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Study on effects of swift heavy ion irradiation on the crystal structure in CeO_2 doped with Gd_2O_3

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ABSTRACT

To simulate the effects of Gd_2O_3-doping and high-energy fission products in UO_2, Gd_2O_3-doped CeO_2 pellets were irradiated with 200-MeV Xe^{14+} ions. Doping and irradiation effects were analyzed using X-ray diffraction (XRD) and extended X-ray absorption fine structure (EXAFS). The lattice constant of CeO_2 decreases and the local structure is disordered with increased doping levels. However, the irradiation induces an expansion of the lattice and a disordering of atomic arrangement near the Gd atoms. The effects of the irradiation become more pronounced with increasing Gd_2O_3-dopant levels. Our results are compared with those of a study involving Er_2O_3-doped CeO_2.

1. Introduction

In current light-water nuclear power plants, to achieve a stable supply of energy and reduce the amount of spent fission fuel (UO_2), a high-burn-up extension for the fission fuel is an effective option. In high-burn-up fission fuel, however, the initial reactivity of the fuel is promoted, because of the use of enriched fission fuels. To control the initial reaction, gadolinium trioxide, which has a high neutron-absorption cross section, has been doped into UO_2 fuels as a burnable poison [1]. UO_2 fuels are exposed to irradiation from high-energy fission products (FPs) which have energies around 100 MeV. The high-energy FPs induce radiation damage in the UO_2 fuel through elastic collisions and high-density electronic excitation [2]. We have suspected that the Gd_2O_3 doping and the energetic FP irradiation affect the structure of UO_2. Therefore, it is important to study these effects on the structure of UO_2 fuels to allow for the safe operation of nuclear power plants.

In this study, CeO_2 pellets were used to simulate UO_2. CeO_2 has the same fluorite structure as that of UO_2, and has properties such as lattice constant and thermal conductivity that are similar to those of UO_2. Therefore, CeO_2 has been widely used to simulate the effects of FPs on UO_2 [3,4]. Hence, we doped CeO_2 pellets with Gd_2O_3 and irradiated them with 200-MeV Xe^{14+} ions to simulate the effects of high-energy FPs. To characterize the effects of Gd_2O_3-doping and ion irradiation, X-ray diffraction (XRD) and extended X-ray absorption fine structure (EXAFS) measurements were used. The result obtained in this study was compared those of a previous study that used Er_2O_3 as a dopant [5].

2. Experimental procedure

The specimens used in this study were Gd_2O_3-doped CeO_2 bulk pellets. CeO_2 and Gd_2O_3 (1, 5, 10 mol%) powders which were 99.9% pure were homogeneously ground and mixed for 24 h using a ball mill. Then the mixtures were compacted into pellets by uniaxial and hydrostatic pressing. The pellets were sintered at 1673 K for 12 h in air.

Gd_2O_3-doped CeO_2 pellets were irradiated at room temperature with 200-MeV Xe^{14+} ions using a high-energy ion accelerator at the Japan Atomic Energy Agency (JAEA-Tokai). The ion fluences were 1×10^{12}, 5×10^{12}, 1×10^{13}, and 2×10^{13} cm^{-2}. Some specimens were not irradiated to serve as control specimens.

The structure of the specimens was investigated using two kinds of measurements. The lattice structure of the specimens was characterized using a conventional Cu-Kα X-ray diffractometer. To study the local structure surrounding the Gd atoms, EXAFS measurements near the Gd L3-edge (7.249 keV) were performed at room temperature on beam line, BL27B, of the photon factory at the High Energy Accelerator Research Organization (KEK-PF).

3. Results and discussion

First, the effects of Gd_2O_3 doping are discussed. Fig. 1a shows the widely-scanned XRD spectra for Gd_2O_3-doped CeO_2

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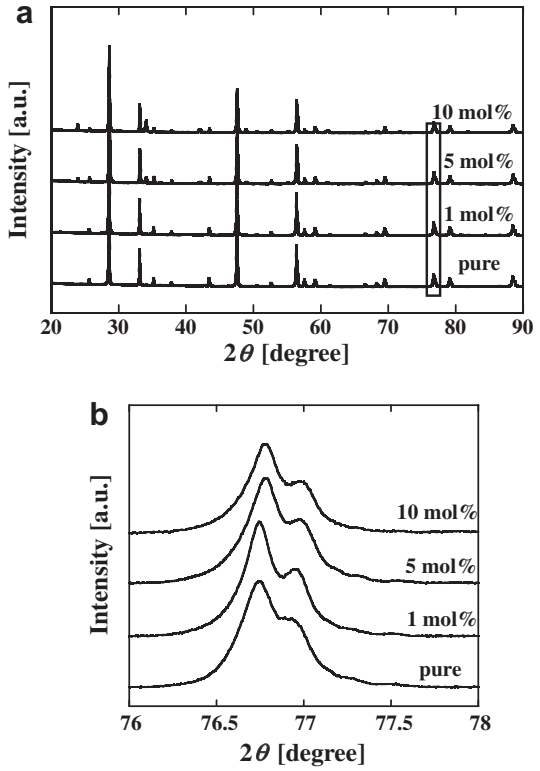

Fig. 1. XRD spectra for the Gd_2O_3-doped specimens; (a) widely-scanned spectra and (b) the (331) peaks.

