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Lattice Parameter Changes for Spent UO_2 Fuels With and Without Gd†

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The microstructural changes of spent nuclear fuels discharged from pressurized water reactors were investigated. The changes in the lattice parameters from the core to the rim of spent UO_2 fuel specimens were measured using a radiation-shielded micro X-ray diffraction system (μ-XRD). For comparison, two different types of spent fuel pellets of 41 GWd/tU burnup were chosen as sample specimens, one containing PWR UO_2 fuel and the other containing Gd-bearing UO_2 fuel. The Gd-bearing UO_2 fuel was chosen to investigate the effect of Gd on the lattice parameter of spent UO_2 fuel. The lattice constant from the core to the rim of the specimen was larger for the spent UO_2 fuel than for the fresh UO_2 before irradiation. Although an overall lattice expansion was observed across the radius, the lattice contracts slightly near the outer surface of the fuel pellet. Based on the position where this lattice contraction was initiated, we predicted the thickness of the 'rim' region as 100 μm. For spent Gd-bearing UO_2 fuel, the lattice across the radius of specimen is larger than that of fresh Gd-bearing UO_2 fuel but smaller than that of fresh UO_2 due to Gd dissolution in the UO_2 lattice.

Key Words: Spent nuclear fuel, UO_2, Gd-bearing UO_2, Rim, Lattice parameter.

INTRODUCTION

The microstructural changes in the outer region, called the rim, of a fuel pellet has been a great concern with the development of high burnup fuel[1-4]. The structure of UO_2 can change during the operation of the reactor as well as during cooling after discharge. The main source of the structural change is related to burnup above approximately 40 GWd/tU, which causes an increase in local burnup in the rim region of a pellet due to the neutron capture of ^{238}U. It has been reported that a rim region with a width of 100 ~ 200 μm was formed in a fuel pellet at burnup above 40 GWd/tU[5]. The notable change in the rim region of a pellet relative to the rest of the pellet is the formation of small grains and micropores. Although the rim region is only a few hundreds of micrometers thick, its volume corresponds to 4~8 % of total fuel volume and the microstructural change in rim region cause fuel to swell and lower its thermal conductivity[6,7]. When compared to fresh fuel, another obvious structural difference in spent fuel is the formation of point defects by radiation damage, which causes the crystal lattice of a UO_2 fuel to expand. The expansion of the crystal lattice caused by the accumulated point defects was observed for burnup up to approximately 70 GWd/tU[8]. At the rim region of a highly burnt fuel, contraction of the UO_2 lattice has been reported by the incorporation of fission products[9-12]. In addition, under high burnup conditions, a fuel can make contact with the cladding due to the fuel swelling, which results in the chemical interaction between the fuel and cladding. This interaction leads to the oxidation of the inner surface of the cladding, thinning of the cladding and in turn affecting the safety of fuel pins.

In this study, to access the changes in the lattice structure of a spent nuclear fuel and its cladding, the diffraction patterns were measured from the core to the rim. The lattice parameters of the spent UO_2 fuel at 41 GWd/tU burnup were obtained and compared with that of 5.98 wt % Gd-doped UO_2 fuel.

EXPERIMENTAL

Preparation of the spent fuel specimens: Spent UO_2 fuel specimens were prepared from a fuel rod discharged from the NPP(Y-2). An axial slice of the fuel pellet with a height of 3 mm and a thickness of 0.5 mm was cut along the diameter of the fuel (Fig. 1). The average burnup of the specimen was 41 GWd/tU, as measured by the Nd-148 method (ASTM, E321-96)[13,14]. A burnable poison fuel, 5.98 wt % Gd_2O_3-UO_2, was also prepared from a fuel rod discharged from the NPP(K-3). The average burnup of the Gd-bearing fuel specimen was 41 GWd/tU. The slices of the spent fuels were embedded in

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epoxy resin and then polished well at the post-irradiation examination facility.

