res.	JAEA Inter.	Com. Use	С	Т	s	Ι	Е	G
3-54						0						4-26						0					
3-55	●					0						4-27						0					
3-56	•					0						4-28						0					
3-57						0						4-29						0	0				
3-58	•					0						4-30				•						0	0
3-59				●OL								4-31				•		0	0				
3-60			•					0				4-32						0				0	
3-61			•					0				4-33				•		0					
3-62			•					0				4-34				•		0					
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3-65			•					0				4-37	•					-				0	
3-66			•					0				4-38	•									Ô	0
3-67			•					0				4-39	•						0	0		-	_
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4-01							0	0	0			4-41								0	-		
4-02				•			0	0	0			4-42	<u> </u>		•					0			
4-03							<u> </u>		\vdash	0	0	4-43								0			
4-04										0	0	4-44							6		0		
4-05						6	0		6		•	4-45							6		0		
4-06						6	-					4-46							6		6		
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4-09				-					6			4-49							6		© @		
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4-12	-							6				4-52 A-53						6					
4-13						6						4-54						6					
4-14					-	F			6	6		A-55						•	6	6	6		
4-15	•							6				4-55				-		6		6	•		
4-10											6							•					
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4-20					-		6	6	6	6	0	5-04									•	0	6
4-22							•	6			•	5-05										0	6
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4-24				OL.								5-07						0	6	6	6	•	-
4-25				OL.								5-08						0	6	0	0		
4-25				●OL								5-08 *2						Ø	0	Ø	0		
Туре	e of R	esear	ch Co	ollabo	ration	1						² Util	izatio	n of I	rradia	tion F	acilit	ies					
Joint R	les. : J	oint r	esearc	ch with	n exte	rnal	use	rs				C : AV	/F Cy	clotro	n Syst	em							
Entr. R	Res. : 1	Resea	rch en	truste	d to JA	AEA						T : 3 N	ΛV Τε	indem	Elec	trostat	ic Ac	cele	rato	r			
Coop.	Res. :	Coop	erativ	ve rese	arch v	vith	pluı	al u	nive	ersiti	es	S:3 N	AV Si	ngle-e	nded	Electro	ostatic	c Ac	cele	rato	r		
through The University of Tokyo								I:400	kV Io	on Imp	plante	r											
JAEA	Inter.	: JAE	A inte	ernal u	ise							E : 2 N	AV El	ectror	n Acce	elerato	r						
Com. U	Use : (Comn	non us	se base	d on '	"JAI	E A- 1	facil	itv-1	use"		G : Co	-60 G	amma	a-Rav	Irradia	ation I	Faci	litie	s			
OL · C)ff lind	e (rese	earch	withou	it the	use	of i	rrad	iatic	m					. ,								
	OL : OII line (research without the use of irradiation facilities)																						

Symbol used in the Appendix

An example of symbol expression is written as following.

- ① Number of last two orders in fiscal year
- ② Kind of publication
 - **J** : Publication in Journal
 - \mathbf{C} : Publication as Proceedings
 - **NP**: Press-Release (Newspaper)
 - TV: TV Programs
 - **PA**: Patent
- \bigcirc Consecutive numbers for the kind of publication
- (4-5) Paper number
- ④ Classification number of research field
 - 1: Space, nuclear and energy engineering
 - 2: Environment conservation and resource security
 - **3**: Biotechnology and medical application
 - 4: Advanced materials, analysis and novel technology
- (5) Consecutive number every research field
- (6) Irradiation facilities utilized for the research
 - \mathbf{C} : AVF Cyclotron
 - \mathbf{T} : 3 MV Tandem Electrostatic Accelerator
 - \mathbf{S} : 3 MV Single-ended Electrostatic Accelerator
 - \mathbf{I} : 400 kV Ion Implanter
 - \mathbf{E} : 2 MV Electron Accelerator
 - G: Co-60 Gamma-ray Irradiation Facilities
 - **O**: Off-line (Research without the utilization of irradiation facilities)

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表1. SI 基本単位								
甘木昌	SI 基本ì	単位						
本平里	名称	記号						
長 さ	メートル	m						
質 量	キログラム	kg						
時 間] 秒	S						
電況	モアンペア	А						
熱力学温度	モケルビン	K						
物質量	もモ ル	mol						
光 度	モカンデラ	cd						

