Nuclear Instruments and Methods in Physics Research B 314 (2013) 191–194

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Effects of high-energy heavy ion irradiation on the crystal structure in CeO_2 thin films

T. Kishino [a,*], K. Shimizu [a], Y. Saitoh [b], N. Ishikawa [c], F. Hori [a], A. Iwase [a]

[a] Department of Materials Science, Osaka Prefecture University, Sakai, Osaka 599-8531, Japan
[b] Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency, Takasaki, Gumma 370-1292, Japan
[c] Tokai Research and Development Center, Japan Atomic Energy Agency, Tokai, Ibaragi 319-9915, Japan

ARTICLE INFO

Article history:
Received 29 November 2012
Received in revised form 9 April 2013
Accepted 10 April 2013
Available online 19 April 2013

Keywords:
CeO_2 thin films
Ion-irradiation
Electronic excitation
Lattice parameter

ABSTRACT

We studied the change in crystal structure of 200 MeV Xe and 10 MeV I ion irradiated CeO_2 thin films by means of X-ray diffraction (XRD). The experimental result showed that the average lattice parameter of CeO_2 decreased and the full width at half maximum increased by the ion irradiation. Their changes are well correlated with the electronic stopping powers for I and Xe ions. The dependence of lattice parameter on the electronic energy loss was analyzed by using the Poisson's law, and the radii of the regions affected by the irradiation were determined.

1. Introduction

In our previous study [1], pure CeO_2 pellets were irradiated with 200-MeV Xe^{14+} ions and the effects of the irradiation on magnetic properties and the lattice structure were measured. And we have reported that the specimens show the ferromagnetic behavior by the ion irradiation. We have explained this phenomenon as follows; the observed ferromagnetic behavior in CeO_2 pellets is attributed to some magnetic interaction of 4f electrons at Ce^{3+} atoms which exist near irradiation-produced oxygen vacancies. It is quite interesting if the high density electronic excitation contributes to the production of oxygen vacancies. However, as the range for the incident ions (about 12 μm) is much smaller than the thickness of CeO_2 bulk (about 0.5 mm), effects of ion irradiation are not uniform in specimens, and it is very difficult to discuss the irradiation effects quantitatively. In this study, we prepared CeO_2 thin films by the RF magnetron sputtering method. They are much thinner than the range of incident ions, and we can obtain uniform irradiation effects in the specimens. The effect of 200 MeV Xe irradiation and 10 MeV I irradiation on the crystal structure is discussed in terms of the energies deposited through the electronic excitation and the elastic collisions.

* Corresponding author. Tel.: +81 72 254 9810.
E-mail address: ss110011@edu.osakafu-u.ac.jp (T. Kishino).

0168-583X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.04.028

2. Experimental procedure

Specimens used in this study were CeO_2 thin films. They were deposited on r-Al_2O_3 substrates by the RF magnetron sputtering method. The sputtering conditions was as follows; the substrate temperature was 650 °C, the sputtering time was 1.5 h, the sputtering gas was Ar + 7%O_2, the sputtering pressure was 7 Pa, and the RF power was 100 W. The thickness of the film was 23 nm. The lattice structure of the as-deposited CeO_2 thin films specimens was estimated by means of X-ray diffraction (XRD).

Then the CeO_2 thin films were irradiated at room temperature with 10-MeV I ions using a tandem accelerator at Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency and with 200-MeV Xe ions at Tokai Research and Development Center, Japan Atomic Energy Agency. The ion fluences were 3.5×10^{12}, 7.0×10^{12}, and 2.0×10^{13} /cm^2, for both of the ions.

In order to investigate the change in the lattice structure of CeO_2 thin film by the irradiations, we used a conventional Cu-Kα X-ray diffractometer.

The electronic stopping power, Se, the nuclear stopping power, Sn, and the projected range, R, were estimated using SRIM-2003. The values of Se, Sn and R for 10 MeV I ion and 200 MeV Xe ion are listed in Table 1. The values of R are much larger than the film thickness (23 nm). Therefore, defects are uniformly introduced along the specimen thickness and the possibility of ion implantation effects can be excluded.

Table 1
Electronic stopping power, Se, nuclear stopping power, Sn, and projected range for 10 MeV I and 200 MeV Xe ion. Target is CeO_2.

	Se(keV/nm)	Sn(keV/nm)	R(μm)
10 MeVI	4.35	0.992	2.23
200 MeV Xe	27.7	0.114	12.7

3. Results and discussion

First, the lattice structure of CeO_2 thin films deposited on r-Al_2O_3 substrates by the RF magnetron sputtering method is discussed. Fig. 1 shows the widely-scanned X-ray diffraction (XRD) spectra for Al_2O_3 substrate and for CeO_2 thin film on it. After the sputtering, (0 0 2) and (0 0 4) diffraction peaks newly appear in addition to Al_2O_3 peaks. These peaks correspond to the fluorite structure. Other diffraction peaks for the fluorite structure are hardly observed, indicating that the CeO_2 thin films are highly c-axis oriented on the Al_2O_3 substrate.