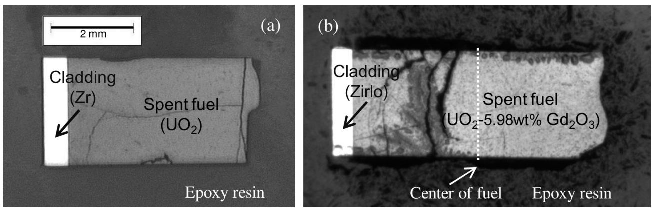

Fig. 1. Spent fuel specimens: (a) UO_2 spent fuel specimen (41 GWd/tU) discharged from NPP(Y-2) and (b) UO_2-5.98 wt % Gd_2O_3 spent fuel specimen (41 GWd/tU) discharged from NPP (K-3)

Instrumentation: A μ-XRD system (D8 Advanced, Bruker-AXS) modified with a microbeam concentrator (exit slit: ≤ 50 μm in width and 4000 μm in length) and a sample micro-positioner was used to obtain the diffraction spectra. The details of this system were fully described in our previous paper[15]. This micro X-ray diffraction (μ-XRD) system was installed inside the lead glove box for radiation safety[16]. For the accurate and precise positioning of each sample, two linear stages (travel distance: 25 mm, minimum movement: 0.1 μm) and a rotation stage (rotation angle: 360º, minimum movement: 0.004º) were equipped with a microscope (magnification at 120 mm: × 13) as shown in Fig. 2. Because the background γ-radiation signal from a spent fuel specimen interrupts the diffracted signal from a sample and results in poor detection, the sample holder[17] and the detector were shielded with 5- to 10-mm-thick tungsten sheets.

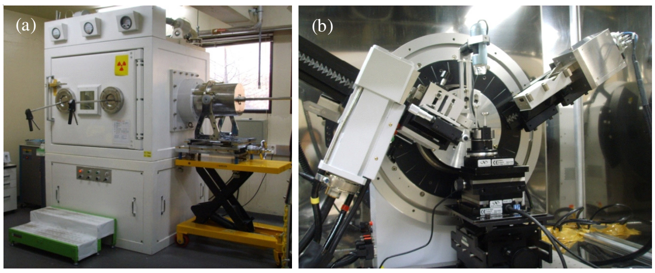

Fig. 2. Radiation-shielded micro-XRD system for the measurement of highly burnt spent fuel and a specimen cask for the transportation of a radioactive sample: (a) Radiation-shielded micro-XRD and specimen cask, (b) micro-XRD system consisting of an X-ray tube, micro-beam concentrator, microscope, shielded detector, sample holder and sample micro-moving stage

This tungsten shield lowered the background signal from the γ-radiation of a spent fuel specimen by a factor of approximately 2000, from 10^5 counts/s to 50 counts/s, as shown in Fig. 3. The measurements were carried out with a scanning step of 0.01º (2θ) for 10 s per count and using a 0.6-mm-wide detector slit. An X-ray diffraction spectrum was obtained by measuring one position of a fuel specimen for approximately 8 h. The CuK_α line filtered through Ni foil and a NaI(Tl) scintillation counter was used with a 40 mA beam current and a 40 kV beam generation power. The lattice parameters of the spent fuel samples were calculated by the TOPAS program (Pawley fitting method, BRUKER-AXS) with diffraction peaks in the range 50º-90º (2θ).

