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Effects of co-implanted oxygen or aluminum atoms on hydrogen migration and damage structure in multiple-beam irradiated Al_2O_3

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Abstract

Depth profiles of implanted H atoms were measured for single crystalline Al_2O_3 samples irradiated at 923 K with dual or triple beams of 0.25 MeV H-, 0.6 MeV He-, 2.4 MeV O-ions or 2.6 MeV Al-ions. The peaks occur at 1.55 and 1.45 μm in the depth profiles measured for the H + Al dual beam irradiation and H + O dual beam case, respectively. The ratio of the peak areas is over 4, which is much larger than the implanted H atom ratio of 1.1, indicating that implanted Al atoms suppress the mobility of H atoms. However, the ratio becomes almost 1 between the triple beam samples with H + He + O-ions and with H + He + Al-ions at comparable doses. The fact demonstrates that implanted He atoms overwhelm the effects of the implanted self-cation/anion excess atoms on the migration behaviors of implanted hydrogen and radiation produced point defects, with the resulting sluggish cavity growth observed. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

The synergetic effects of atomic displacement damage and implanted H, He atoms in Al_2O_3 have been studied using samples irradiated with multiple beams of hydrogen (H), helium (He) and oxygen (O) ions from TIARA (Takasaki Ion Accelerator for Advanced Radiation Application) at Takasaki establishment of JAERI. These works have been conducted in view of the importance of the synergetic effects since Al_2O_3 has been proposed for electrical insulators and diagnostic materials in fusion reactors. Although important findings have been reported already, such as the fact that hydrogen atoms enhance the growth of dislocation loops decorated with tiny cavities, while the concurrent existence of He atoms retards the dislocation growth [1,2], the new experiments have been made using self-O ions along with H and/or He-ions. It remains unclear how the implanted excess O atoms play roles in developing the damage structures or migration behaviors of H and He atoms.

In the present study, we have measured depth profiles of implanted H atoms in Al_2O_3 samples simultaneously irradiated with beams of H, He, O or Al ions and also investigated the damage structures by cross-sectional transmission electron microscopy (XTEM). The results are discussed in terms of mobilities of point defects influenced by the presence of excess O and Al atoms as well as H and He atoms under irradiation.

2. Experimental procedure

The materials used in this study were single crystal alumina (α-Al_2O_3).The procedures of sample preparation and irradiations have been described elsewhere [3]. The samples were irradiated mainly with dual and triple

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Table 1
Irraditation conditions and corresponding damage parameters obtained by TRIM calculations[a]

Ion beams (sample ID)	Energy (MeV/Ion)	Flux (10^{16}(ions/m^{2}s)	Fluence (10^{20}ions/m^2)	Peak damage (dpa)	Peak ion concentration (at.%)
Single					
(#97-01)	0.25 H^+	6.0	6.6	0.10	4.8
Dual					
(#95-11)	0.25 H^+	4.0	5.0	0.09	3.7
	2.4 O^{2+}	6.0	7.6	10.5	2.8
(#96-08)	0.25 H^+	3.2	5.8	0.1	4.2
	2.6 Al^{2+}	2.1	3.8	10.1	1.1
Triple					
(#94-05)	0.25 H^+	3.6	2.3	0.04	1.7
	0.6 He^+	2.4	1.5	0.3	0.9
	2.4 O^{2+}	3.3	2.1	3.3	1.3
(#96-03)	0.25 H^+	2.7	2.2	0.04	1.6
	0.6 He^+	2.3	1.9	0.35	1.05
	2.6 Al^{2+}	1.7	1.4	3.7	0.4

[a] Depths (μm) of (damage peaks and average project ranges) for 0.25 H^+, 0.6 He^+, 2.4 O^{2+} and 2.6 Al^{2+} are: (1.38, 1.44), (1.34, 1.39), (1.34, 1.41) and (1.47, 1.56), respectively.

beams of (0.25 MeV H^+, 0.6 MeV He^+, 2.4 MeV O^{2+}or 2.6 MeV Al^{2+}) ions at 923 K (0.4 Tm). The ion energies are so chosen that the projected ranges of the irradiated ions in Al_2O_3 coincide with depths of 1.4–1.6 μm. Table 1 summarizes the irradiation conditions examined and respective damage parameters obtained with TRIM89 code [4], using a common displacement energy of 40 eV for Al and O atoms. After irradiation, XTEM observations were performed using a JEM-2000FX electron microscope operating at 200 kV.

Depth profiles of implanted H atoms in the as-irradiated samples were measured utilizing a resonant nuclear reaction analysis (NRA) of ^{15}N, $^{15}N(p, \alpha, \gamma)^{12}C$, details of which have been already described in Ref. [5].