A2. 本平平位を用いてAC4しQ31組立平位のP	表2.	基本単位を用い	て表されるSI組立単位の例	
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선수료	SI 基本単位							
和立里	名称	記号						
面積	平方メートル	m^2						
体積	立法メートル	m ³						
速さ、速度	メートル毎秒	m/s						
加 速 度	メートル毎秒毎秒	m/s^2						
波 数	毎メートル	m-1						
密度(質量密度)	キログラム毎立法メートル	kg/m^3						
質量体積(比体積)	立法メートル毎キログラム	m ³ /kg						
電流密度	アンペア毎平方メートル	A/m^2						
磁界の強さ	アンペア毎メートル	A/m						
(物質量の)濃度	モル毎立方メートル	$mo1/m^3$						
輝 度	カンデラ毎平方メートル	cd/m^2						
屈 折 率	(数 の) 1	1						

	表 5. SI 接頭語									
乗数	接頭語	記号	乗数	接頭語	記号					
10^{24}	э 9	Y	10^{-1}	デシ	d					
10^{21}	ゼタ	Z	10^{-2}	センチ	с					
10^{18}	エクサ	E	10^{-3}	ミリ	m					
10^{15}	ペタ	Р	10^{-6}	マイクロ	μ					
10^{12}	テラ	Т	10^{-9}	ナノ	n					
10^{9}	ギガ	G	10^{-12}	ピコ	р					
10^{6}	メガ	Μ	10^{-15}	フェムト	f					
10^{3}	キロ	k	10^{-18}	アト	а					
10^{2}	ヘクト	h	10^{-21}	ゼプト	Z					
10^{1}	デカ	da	10^{-24}	ヨクト	v					

乗

 10^{1}

da

表3. 固有の名称とその独自の記号で表されるSI組立単位

			31 旭立中位	
組立量	名称	記号	他のSI単位による	SI基本単位による
			表し方	表し方
平 面 角	ラジアン ^(a)	rad		$m \cdot m^{-1} = 1^{(b)}$
立 体 角	ステラジアン ^(a)	$\mathrm{sr}^{(\mathrm{c})}$		$m^2 \cdot m^{-2} = 1^{(b)}$
周 波 数	ヘルツ	Hz		s ⁻¹
力	ニュートン	Ν		m•kg•s ⁻²
圧力,応力	パスカル	Pa	N/m^2	$m^{-1} \cdot kg \cdot s^{-2}$
エネルギー,仕事,熱量	ジュール	J	N•m	m ² • kg • s ⁻²
工率,放射束	ワット	W	J/s	m ² • kg • s ⁻³
電荷, 電気量	クーロン	С		s•A
電位差(電圧),起電力	ボルト	V	W/A	$m^2 \cdot kg \cdot s^{-3} \cdot A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2} \cdot kg^{-1} \cdot s^4 \cdot A^2$
電気抵抗	オーム	Ω	V/A	$m^2 \cdot kg \cdot s^{-3} \cdot A^{-2}$
コンダクタンス	ジーメンス	S	A/V	$m^{-2} \cdot kg^{-1} \cdot s^3 \cdot A^2$
磁東	ウエーバ	Wb	V•s	$m^2 \cdot kg \cdot s^{-2} \cdot A^{-1}$
磁束密度	テスラ	Т	Wb/m^2	$kg \cdot s^{-2} \cdot A^{-1}$
インダクタンス	ヘンリー	Н	Wb/A	$m^2 \cdot kg \cdot s^{-2} \cdot A^{-2}$
セルシウス温度	セルシウス度 ^(d)	°C		K
光束	ルーメン	1m	$cd \cdot sr^{(c)}$	$m^2 \cdot m^{-2} \cdot cd=cd$
照度	ルクス	1 x	1m/m^2	$m^2 \cdot m^{-4} \cdot cd = m^{-2} \cdot cd$
(放射性核種の)放射能	ベクレル	Bq		s ⁻¹
吸収線量, 質量エネル	ガレイ	Gw	T/ka	m ² • a ⁻²
ギー分与, カーマ		0 y	J/ ng	ш - 5
線量当量,周辺線量当				0 0
量,方向性線量当量,個	シーベルト	Sv	J/kg	$m^2 \cdot s^{-2}$
人線重当重,組織線重当				

