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Study of high energy ion-irradiation effects in CeO_2 by using X-ray Photoelectron Spectroscopy

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Introduction

High-burnup extension of light-water reactor (LWR) nuclear fuel (UO_2) is a good option to reduce the total amount of spent fuel and the fuel cycle costs. In high-burnup nuclear fuel pellets, however, so-called "rim structure"[1], is produced by high energy(around 100MeV) fission products (FPs). This structure includes highly dense small-sub grains whose size is ~100nm, and small pores with an average size around 1μm. The rim structure may influence the fuel performance, including, e.g. fuel temperature and thermal conductivity. This is the matter of concern on the atomic power generation systems.

This study has been performed to clarify the effects of high energy FPs on nuclear fuels by using cerium dioxide(CeO_2) with the fluorite structure as a simulation material of UO_2 fuel. To evaluate the effects of high energy FPs, heavy ion irradiation method using energetic ion accelerators is effective. In the present experiment, to evaluate the effect of high energy ion irradiation on CeO_2, we have used X-ray Photoelectron Spectroscopy (XPS) on the beamline BL27A of the KEK photon factory.

Experimental procedure

Specimens in this study were CeO_2 bulk pellets which were prepared by sintering CeO_2 powder at 1400C. The dimension of the pellets was 8 mm in diameter and 1mm thick. The specimens were irradiated with 200 MeV Xe ions at room temperature by using a high energy ion accelerator at JAEA-Tokai. The irradiation fluences were $6x10^{12}/cm^2$, $3x10^{13}/cm^2$ and $6x10^{13}/cm^2$. X-ray photoelectron spectra (XPS) of the ion-irradiated CeO_2 pellets and unirradiated one were acquired at room temperature at the end of the station of the 27A beam line in the Photon Factory at High Energy Accelerator Research Organization (KEK-PF). The monochiromatized photon energy for the measurements was just 2200.0 eV. The energy solution of the X-rays near 2200 eV was 0.1 eV. The binding energy, EB, was normalized by Au 4f7/2 photoelectron peak (E_B=84.0 eV) from metallic gold.

Results and discussion

Figure.1 (a) shows the typical Ce 3d XPS spectrum for Ce 4+ valence state, which is obtained from an unirradiated CeO_2 pellet. It has six peaks which are produced due to the multielectron interaction[2]. Fig.1 (d) shows the reference spectrum for Ce3+ valence state[3]. Fig. 1(b and c) shows the ion-fluence variation of Ce 3d XPs spectra for CeO_2 pellet irradiated with 200 MeV Xe ions. With increasing the ion-fluence, the intensity of the peaks around 916 eV, 907 eV and 888 eV, which correspond to Ce4+ state decrease and those around 903 eV and 884 eV increase gradually. This result implies that in the irradiated samples, both Ce4+ and Ce3+ oxidation states coexist and the amount of Ce3+ state increases by the irradiation. In the fluorite structure, the appearance of Ce3+ valence state has to be accompanied by oxygen vacancies as a result of oxygen displacements from the regular sites. The amount of oxygen atom displacements cannot be explained if we only consider the effect of the elastic interaction between CeO_2 and 200 MeV Xe ions, because the value of dpa(displacement per atom) is below 0.01 even for the Xe ion fluence of $10^{14}/cm^2$. To understand the change in Ce valence and accompanying oxygen atom displacements by the irradiation, the effect of high density electronic excitation on atomic movements has to be discussed.

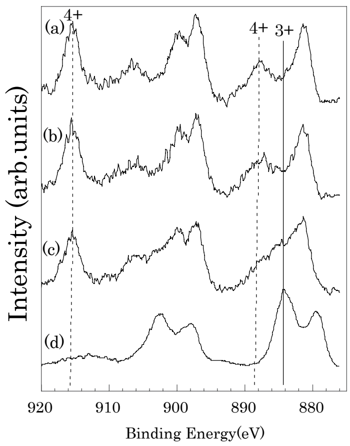

Fig. 1 : XPS spectra of Ce 3d electrons for CeO_2 unirradiated (a) and irradiated with 200MeV Xe ions of the fluence $6x10^{12}ions/cm^2$(b), $1x10^{14}ions/cm^2$(c), and for the standard specimen with the valence of Ce^{3+}(d)

References

[1] M.Kinoshita et al., in: Proceedings of the 2004 International Meeting on LWR Fuel Performance, Orlando, Florida, USA (2004), Paper 1102.
[2] Atsushi Fujimori, Phys. Rev. B, 28(1983)2281
[3] Juan P. Holgado, Rafael Alvarez, Guillermo Munuera, Applied Surface Science 161 (2000) 301

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