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# A study of defects in electron- and ion-irradiated ZrCuAl bulk glassy alloy using positron annihilation techniques

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**Abstract.** Free volume changes in  $Zr_{50}Cu_{40}Al_{10}$  bulk glassy alloys irradiated by 200 and 2.5 MeV Xe ions, 180 keV He ions, and 2 MeV electrons were investigated at room temperature using positron annihilation lifetime and Doppler broadening techniques. In addition, a slow positron beam was used to probe the change in free volume in the 180 keV He ion-irradiated sample. X-ray diffraction revealed that no crystallization took place in any of the irradiated samples. The Doppler broadening spectra from the annihilated gamma rays remained essentially constant in all ion-irradiation cases; however, an extremely minor change of positron mean lifetime was detected in each case. For electron- and He ion-irradiated samples the positron lifetime increased, and the opposite was seen in heavy-ion irradiated samples. The Doppler broadening S parameter increased with He-ion radiation dose, and the depth profile correlated well to the damage profile.

## 1. Introduction

Bulk glassy alloys show promise for various applications because they possess superior mechanical properties such as hardness, strength, corrosion resistance and micro-formability [1-3]. We propose that the properties of bulk glassy alloys can be further improved via irradiation treatments. To date, thin-film metallic glasses treated with electron- and ion radiation have been extensively studied [4-8]. In these thin films, both electron [4] and ion irradiation [7-8] was found to crystallize the metallic glass. However, it has become apparent that the local structure and properties in thin films are different to bulk glassy alloys, and furthermore, it is known that glassy alloys contain internal stresses. We suspect that the larger sample size has a direct influence on the effects of radiation on bulk glassy alloys, and have previously reported on the effects of electron-and swift heavy-ion radiation on the

free volume and mechanical properties of ZrCuAl bulk glassy alloys [9]. In the present work we investigate the influence of radiation species on the radiation effects and free volume changes during low-energy He-ion radiation of ZrCuAl bulk glasses using positron annihilation techniques.

## 2. Experimental procedure

Zr<sub>50</sub>Cu<sub>40</sub>Al<sub>10</sub> bulk glassy alloy samples were fabricated by the tilt casting method in an arc furnace and cut into 0.6-mm-thick disks of 8 mm in diameter following the procedure given in [9]. The samples were irradiated at room temperature with 200 and 2.5 MeV Xe ions, 2 MeV electrons, or 180 keV He ions using a tandem-type accelerators at the Japanese Atomic Energy Research Agency (JAEA) Takasaki, the Wakasa-Wan Energy Research Center (WERC), and JAEA Tokai Japan, respectively. The total doses of radiation used are listed in Table 1. The irradiation fluence of He ions used was  $1.0 \times 10^{15}$ ,  $5.0 \times 10^{15}$  and  $1.0 \times 10^{16}$  ions/cm<sup>2</sup>. Positron annihilation lifetime measurement was performed for the electron-irradiated and 200 MeV Xe ion-irradiated samples using a classical sandwich configuration (CSC) of the positron source between two identical samples. Furthermore, coincidence Doppler broadening (CDB) measurements were performed for electron, He and Xe ion-irradiated samples using the CSC method. Positron lifetime and Doppler broadening measurements for the 180 keV He and the 2.5 MeV Xe ion-irradiated samples were carried out in vacuum using slow positron beam apparatus with an energy range of 1 to 30 keV, which corresponds to mean implantation depth of incident positrons from 0.05 to 1.3 mm in this ZrCuAl alloy. The statistical error of the fits was less than 1 ps, however, the overall accuracy was 2 ps due to the reproducibility of the experiments. The region of  $|p_L| < 3.6 \times 10^{-3}$  mc in the Doppler broadening spectrum was selected as the window for the S parameter, where  $p_L$  is an electron-momentum,  $c$  the speed of light and  $m$  the electron-mass at rest. X-ray diffraction (XRD) was performed using a Rigaku Ultima IV to determine the crystallinity of the samples.

## 3. Results and discussion

No crystalline peaks were observed in any of the XRD patterns from the bulk glassy alloys, whether obtained either before or after irradiation. This is different to behavior reported for thin-film metallic glasses where nanocrystals are formed after both light- and heavy-ion irradiation [4, 8]. This difference must therefore be due to the difference in sample size.