3. Results and discussion

Depth profiles of H atoms concentrations as measured and expressed with γ-ray counts are given in Fig. 1 for the sample irradiated with the single beam of 0.25 MeV H^+ions, comparing with the calculated result by TRIM89 code. In Fig. 2, the corresponding measurement results for the sample irradiated with the dual beams of (0.25 MeV H^+, 2.6 MeV Al^{2+}) ions are compared with those for dual (0.25 MeV H^+, 2.4 MeV O^{2+}) beams. And in Fig. 3, the results are compared for the sample irradiated with triple beams of (0.25 MeV H^+, 0.6 MeV He^+, 2.6 MeV Al^{2+}) ions and the one with (0.25 MeV H^+, 0.6 MeV He^+, 2.4 MeV O^{2+}) ions.

In these comparisons of migration behaviors of H atoms, the depth profile of H atoms in the sample irradiated at 923 K with single H beam to a peak concentration of 4.8 at.% under the lowest displacement damage of 0.1 dpa at the peak is taken for the reference.

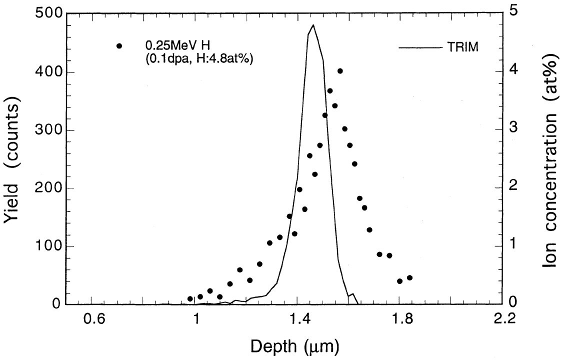

Fig. 1. Depth profiles of hydrogen concentrations as measured by NRA in single crystal α-Al_2O_3 irradiated with single 0.25 MeV H^+ ion beam irradiation, in comparison with a calculation by TRIM code.

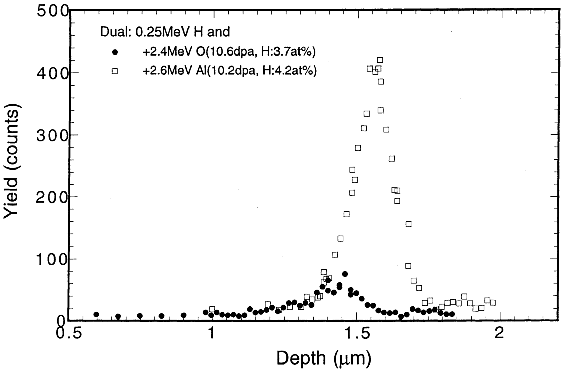

Fig. 2. Depth profiles of hydrogen concentrations as measured by NRA in single crystal α-Al_2O_3 irradiated with dual (0.25 MeV H^+, 2.6 MeV Al^{2+}) beams at 923 K, which are compared with those irradiated with dual beams of (0.25 MeV H^+, 2.4 MeV O^{2+}) ions.

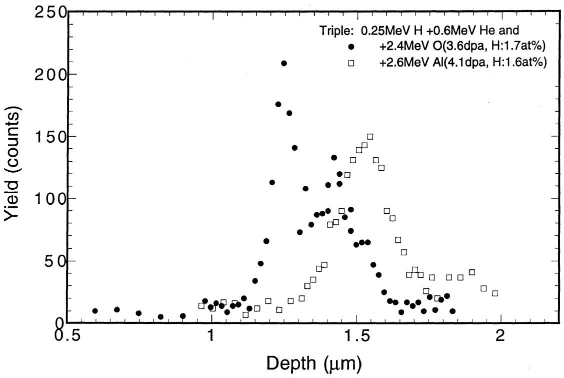

Fig. 3. Depth profiles of hydrogen concentrations as measured by NRA in single crystal α-Al_2O_3 irradiated with triple (0.25 MeV H^+, 0.6 MeV He^+, 2.6 MeV Al^{2+}) beams at 923 K, which are compared with those of triple (0.25 MeV H^+, 0.6 MeV He^+, 2.4 MeV O^{2+}) beam irradiation.

The measured profile exhibits a peak at a depth of 1.55 μm (Fig. 1), which is 6% deeper than that predicted by the TRIM code, i.e., by 0.1 μm. The full width at a half maximum for the measured peak is 0.24 μm, which is 1.8 times larger than the predicted one. Moreover, the profile shows a long tail towards the irradiation surface. These facts indicate that the considerable diffusion of H atoms occurs in this sample under the lowest displacement damage of 0.1 dpa along the steep gradient of implanted H atom concentrations.