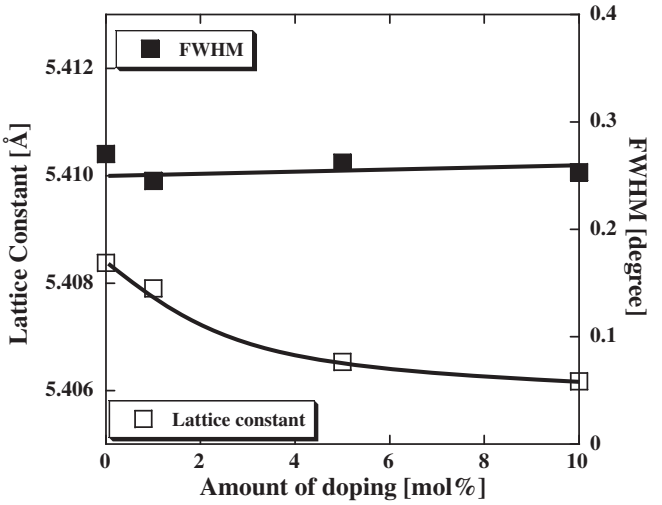

Fig. 2. Lattice constant and FWHM of the (331) peaks of the Gd_2O_3-doped specimens as a function of the amount of Gd_2O_3-dopant.

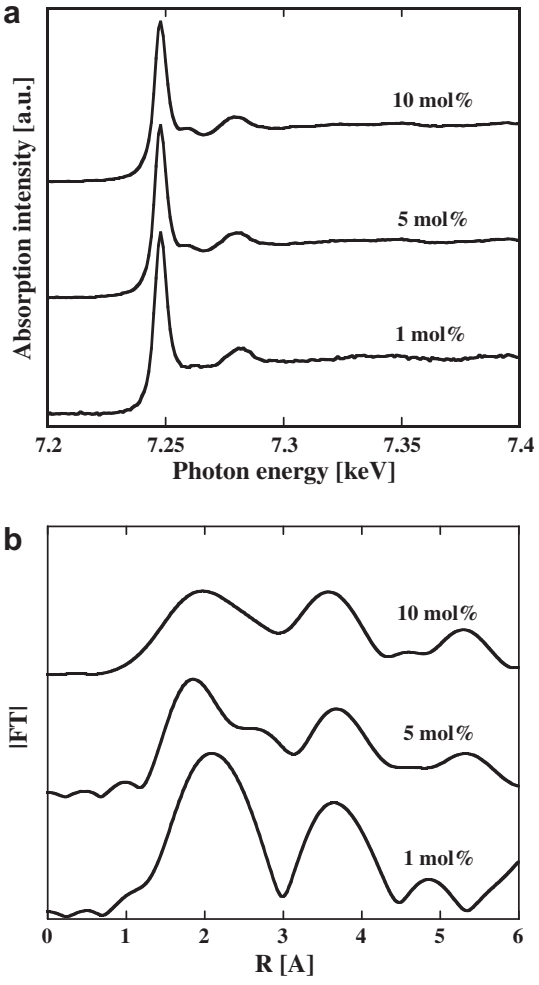

Fig. 3. Gd L3-edge EXAFS spectra for the Gd_2O_3-doped specimens; (a) normalized spectra and (b) corresponding Fourier transforms.

and undoped CeO_2. In the figure, no new phases can be seen and the lattice structure remains unchanged after doping. This result suggests that Gd atoms occupy regular Ce lattice sites in the Ce-crystal structure in Gd_2O_3-doped CeO_2. However, when analyzing diffraction peaks at each diffraction angle, we observe a peak sift to higher angles with increasing amounts of Gd_2O_3-dopant. For example, Fig. 1b shows the shift of the (331) diffraction peaks caused by Gd_2O_3-doping. This result means the lattice constant decreases through Gd_2O_3-doping. In this figure, two peaks, which correspond to Cu-Kα_1 and Cu-Kα_2 X-rays, can be observed. Note that in the data analysis in the current experiment, the peak components of Kα_2 were subtracted out of the data and the peaks corresponding to Kα_1 were used to calculate the lattice constant and the peak width. Fig. 2 shows the lattice constant and full width at half maximum (FWHM) for Gd_2O_3-doped CeO_2 as a function of the amount of Gd_2O_3 dopant, which was calculated from the (331) peaks. The lattice constant decreases with increasing amounts of Gd_2O_3. This can be explained from the difference in atomic size between the Ce atoms and Gd atoms. In the lanthanide series, the atomic size decreases with increasing atomic number. Therefore, the atomic size of Gd is smaller than that of Ce, and the doping of Gd_2O_3 into CeO_2 causes the shrinkage of the lattice. On the other hand, the FWHM changes little after Gd_2O_3-doping.

Fig. 3a shows the normalized EXAFS spectra near the Gd L3-edge for Gd_2O_3-doped CeO_2. EXAFS oscillations above the Gd edge are clearly observed in each spectrum. Fig. 3b shows the k^3-weighted Fourier transforms (FT) corresponding to the EXAFS spectra in Fig. 3a. The first peak at 2 Å for each spectrum corresponds to the first coordination of the Gd atoms for the fluorite structure (i.e. O atoms) and the second peak at 3.5 Å for each spectrum corresponds to the second coordination of the Gd atoms for the fluorite structure (i.e. primarily Ce atoms). The intensity of the peaks decreases and their widths increase with increasing dopant levels. Ohashi et al., have reported the similar effects from Gd_2O_3-doping in CeO_2 based on EXAFS analysis [6]. This means that the local structure surrounding the Gd atoms is more disordered with increasing amounts of dopant. This result suggests that the differ-