All the positron lifetime spectra were analyzed based on the one lifetime component shown in Table 1. The positron lifetime increases only after electron and 180 keV He irradiation. This indicates that the radiation has increased the open volume size (free volume). On the other hand, the positron lifetime was unchanged by irradiation with heavy Xe-ions

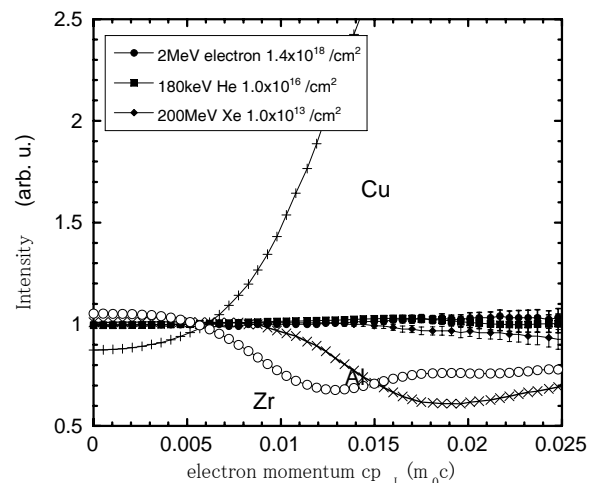


Figure 1 CDB spectra of ZrCuAl bulk glassy alloy irradiated with electron, He and 200MeV Xe ions, expressed as a ratio of CDB intensity of irradiated specimens to that of the as-prepared specimen.

Table 1 Positron lifetime of bulk ZrCuAl glassy alloy before and after irradiation.

	total dose (/cm <sup>2</sup> )	incident positron energy	positron lifetime difference $\Delta\tau$ (ps)
before irradiation		15 keV, CSC	[191]
180 keV He	$1 \times 10^{16}$	15 keV	+23
2.5 MeV Xe	$1 \times 10^{14}$	15 keV	-3
200 MeV Xe	$1 \times 10^{13}$	CSC	-3
2 MeV electron	$1 \times 10^{18}$	CSC	+6

CSC: classical sandwich configuration method using RI source

at either energy of 2.5 or 200 MeV. In the case of the 200 MeV Xe ion-irradiated sample measured by the CSC method, the change in positron lifetime of 3 ps is underestimated because the maximum damage depth of  $\sim 14 \mu\text{m}$  is shallower than implantation depth of positrons emitted from the  $^{22}\text{Na}$  radioactive source. We suggest that this difference in behavior can be attributed to the difference in displacements per radiation particle (dpp), because the dpp estimated for 2 MeV electrons and 180 keV He ions is approximately three or more orders of magnitude less than that for heavy Xe ions. Under these radiation sources, an increasing positron lifetime indicates the introduction of vacancy-type defects, and decreasing positron lifetime reveals the disappearance and shrinking of free volume due to structural relaxations and/or by crystallization. Similar changes due to structural relaxation and crystallization caused by heat treatment have been reported elsewhere [10-12]. In our experiments, however, the XRD results show that crystallization does not take place under any irradiation investigated, therefore, the decrease of positron lifetime with Xe-ion irradiation can only be due to structural relaxations, with the energy provided not through heat treatment but via the particle energy. It follows then that the relaxation processes caused by irradiation are not necessarily the same as those from the movement of thermally activated atoms. No significant changes were observed in the CDB spectra for any irradiation, as shown Figure 1. In the case of He- and Xe-ion irradiation using the CSC method, not all positrons annihilate within the damaged layer, hence the change in CDB ratio is small. It has been reported that the CDB ratio changes during crystallization but does not change during structural relaxation [12, 13]. Therefore, we can consider these irradiation treatments are not especially effective for local atomic reordering around the free volume.

Figures 2(a), (b), and (c) show the Doppler broadening S parameter as a function of depth after 180 keV He-ion irradiation. Also shown is the irradiation damage profile from 180 keV He-ion irradiation on