The measured depth profile of H atoms implanted with dual beams of H + O ions to respective peak concentrations of 3.7 and 2.8 at.% and to 10.6 dpa at peak is observed to be slightly peaked at a depth of 1.45 μm, while the higher peak occurs at a depth of 1.55 μm for the dual beam irradiations with H + Al ions to respective peak concentrations of 4.2 and 1.1 at.% and to 10.2 dpa (Fig. 2). The peak area in the measured depth profiles of H atoms for H + Al dual irradiation, which represents the yet-migrated-away H atoms, is over four times larger than that for the H + O dual case. This value is markedly larger as compared with the ratio of 1.1 for implanted H atom concentrations under the two comparable displacement damage levels. Moreover, it should be noted that the profile of the H + Al dual case is less broadened and more symmetric around the peak than that observed in the single H beam irradiated sample (Fig. 1). The facts indicate that implanted Al atoms, which behave as excess cations in Al_2O_3 suppress mobilities of H atoms implanted. Furthermore, as is shown in Fig. 4, cavities observed in an extremely high density around the ions' ranges for H + Al dual irradiation are much smaller sized, 3–4 nm than those observed to be over 15 nm for the H + O dual case, with a low density of developed dislocation loops around depths of 1.4–1.7 μm [5,6]. This also suggests that implanted Al atoms suppress mobilities of radiation produced point defects. In addition, it is worth noting that suppressed mobility of implanted H atoms and retarded formation of cavities as well are also observed in the sample irradiated with dual

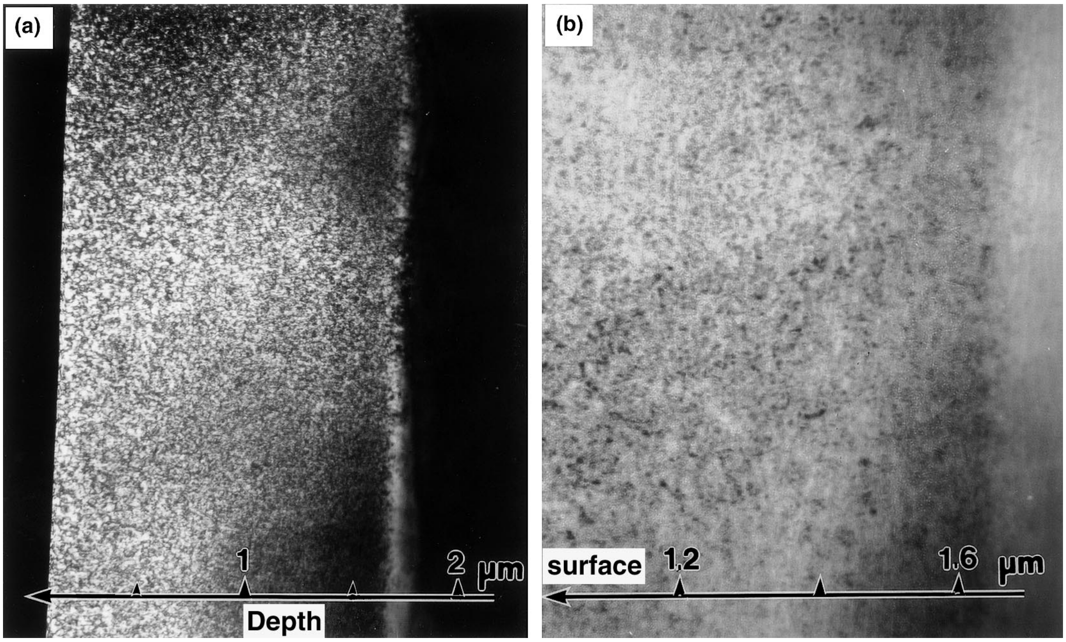

Fig. 4. Cross-sectional damage structures of α-Al_2O_3 irradiated with dual (0.25 MeV H^+, 2.6 MeV Al^{2+}) beam at 923 K (a), and structures around ion ranges (b).

