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Radiation damage of UO_2 implanted with 100 MeV iodine ions

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Abstract

In connection with the "rim effect", a microstructural change, of the light water reactor fuels irradiated up to high burn-ups, the radiation damage of UO_2 implanted with 100 MeV iodine ions was studied by scanning electron microscopy (SEM) and X-ray diffraction analysis. SEM observation revealed that as-fabricated pores had diminished with a melting-like morphology change at the ion-implanted surface, which can be ascribed to the large energy deposition by incident ions at the surface. The lattice parameter change for the implanted specimens increased to a maximum value of about 0.35% with increasing ion dose up to 10^{20} ions m^{-2}. Broadening of the X-ray diffraction peaks was also observed at increased doses, which could be attributed to the lattice distortion and/or decrease in the crystallite size.

1. Introduction

Uranium dioxide (UO_2) has been used as the fuel of nuclear reactors, including light water reactors (LWRs). In the course of burn-up extension programs of LWR fuel exceeding 50–70 GWd t^{-1}, attention has been directed to the "rim effect", which is the unexpected release of fission gases, Kr and Xe, from the UO_2 matrix in the rim region of the fuel pellet. The release is accompanied by a change in the microstructure of the UO_2 matrix, i.e., a loss of definable grain structure and an increase in porosity [1], and polygonization with a subgrain boundary structure [2]. These microstructural changes have been considered to be related to the radiation damage introduced by energetic fission fragments at low temperatures in the rim region [2].

Besides neutron [3] and α particle irradiation [4], the radiation damage of the nuclear fuel has been studied by ion implantation, so far mostly with relatively low energy (less than 1 MeV) ions [5]. Low energy ion implantation is effective in inducing radiation damage through the nuclear energy deposition process. In real fission events, however, much larger energy is deposited through electronic (inelastic) collisions between the UO_2 target and the incident fission fragments with energies at about the 100 MeV level.

In the present study 100 MeV iodine ions have been adopted for studying the radiation damage in the fission fragment energy region.

2. Experimental procedure

The specimens used were sintered UO_2 discs, 10.4 $mm^{\varnothing} \times$ ca. mm^{-t}, sliced from LWR fuel pellets. The O/U ratio of the as-fabricated fuel was 2.00 [6] and the density was 95% of the theoretical density (TD). The specimen discs were polished with emery paper and diamond paste of about 1 μm particle size.

The specimens were irradiated with 100 MeV iodine ions using a Tandem accelerator at the Tokai Establishment of the Japan Atomic Energy Research Institute (JAERI). The ion beam flux was $(4–9) \times 10^{15}$ ions m^{-2} s^{-1} and the fluence was 1.0×10^{18}–3.1×10^{20} ions m^{-2}. Irradiation was carried out at room temperature with a sample holder cooled by water kept at 20 °C. The temperature of the specimens was measured by a chromel–alumel (CA) thermocouple in contact with the specimen surface under ion beam incidence. The measured temperature was raised to 270 °C at maximum by ion beam heating.

Thus-irradiated specimens were observed by optical and scanning electron microscopy (SEM). The specimens were also analysed by an X-ray diffraction apparatus, RU-200B of Rigaku Ltd., with a Cu Kα beam. In order to limit the X-ray analysis area within the ion beam spot (5 mm in diameter), the specimens were masked with a molybdenum plate 0.2 mm thick with a 2 mm hole in the centre, instead of a lead mask as in a previous measurement [7]. The lattice parameter was obtained by a least-squares fitting of seven dif-

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fraction lines at higher diffraction angles (78°, 94°, 105°, 113°, 126°, 135° and 138° in 2θ).

3. Results and discussion

3.1. Surface observation

The ion beam incidence region could be distinguished even with the naked eye, in particular for higher dose specimens, as a white spot in the surrounding black unirradiated area. With optical microscopy, decreases in the density and size of as-fabricated pores were observed in the irradiated region.

The typical surface appearance observed by SEM is shown in Fig. 1. As-fabricated pores were seen in a certain density for the unirradiated specimen with 95% TD (Fig. 1(a)), while the pores have diminished in the irradiated specimen (Fig. 1(b)). Moreover, the irradiated surface has become smooth, which is well recognized in an enlarged view of the edges of the pores (Fig. 1(c)). It appears that the surface was subjected to melting or highly enhanced surface diffusion.

