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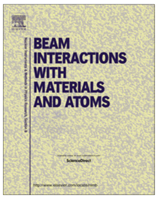

Heavy and light ion irradiation damage effects in δ-phase $Sc_4Hf_3O_{12}$

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ABSTRACT

Polycrystalline δ-phase $Sc_4Hf_3O_{12}$ was irradiated with light and heavy ions to study the radiation stability of this compound. In order to explore the ion species spectrum effect, the irradiations were performed with 400 keV Ne^{2+} ions to fluences ranging from 1×10^{14} to 1×10^{15} ions/cm^2, 600 keV Kr^{3+} ions to fluences ranging from 5×10^{14} to 5×10^{15} ions/cm^2, and 6 MeV Xe^{26+} ions to fluences ranging from 2×10^{13} to 1×10^{15} ions/cm^2. Irradiated samples were characterized by various techniques including grazing incidence X-ray diffraction (GIXRD) and transmission electron microscopy (TEM). A complete phase transformation from ordered rhombohedral to disordered fluorite was observed by a fluence of 1×10^{15} ions/cm^2 with 400 keV Ne^{2+} ions, equivalent to a peak ballistic damage dose of ~0.33 displacements per atom (dpa). Meanwhile, the same transformation was also observed by 600 keV Kr^{3+} ions at the same fluence of 1×10^{15} ions/cm^2, which however corresponds to a peak ballistic damage dose of ~2.2 dpa. Only a partial O-D transformation was observed for 6 MeV Xe^{26+} ions in the fluence range used. Experimental results indicated that the O-D transformation is observed under both electronic and nuclear stopping dominant irradiation regimes. It was also observed that light ions are more efficient than heavy ions in producing the retained defects that are presumably responsible for the O-D phase transformation. The O-D transformation mechanism is discussed in the context of anion oxygen Frenkel defects and cation antisite defects. We concluded that the irradiation induced O-D transformation is easier to occur in δ-phase compounds with partial order of cations than in that with fully disordered cation structures.

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1. Introduction

With the expanded application of nuclear power all over the world in the past half-century, substantial quantities of nuclear waste, such as plutonium and minor actinides (Np, Am and Cm), have been generated from nuclear fuel cycles. A considerable amount of research has been done in an effort to search for radiation-tolerant materials that can be used as an inert matrix in nuclear fuels and as host materials for minor actinides in high-level radioactive waste [1–5]. Such waste form materials should have a high capacity for incorporating actinides, possess a high radiation resistance and excellent chemical stability. Crystalline oxides that possess the fluorite (CaF_2) structure or the derivatives of the fluorite structure have received considerable attention as candidate materials for nuclear-waste host. The compounds of interest in this investigation are oxygen-deficient fluorite structural derivatives MO_{2-x} (M represents a metal cation, while O refers to an oxygen anion). The compounds stoichiometry plays a great role on the effect of structural stability under irradiation in certain fluorite derivative structures. Isometric pyrochlore structure, $A_2B_2O_7$, is a promising host phase for the immobilization of actinides [6–11]. For instance, in the binary system $Gd_2Ti_{2-x}Zr_xO_7$, the resistance to ion beam irradiation-induced amorphization increases dramatically with increasing Zr content, because of the cation and anion substructures disordered gradually as Zr content increases [2,12–14]. These studies illustrate that the irradiation tolerance of pyrochlore structure ($A_2B_2O_7$) is not only affected by the radius ratio r_A/r_B and type of A–O and B–O bonds, but also affected by the level of initial cation structural disordering [11,15].

Another family of radiation tolerant fluorite-derivative oxides $A_4B_3O_{12}$ known as δ-phase possesses rhombohedral symmetry (space group $R\bar{3}$) of the fluorite structure with ordered oxygen vacancies along the $\langle 1\,1\,1 \rangle$ direction. One-seventh of all cation sites are in a 6-fold coordination by oxygen on a site of trigonal symmetry. The remaining six-sevenths of the cations are surrounded by seven oxygen atoms. The δ-phase materials have been shown to exhibit astonishing amorphization resistance characteristics due

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to their natural tendency to accommodate lattice disorder. In previous studies, no “4:3:12” δ-phase compounds have exhibited irradiation-induced amorphization. However, δ-phase compounds do undergo an order-to-disorder (O-D) phase transformation from an ordered rhombohedral to a disordered cubic fluorite structure. In particular, δ-$Sc_4Zr_3O_{12}$ does undergo O-D transformation and no amorphization has been observed even at very high radiation dose (~70 dpa) [16].

