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Radiation damages in CeO_2 thin films irradiated with ions having the same nuclear stopping and different electronic stopping powers

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ABSTRACT

In order to characterize a possible modification due to high-density electronic excitation, thin films of CeO_2 have been irradiated with 10-MeV Ni and 120-MeV Xe ions, both having the same nuclear stopping powers, while having different electronic stopping powers. The comparison of the fluence dependence of X-ray diffraction (XRD) peaks suggests that the defect production cross section for 120-MeV Xe is nearly one order of magnitude higher than that for 10-MeV Ni. The comparative study demonstrates the prominent damage observed for 120-MeV Xe irradiation is due to high electronic energy deposition.

1. Introduction

Oxide ceramic fuels in nuclear reactors are subjected to various high-energy particles. Especially, fission fragments have high kinetic energy of typically about 70–100 MeV, and create radiation damages in UO_2 oxide fuel [1], leading to degradation of fuel properties such as swelling and decrease in thermal conductivity. For investigating damage mechanism of oxide fuels, ion accelerator can be a powerful tool to simulate damage behavior under high-energy heavy ions, since accelerator provides ions having well-defined energy, and the fluence can be precisely controlled. For the simulation of radiation damage of oxide fuels, in the present study cerium dioxide, CeO_2, is adopted as an irradiating target, since it has the same structure (fluorite structure) as UO_2.

One of the intriguing characteristics of the high-density electronic excitation effect is the formation of continuous ion tracks along ion-paths [2], whose damage structure is different from the damage created via elastic displacement process. The crystallographic structure inside the ion track is usually modified in a way different from that of an elastic collision cascade process [3].

It is already known by transmission electron microscopy observation [4] that ion tracks are observed for CeO_2 polycrystalline sample irradiated with 210-MeV Xe ions. It is important from the fundamental point of view how the electronic energy deposition contributes to structural change of CeO_2 irradiated with high-energy heavy ions. The energy deposition from ions consists of two independent processes; the electronic energy deposition which is defined by the electronic stopping power, and the energy deposition due to elastic collisions which is defined by the nuclear stopping power. Since both of these processes may contribute to defect creation at the same time, it is difficult to characterize effects only by the electronic energy deposition. In this study the effect due to 10-MeV Ni ion irradiation and that due to 120-MeV Xe ion irradiation are compared. Here, these ions have the same nuclear stopping power, while having different electronic stopping powers. The purpose of the present study is to demonstrate that the effect originated only from the difference in the electronic stopping power can be characterized by such comparative investigation.

2. Experimental procedure

Thin films of CeO_2 were prepared on single crystal sapphire substrates by a sputtering method. The film thickness was about 0.3 μm. The films were irradiated with 120-MeV Xe ions from the tandem accelerator at JAEA-Tokai and with 10-MeV Ni ions from the tandem accelerator at JAEA-Takasaki. The both irradiations were performed at room temperature. In order to investigate the irradiation-induced modification of crystal structure, Wide-angle X-ray diffraction (XRD) profiles were measured before and after the irradiations by X-ray diffractometer (MAC Science, MXP model 1030) using Cu radiation and operated by 40 kV and 40 mA. In this study, the irradiation-induced change of the XRD peak corresponding to (0 0 2) reflection is analyzed.

The irradiating ions were chosen by taking into account of the values of the nuclear stopping power and the electronic stopping power. Table 1 shows that both 10-MeV Ni ion and 120-MeV Xe have the same nuclear stopping power when irradiated to CeO_2, suggesting that both ions are expected to have the same effect when it comes to elastic displacement process. On the other hand,

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Table 1
Nuclear stopping power (S_n) and electronic stopping power (S_e) for 10-MeV Ni and 120-MeV Xe when irradiated to CeO_2.

	S_n (keV/nm)	S_e (keV/nm)
10-MeV Ni	0.18	6.1
120-MeV Xe	0.18	23.5

these ions have different electronic stopping powers, suggesting that the difference in observed damage, if any, is attributable to the difference in the electronic stopping power. By using such combination of ions, the influence originated from the difference in the electronic stopping power can be investigated.

The nuclear stopping power, the electronic stopping power and the projected range were estimated using SRIM-2008 [5,6]. The projected ranges for 10-MeV Ni ions and 120-MeV Xe ions are 3.1 μm and 8.6 μm, respectively. They are much larger than the film thickness (0.3 μm). Therefore, defects are uniformly introduced along the sample thickness, and the possibility of ion implantation can be excluded.