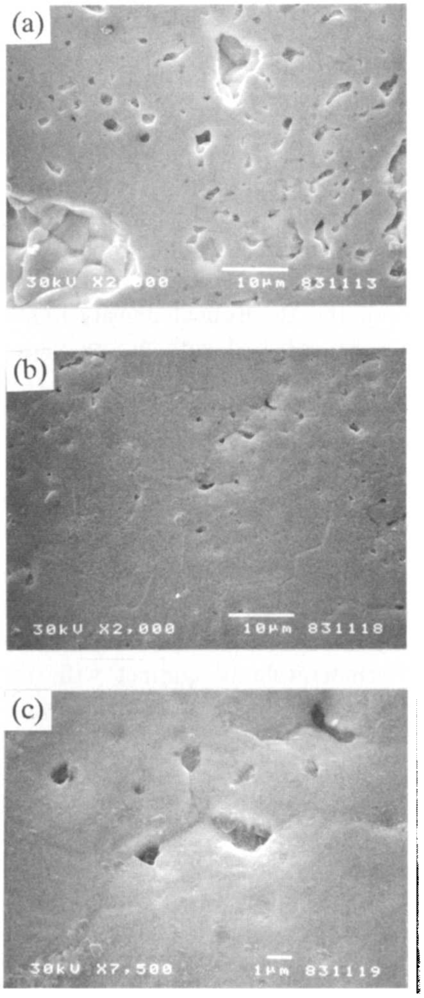

Fig. 1. Surface appearance observed by SEM: (a) unirradiated specimen with 95% TD; (b) irradiated with 100 MeV iodine ions to 9.2×10^{19} ions m^{-2} at 270 °C; (c) enlarged view of pores in (b).

The surface morphology change observed is considered to be due to the large energy deposition. In this energy region, electronic (or inelastic) energy deposition is predominant in the total energy deposition. However, the relative importance of nuclear and electronic energy depositions in inducing the surface morphology change cannot be decided by the present results only.

3.2. X-Ray diffraction

The change in the X-ray diffraction peak profile with increasing ion fluence is depicted in Fig. 2 for the (531) plane as a typical example. Compared with the profile for the unirradiated specimen, the peak positions of the irradiated specimens shifted towards lower angles with the ion fluence, indicating increases in the lattice parameter.

The lattice parameter change is plotted as a function of the ion fluence in Fig. 3. The change first increases with the fluence and then reaches a maximum value of about 0.35%. The maximum value for the present experiment is larger than the maximum value of about 0.1% derived by Nakae et al. from their neutron fission experiment [3]. A possible reason for the difference is the higher damage rate in the present experiment.

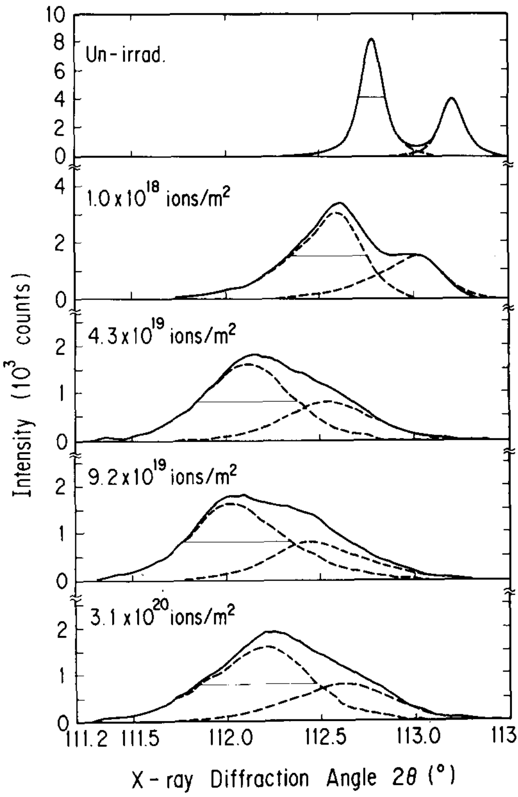

Fig. 2. X-Ray diffraction peaks for UO_2 (531) plane at various fluences of 100 MeV iodine ions.

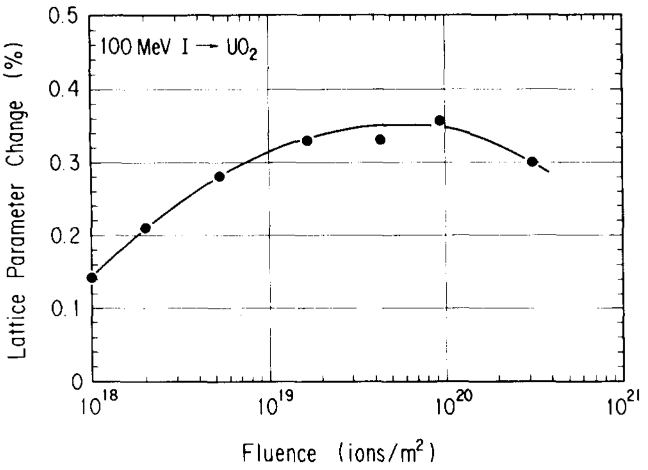

Fig. 3. Lattice parameter change of UO_2 as a function of iodine ion fluence.

Furthermore, broadening of the X-ray diffraction peaks was observed for the irradiated specimens, as shown in Fig. 2. The X-ray analysis with the 2 mm hole mask was made at three different positions within the ion beam spot, 5 mm in diameter; and similar degrees of broadening together with similar lattice parameter changes were obtained for the different positions. Therefore the broadening cannot be attributed to overlapping of different ion fluences in the X-ray-analysed area of the ion beam spot. Instead, the broadening could be ascribed to lattice distortion and/or a decrease in crystallite size of the UO_2 matrix; both of these can be induced by energetic ion beam incidence. Quantitative characterization of the broadening is in progress.

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