In this study, we choose to examine the ion induced radiation response of another δ-phase compound. The compound we chose to examine is $Sc_4Hf_3O_{12}$ which possesses a different cation arrangement from δ-$Sc_4Zr_3O_{12}$ which has been the focus of many studies [16–20]. In $Sc_4Hf_3O_{12}$ the cations are partially ordered compared to the complete disordering in $Sc_4Zr_3O_{12}$ [21]. The purpose of the study presented here is to investigate radiation damage effects and ion species spectrum effect in δ-$Sc_4Hf_3O_{12}$ by irradiating this compound with light and heavy ions. In another aspect, we will elucidate how cation ordering level affects the radiation damage behavior in δ-phase compounds. Finally, we discuss the effect of irradiation-induced cation antisite and anion oxygen Frenkel-pair defects on the O-D transformation in δ-$Sc_4Hf_3O_{12}$.

2. Experimental procedure

Polycrystalline pellets of the δ-$Sc_4Hf_3O_{12}$ were synthesized from high purity Sc_2O_3 (Alfa Aesar, 99.99%) and HfO_2 (Aldrich Chemical company, 99.99%) powders by conventional ceramic processing procedures. The pellets were then cut and polished to a mirror finish using 0.25 μm diamond suspension. X-ray diffraction measurements revealed that the pristine oxide ceramics possess a rhombohedral symmetry, δ-phase $Sc_4Hf_3O_{12}$ structure. The measured density of the $Sc_4Hf_3O_{12}$ sample was 6.40 g/cm^3, close to ~93% of the theoretical value ($\rho_{\text{theoretical}} = 6.90$ g/cm^3).

Samples of δ-$Sc_4Hf_3O_{12}$ were irradiated with 400 keV Ne^{2+} and 600 keV Kr^{3+} ions at cryogenic temperature (~77 K) in the Ion Beam Materials Laboratory at Los Alamos National Laboratory, using a 200 kV Danfysik High Current Research Ion Implanter. The 400 keV Ne^{2+} ions were implanted at normal incidence using ion fluences ranging from 1×10^{14} to 1×10^{15} ions/cm^2 and the average ion flux was maintained at $\sim 1 \times 10^{12}$ Ne/cm^2/s during the implantations, while the heavy ion irradiation was performed with 600 keV Kr^{3+} ions to fluences ranging from 5×10^{14} to 5×10^{15} ions/cm^2, with an ion flux of $\sim 1 \times 10^{12}$ Kr/cm^2/s. The 6 MeV Xe^{26+} ion irradiation was carried out at room temperature (~300 K) to ion fluences ranging from 2×10^{13} to 1×10^{15} ions/cm^2 with an ion flux of $\sim 1 \times 10^{11}$ Xe/cm^2/s on 320 kV platform for multi-discipline research with highly charged ions at the Institute of Modern Physics, Chinese Academy of Sciences (CAS). The projected ranges of the above ion species into δ-$Sc_4Hf_3O_{12}$ were estimated using the Monte Carlo program SRIM [22] and the results are listed in Table 1. The electronic to nuclear stopping power ratio (ENSP) and displacements per atom (dpa) were also calculated using SRIM [22]. The threshold displacement energies of Sc, Hf and O were assumed to be 40 eV in the simulation (because neither the theoretical or experimental values are known). The plots showing the ENSP simulation results are shown in Fig. 1a and it shows that electronic stopping plays a greater role in the total stopping power for 400 keV Ne and 6 MeV Xe ions, while the nuclear stopping plays a greater role for 600 keV Kr ions. Fig. 1b shows the displacement damage for Ne, Kr and Xe ions at the fluence of 1×10^{15} ions/cm^2. The maximum peak displacement damage is approximately 0.33 dpa at the depth of 0.39 μm for Ne^{2+} ions, ~2.2 dpa at the depth of 0.12 μm for Kr^{3+} ions and ~4.86 dpa at the depth of 1.05 μm for Xe^{26+} ions.