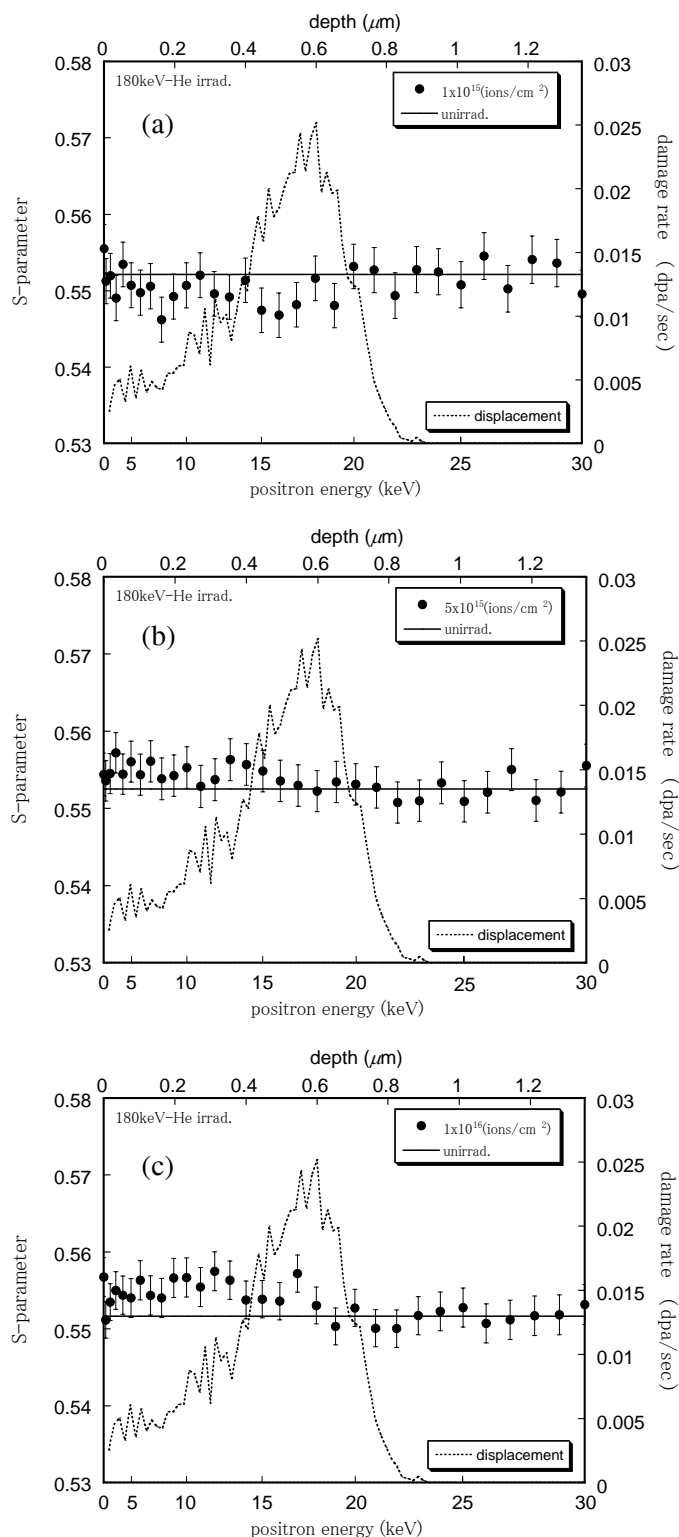


Figure 2 Positron annihilation Doppler S parameter in ZrCuAl bulk glassy alloy as a function of depth after 180 keV He-ion irradiation. Also shown is the irradiation damage profile. Total dose: (a)  $1 \times 10^{15}$  ions/cm<sup>2</sup>, (b)  $5 \times 10^{15}$ , and (c)  $1 \times 10^{16}$  ions/cm<sup>2</sup>.

the ZrCuAl bulk glassy alloy. The S parameter bulk value is defined as the mean value in the non-irradiated area between 22 and 30 keV in each sample, and the damage profile was calculated using SRIM2006 code [14]. The maximum damage depth for this radiation was estimated to be approximately 0.8  $\mu\text{m}$ . In Figure 2(a), the S parameter does not increase despite the high irradiation fluence of  $1 \times 10^{15}$  ions/cm<sup>2</sup>. However, at higher doses the S parameter is slightly increased in the damaged layer from the surface to 0.6  $\mu\text{m}$ , as shown in Figures 2(b) and (c). This result reveals that the He-ion radiation is effective at introducing vacancy-type atomic defects producing excess free volume. According to the positron lifetime results given in Table 1, the average size of the open volume in the bulk glassy alloy before, and then after He-ion irradiation, corresponds roughly to single or di-vacancy, or larger than metallic tri-vacancy, respectively [15]. Considering the fact that in this case the irradiated material is not crystalline but amorphous, a conclusive determination of the number of vacant sites included in the open volume and its structure cannot be made. Consequently, all that can be concluded is that the open volume, which is not necessarily the same intrinsic free volume in the as-prepared glassy alloy, increases with 180 keV He-ion irradiation.

#### 4. Summary

Free volume changes in a Zr<sub>50</sub>Cu<sub>40</sub>Al<sub>10</sub> bulk glassy alloy after electron and ion irradiation were studied by employing positron annihilation spectroscopy. Different trends in the free volume change were observed for the various radiation species. Specifically, the free volume increased only under electron and 180 keV He-ion radiation. The increase in free volume within the damaged region was measured during 180 keV He-ion irradiation using the slow positron beam technique. We found that no crystallization in the bulk glassy alloy took place under any radiation source or by changing irradiation parameters such as energy, flux, total dose or irradiation temperature. We also found that the radiation effect in ZrCuAl bulk glassy alloy is dependent on the displacement per radiation particle.

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