(a) ラジアン及びステラジアンの使用は、同じ次元であっても異なった性質をもった量を区別するときの組立単位の表し方として利点がある。組立単位を形作るときのいくつかの

用例は表4に示されている。 (b)実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位として の記号"1"は明示されない。 (c)測光学では、ステラジアンの名称と記号srを単位の表し方の中にそのまま維持している。 (d)この単位は、例としてミリセルシウス度m℃のようにSI接頭語を伴って用いても良い。

表4. 単位の中に固有の名称とその独自の記号を含むSI組立単位の例

和平量	SI 組立単位							
和立里	名称	記号	SI 基本単位による表し方					
粘度	パスカル秒	Pa•s	$m^{-1} \cdot kg \cdot s^{-1}$					
力のモーメント	ニュートンメートル	N • m	$m^2 \cdot kg \cdot s^{-2}$					
表 面 張 力	ニュートン毎メートル	N/m	kg • s ⁻²					
角 速 度	ラジアン毎秒	rad/s	$m \cdot m^{-1} \cdot s^{-1} = s^{-1}$					
角 加 速 度	ラジアン毎平方秒	rad/s ²	$m \cdot m^{-1} \cdot s^{-2} = s^{-2}$					
熱流密度,放射照度	ワット毎平方メートル	W/m^2	kg • s ⁻³					
熱容量、エントロピー	ジュール毎ケルビン	J/K	$m^2 \cdot kg \cdot s^{-2} \cdot K^{-1}$					
質量熱容量(比熱容量), 質量エントロピー	ジュール毎キログラム 毎ケルビン	J/(kg \cdot K)	$m^2 \cdot s^{-2} \cdot K^{-1}$					
質量エネルギー (比エネルギー)	ジュール毎キログラム	J/kg	$m^2 \cdot s^{-2} \cdot K^{-1}$					
熱 伝 導 率	ワット毎メートル毎ケ ルビン	W/(m \cdot K)	$\mathbf{m} \cdot \mathbf{kg} \cdot \mathbf{s}^{-3} \cdot \mathbf{K}^{-1}$					
体積エネルギー	ジュール毎立方メート ル	J/m^3	$m^{-1} \cdot kg \cdot s^{-2}$					
電界の強さ	ボルト毎メートル	V/m	$\mathbf{m} \cdot \mathbf{kg} \cdot \mathbf{s}^{-3} \cdot \mathbf{A}^{-1}$					
体 積 電 荷	クーロン毎立方メート ル	C/m^3	$m^{-3} \cdot s \cdot A$					
電 気 変 位	クーロン毎平方メート ル	C/m^2	$m^{-2} \cdot s \cdot A$					
誘 電 率	ファラド毎メートル	F/m	$m^{-3} \cdot kg^{-1} \cdot s^4 \cdot A^2$					
透 磁 率	ヘンリー毎メートル	H/m	$\mathbf{m} \cdot \mathbf{kg} \cdot \mathbf{s}^{-2} \cdot \mathbf{A}^{-2}$					
モルエネルギー	ジュール毎モル	J/mo1	$m^2 \cdot kg \cdot s^{-2} \cdot mol^{-1}$					
モルエントロピー,	ジュール毎モル毎ケル	T/(1 V)	2211					
モル熱容量	ビン	J/ (mol • K)	m • kg • s • • K • • mol					
照射線量(X線及びγ線)	クーロン毎キログラム	C/kg	kg ⁻¹ • s • A					
吸収線量率	グレイ毎秒	Gy/s	$m^{2} \cdot s^{-3}$					
放 射 強 度	ワット毎ステラジアン	W/sr	$m^4 \cdot m^{-2} \cdot kg \cdot s^{-3} = m^2 \cdot kg \cdot s^{-3}$					
放 射 輝 度	ワット毎平方メートル	$W/(m^2 \cdot sr)$	$m^2 \cdot m^{-2} \cdot kg \cdot s^{-3} = kg \cdot s^{-3}$					