The pristine and ion irradiated samples were characterized using both grazing incidence X-ray diffraction (GIXRD) and transmission electron microscopy (TEM). GIXRD measurements were made using a Bruker AXS D8 Advanced X-ray Diffractometer, with Cu-Kα radiation operating in θ–2θ geometry. The X-ray incidence angle for these measurements was chosen as $\gamma = 0.25°$, which is slightly smaller than the calculated critical angle $\alpha_c = 0.3°$ [23,24]. Under these conditions, X-rays are scattered from the surface of these samples to a depth of less than 100 nm [24,25]. This is much shallower than the 400 keV Ne, 600 keV Kr and 6 MeV Xe ion ranges shown in Fig. 1b. Therefore, we believe that the GIXRD measurements examined only the irradiated section, in the vicinity of

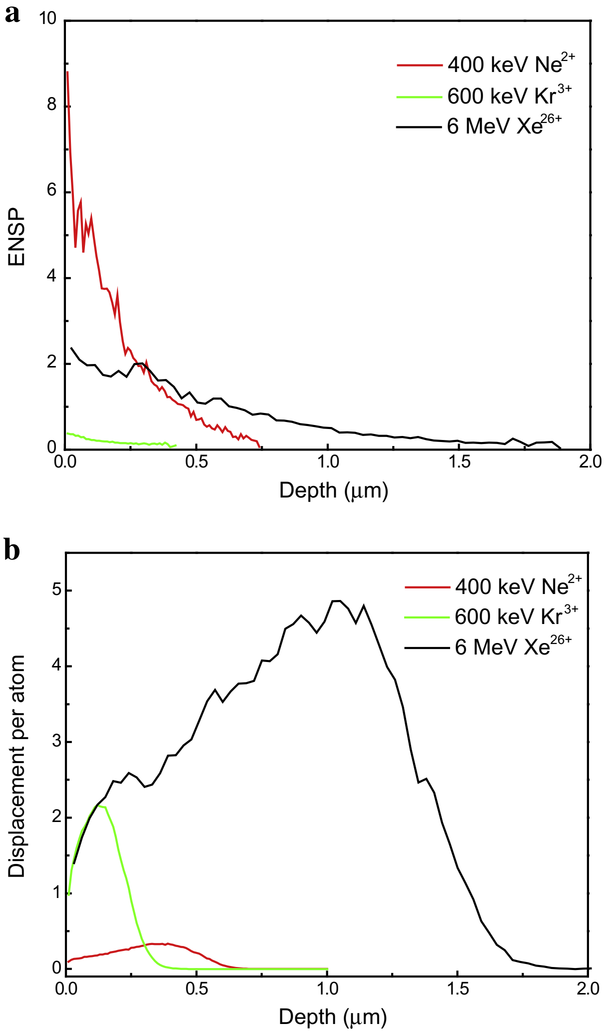

Fig. 1. (a) Electronic to nuclear stopping power ratio (ENSP) as a function of penetration depth calculated from data obtained from SRIM simulations for 400 keV Ne, 600 keV Kr and 6 MeV Xe into $Sc_4Hf_3O_{12}$. (b) SRIM calculation results plotting displacement per atom as a function of penetration depth for 400 keV Ne, 600 keV Kr and 6 MeV Xe ion irradiations into $Sc_4Hf_3O_{12}$ at a fluence of 1×10^{15} ions/cm^2. (We used the density of 6.90 g/cm^3 and the threshold displacement energy of 40 eV for Sc, Hf and O).

Table 1
SRIM projected range and longitudinal straggling, and measured O-D transformation threshold dose of three ion irradiation conditions used.

Ion beam type	400 keV Ne^{++}	600 keV Kr^{3+}	6000 keV Xe^{26+}
Projected range (μm)	0.65	0.32	1.50
Straggling (μm)	0.13	0.07	0.30
Threshold dose (dpa)	0.33	2.20	>4.86

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