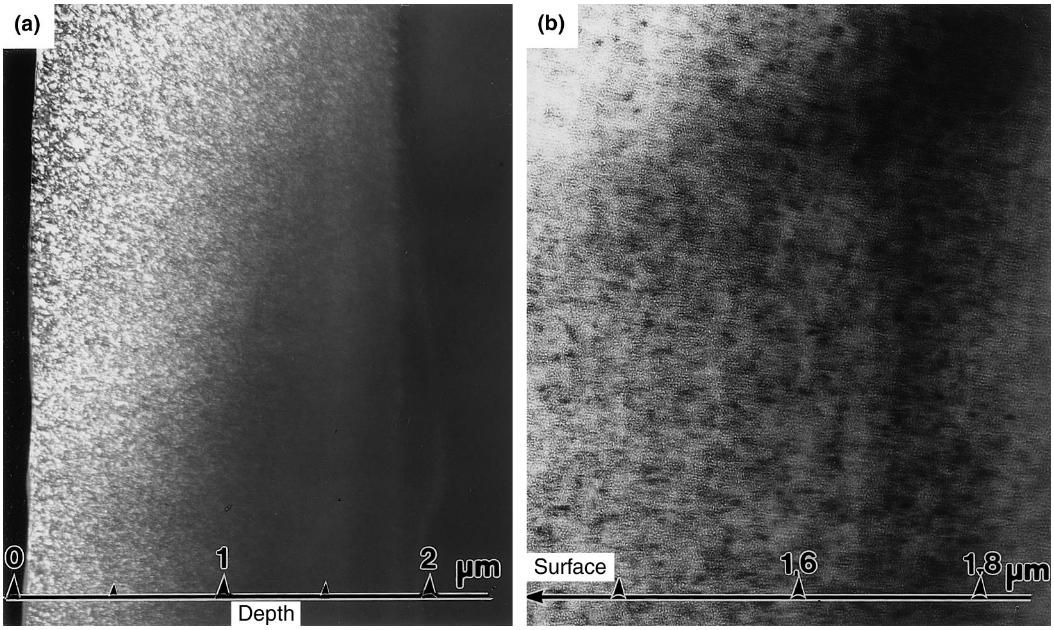

Fig. 5. Cross-sectional damage structures of α-Al_2O_3 irradiated with triple (0.25 MeV H^+, 0.6 MeV He^+, 2.6 MeV Al) beams at 923 K (a), and structures around ion ranges (b).

beams of (0.25 MeV H^+, 2.8 MeV Si^{2+}) ions to respective peak concentrations of 3.5 and 0.9 at.% and to 6.6 dpa at peak at 923 K [7]. The fact confirms that implanted cation atoms suppress mobilities of both implanted H atoms and radiation produced point defects.

The difference in the peak areas for the triple beam irradiations with a He-ion beam involved becomes diminished between the two triple beam irradiations; the one with H + He + O ions to respective peak concentrations of 1.7, 0.9 and 1.3 at.% and to 3.6 dpa at peak, and the other with H + He + Al ions to respective peak concentrations of 1.6, 1.1 and 0.4 at.% and to 3.7 dpa, as shown in Fig. 3. This equivalency of the peak areas for both the samples indicates that most of the implanted H atoms are trapped in cavities nucleated as agglomerates of He atoms and vacancies. However, for the triple beam irradiation with H + He + O ions, two peaks occur at 1.25 and 1.45 μm (Fig. 3); the peak height of the former is 1.5 times larger than the latter, with the area of both being almost same. On the other hand, the large peak occurs at 1.55 μm for the H + He + Al ion irradiation with the lower second peak at 1.9 μm being less clear. In addition to an appearance of the double peaks, for the H + He + O irradiation, note that respective peak position is deviated largely from that observed in the single H beam irradiation by 0.3 and 0.15 μm toward the irradiation surface. It is considered that large front peak at 1.25 μm would be produced by H atoms diffusing toward the surface along the steep gradient of displacement damages the O ions under the enhanced mobilities of H atoms due to a co-existence of O atoms.

The occurrence of the large peak at 1.55 μm for the H + He + Al ion irradiation is seen to exhibit the superposed effects for Al and He atoms to trap H atoms. Suppressed mobilities of both H atoms and the radiation produced point defects are evidenced by comparing the depth dependent microstructures which are given in Fig. 5 for the H + He + Al ion irradiation and in Ref. [2] for the triple beam irradiation with H + He + O ions. Tiny cavities sized at a narrow range of 2–3 nm are formed uniformly at depths of 0.5–1.8 μm in the H + He + Al ion irradiated sample and an irresolvably high density of dislocation loops is formed uniformly from the irradiation surface to 1.8 μm. On the other hand, cavities sized 13 nm in an average were distributed in the restricted range of depths from 1.2–1.7 μm in the H + He + O ion irradiated sample [2], exhibiting comparatively enhanced cavity growth.

4. Summary

Migration behaviors of hydrogen atoms in single crystalline Al_2O_3 samples irradiated at 923 K with dual or triple beams of 0.25 MeV H-, 0.6 MeV He-, 2.4 MeV O-ions or 2.6 MeV Al-ions were examined by measuring depth profiles of implanted H atoms through a resonant nuclear reaction analysis with ^{15}N.

The present results demonstrate that implanted Al atoms in Al_2O_3 suppress mobilities of H atoms implanted, as compared with the observed enhanced mobilities under concurrently implanted excess O atoms. It is further suggested that implanted Al atoms suppress mobilities of radiation produced point defects also. However, implanted He atoms overwhelm the influence of implanted self-cation/anion excess atoms on the migration behaviors of implanted hydrogen and radiation produced point defects.

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