表6. 国際単位系と併用されるが国際単位系に属さない単位

<u>10⁻²⁴</u>

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86400 s
度	0	$1^{\circ} = (\pi / 180)$ rad
分	,	1' = $(1/60)^{\circ}$ = $(\pi/10800)$ rad
秒	"	1" = $(1/60)$ ' = $(\pi/648000)$ rad
リットル	1, L	$11=1 \text{ dm}^3 = 10^{-3} \text{m}^3$
トン	t	1t=10 ³ kg
ネーパ	Np	1Np=1
ベル	В	1B=(1/2)ln10(Np)

表7. 国際単位糸と併用されこれに属さない単位で SI単位で表される数値が実験的に得られるもの									
名称	記号	SI 単位であらわされる数値							
電子ボルト	eV	1eV=1.60217733(49)×10 ⁻¹⁹ J							
統一原子質量単位	u	1u=1.6605402(10)×10 ⁻²⁷ kg							
天 文 単 位	ua	1ua=1.49597870691(30)×10 ¹¹ m							

表8. 国際単位系に属さないが国際単位系と

伊用されるその他の単位							
名称	記号	SI 単位であらわされる数値					
海 里		1 海里=1852m					
ノット		1 ノット=1 海里毎時=(1852/3600)m/s					
アール	а	$1 \text{ a=} 1 \text{ dam}^2 = 10^2 \text{m}^2$					
ヘクタール	ha	$1 \text{ ha}=1 \text{ hm}^2=10^4 \text{m}^2$					
バール	bar	1 bar=0.1MPa=100kPa=1000hPa=10 ⁵ Pa					
オングストローム	Å	1 Å=0. 1nm=10 ⁻¹⁰ m					
バーン	b	$1 \text{ b}=100 \text{ fm}^2=10^{-28} \text{m}^2$					

表9. 固有の名称を含むCGS組立単位

A.J.	旧口	12日日日1000地工工工工	
名称		記号	SI 単位であらわされる数値
エル	グ	erg	1 erg=10 ⁻⁷ J
ダイ	ン	dyn	1 dyn=10 ⁻⁵ N
ポーア	ズ	Р	1 P=1 dyn•s/cm ² =0.1Pa•s
ストーク	ス	St	1 St $=1 \text{ cm}^2/\text{s}=10^{-4} \text{m}^2/\text{s}$
ガウ	ス	G	1 G ≙10 ⁻⁴ T
エルステッ	・ド	0e	1 Oe ≙(1000/4π)A/m
マクスウェ	ル	Mx	1 Mx ≙10 ⁻⁸ Wb
スチル	ブ	sb	$1 \text{ sb } = 1 \text{ cd/cm}^2 = 10^4 \text{ cd/m}^2$
朩	ŀ	ph	1 ph=10 ⁴ 1x
ガ	ル	Gal	$1 \text{ Gal} = 1 \text{ cm/s}^2 = 10^{-2} \text{m/s}^2$

表10. 国際単位に属さないその他の単位の例				
名称			記号	SI 単位であらわされる数値
キュ	IJ	ĺ	Ci	1 Ci=3.7×10 ¹⁰ Bq
レン	トゲ	\sim	R	$1 R = 2.58 \times 10^{-4} C/kg$
ラ		ド	rad	1 rad=1cGy=10 ⁻² Gy
ν		A	rem	1 rem=1 cSv=10 ⁻² Sv
X 線	単	位		1X unit=1.002×10 ⁻⁴ nm
ガンマ			γ	$1 \gamma = 1 nT = 10^{-9}T$
ジャン	レスキ	1	Jy	$1 \text{ Jy}=10^{-26} \text{W} \cdot \text{m}^{-2} \cdot \text{Hz}^{-1}$
フェ	ル	111		$1 \text{ fermi=1 fm} = 10^{-15} \text{m}$
メートル系カラット				1 metric carat = 200 mg = 2×10^{-4} kg
ŀ		ル	Torr	1 Torr = (101 325/760) Pa
標準	大 気	圧	atm	1 atm = 101 325 Pa
力 口	IJ	ļ	cal	
3 <i>1</i>	17	~	11	$1 - 1 - 1 - 10^{-6}$ m