3. Results and discussion

Since the CeO_2 thin films are highly oriented, only (0 0 2) and (0 0 4) peaks are the observable peaks in the reflection angle range of $2\theta = 5–100°$. Large peaks originated from single crystalline Al_2O_3 substrate are always observed, but they do not disturb the observation of CeO_2 peaks. Fig. 1 shows the evolution of the (0 0 2) diffraction peak for CeO_2 irradiated with 10-MeV Ni ions (Fig. 1(a)) and with 120-MeV Xe ions (Fig. 1(b)). A monotonic decrease in the peak intensity as a function of the fluence is observed for both irradiations. The same tendency of the peak intensity is observed for (0 0 4) peak, therefore only the result for (0 0 2) peak is presented in this paper. The intensity decrease can be attributed to a defect creation by ion-irradiation. The fluence dependence of the lattice parameter estimated from the shift of the peak angle is shown in Fig. 2. In the figure, the fluence axis of 10-MeV Ni ion is shifted one order of magnitude relative to that of 120-MeV Xe ion in order to facilitate the comparison of the data. Both of the fluence dependences do not exhibit a monotonic behavior. It should be noted here that such a complex fluence dependence is not explainable before taking into account the break of peak symmetry appeared after the irradiations. The detailed analysis will be performed in the last part of this section. The fluence dependence of the peak width (full width at half maximum) of the observed peak is shown in Fig. 3. The fluence dependence of the maximum intensity of the observed peak is shown in Fig. 4. In these figures, the fluence axis of 10-MeV Ni ion is again shifted one order of magnitude relative to that of 120-MeV Xe ion. Since the change in peak profiles is attributed to defect creation, the results can be interpreted that defects created by 120-MeV Xe ion amount to nearly 1 order of magnitude larger than those created by 10-MeV Ni ion. The comparison of the fluence dependence of the peak width also makes a result consistent with this interpretation. The one order of magnitude difference in the defect density per incident ion is attributable to the difference in the electronic stopping power. Here, it is reasonable to assume that the energy deposition via elastic collisions for 120-MeV Xe ion and for 10-MeV Ni equally contributes to defect creation, since these ions have the same nuclear stopping power. Therefore, the present comparative study clearly demonstrates that the difference in the damage creation behavior is attributed to the difference in the electronic energy deposition, leading to a much higher contribution to damage creation for 120-MeV Xe ion irradiation than that for 10-MeV Ni ion irradiation.

It is already demonstrated that, by analyzing the fluence dependence of XRD profile, the size of the region continuously modified

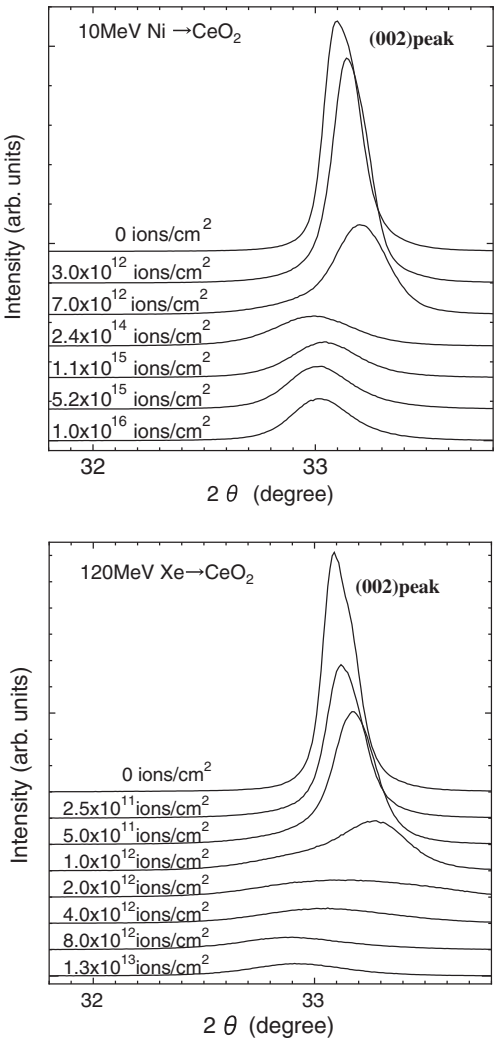

Fig. 1. Evolution of the X-ray diffraction peak assigned for (0 0 2) reflection of CeO_2 irradiated with (a) 10-MeV Ni ions and (b) 120-MeV Xe ions.

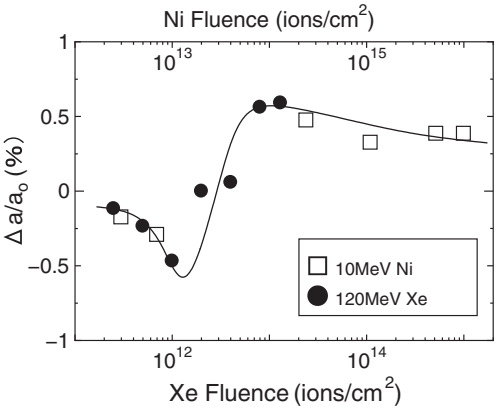

Fig. 2. Fluence dependence of lattice parameter change derived from the shift of the maximum position of the peak for 10-MeV Ni ion irradiation and 120-MeV Xe ion irradiation. The change in lattice parameter, Δa, is normalized to the lattice parameter before irradiation, a_o. The solid curve is just a guide for the eyes.

by the electronic energy deposition (the size of the ion-track) can be estimated when the modified region is amorphized [7–10]. This method is based on the assumption that the irradiation-induced amorphized region does not contribute to X-ray diffraction, and the diffraction from undamaged matrix is diminished as increasing