Next, in order to investigate the irradiation-induced modification of crystal structure, the (0 0 4) peaks before and after the irradiation were analyzed in detail. Fig. 2 shows the (0 0 4) diffraction peaks for CeO_2 thin films irradiated with 10-MeV I (Fig. 2a) and for those with 200-MeV Xe (Fig. 2b) for various ion fluences. With increasing the ion fluence, the peaks shift to higher angles, and a monotonic decrease in peak intensity and a peak broadening are also observed for both irradiations. These phenomena indicate that the average lattice parameter of CeO_2 films decreases and the crystal structure is disordered by the irradiation. But the peaks for the Al_2O_3 substrate scarcely change their position by the irradiation. This result implies that the structure of the substrates is not influenced by the irradiation, and that the CeO_2 peak shift surely shows the lattice shrinkage of CeO_2 films by the irradiation. Fig. 3(a) shows the ion fluence dependence of the lattice parameter of the CeO_2 films for 10-MeV I and 200-MeV Xe irradiations, which has been derived from the shift of the XRD peaks by the irradiations. The lattice parameter was calculated by using the Bragg's equation. With increasing the ion fluence, a monotonic decrease in lattice parameter is observed, and the effect of 200 MeV Xe ions is larger than that of 10 MeV I ions when compared at the same ion fluence. In our previous study, however, the average lattice parameter of CeO_2 bulk specimens was increased by 200 MeV Xe irradiation [1,2]. In CeO_2 bulk specimen, the lattice expansion has been explained as originating from the irradiation-induced oxygen deficiencies. When oxygen vacancies were introduced by the irradiation, Ce atoms would be repulsed each other, leading to the lattice expansion. Here, we discuss the difference in the irradiation

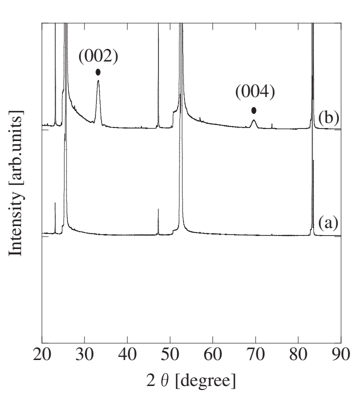

Fig. 1. XRD spectra for (a) Al_2O_3 substrate and (b) CeO_2 deposited on Al_2O_3 substrate.

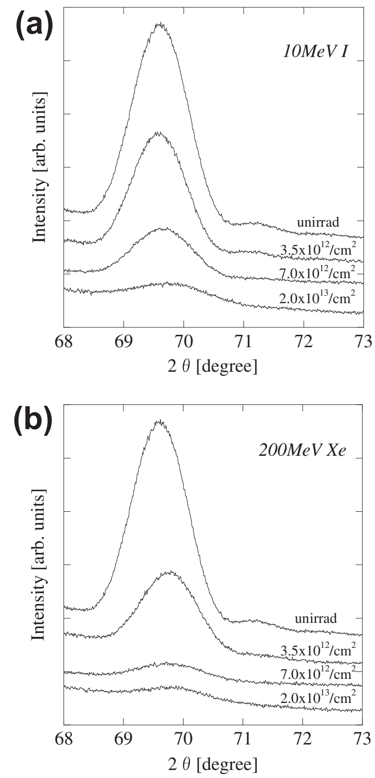

Fig. 2. (0 0 4) diffraction peaks for (a) 10-MeV I^{3+} irradiation and (b) for 200-MeV Xe^{14+} irradiation. The ion fluences are 3.5×10^{12}, 7.0×10^{12} and $2.0 \times 10^{13}/cm^2$.

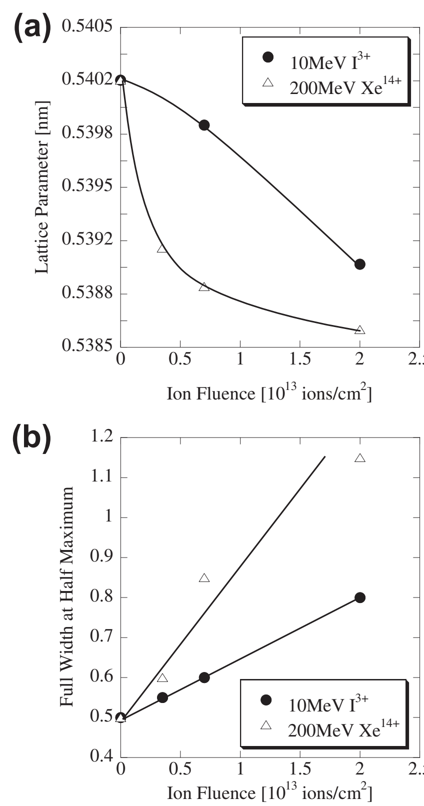

Fig. 3. (a) Change in lattice parameter of CeO_2 thin films and (b) change in full width at half maximum of (0 0 4) peaks by the irradiation with 10-MeV I^{3+} and 200-MeV Xe^{14+}.