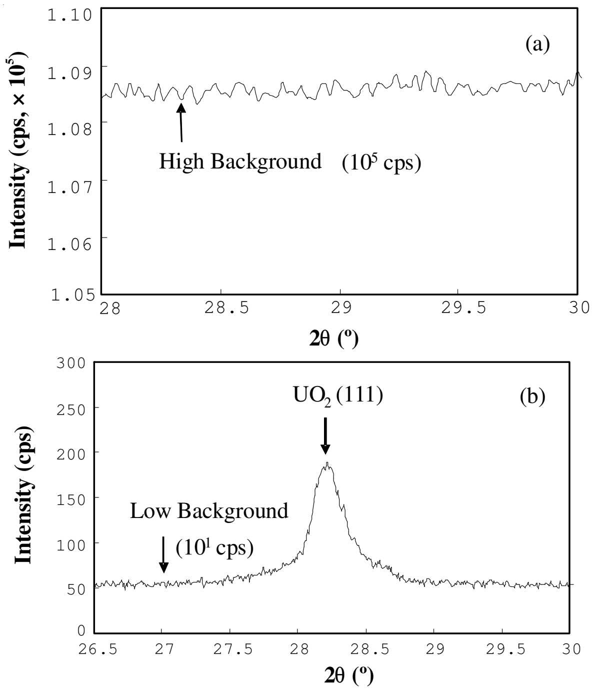

Fig. 3. Comparison of the peak intensity and background signal for UO_2 spent fuel obtained before (a) and after (b) the shield for the detector in the micro-XRD system

RESULTS AND DISCUSSION

Microstructural changes in UO_2 spent fuel specimens: UO_2 has a fluorite structure (fcc), like CaF_2 and can be described as an array of simple cubic cells of side $a_0/2$ of oxygen with alternating cube centers occupied by uranium. During the reactor operation and cooling after discharge, the lattice spacing of the UO_2 matrix of spent fuel can be affected by a number of processes. The main factors are related to the burnup and decay time of a spent fuel. In this work, the variation of lattice parameters along the radius of spent PWR fuels was investigated by μ-XRD.

The X-ray diffraction spectra were obtained at 50 μm intervals from the outer surface inwards (Fig. 4). The obtained lattice parameters of the spent UO_2 fuel were larger than that of fresh UO_2 (a = 547 pm), which seems to be caused by the radiation damage that occurred during the reactor operation and cooling after discharge. The radiation damage may cause the accumulation of point defects, which in turn expand the lattice, as mentioned by Weber[2] and Une *et al.*[3]. Weber[2] reported that the expansion of the UO_2 lattice was caused by an increase in the alpha irradiation gradient. Similar results were also reported by Une *et al.*[3], who found that the UO_2 lattice parameter increased with increasing fuel burnup, reaching saturation at a burn-up of approximately 50 GWd/tU. Beyond the burn-up limit, the supersaturated vacancies would create a porous structure.

In Fig. 4, although the contraction was not as large as reported by Spino *et al.*[8], a slight lattice contraction was observed in the rim region of the spent UO_2 fuel pellets. The maximum value was measured 100 μm from the outer surface of the fuel, whereas a decrease was observed toward the center, as shown in Fig. 4. According to Spino and Papaioannou report, the onset of the rim region was defined as the location of the maximum lattice parameter. Based on this comment, the thickness of the rim region for this sample is estimated as *ca*. 100 μm.

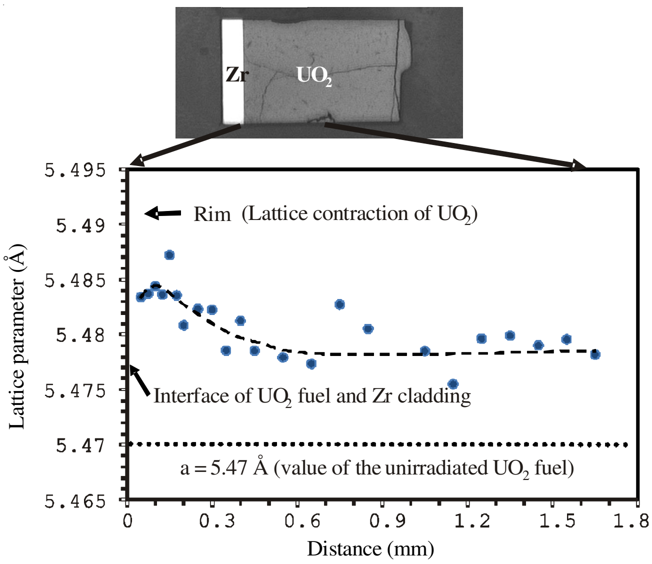

Fig. 4. Variations of the lattice parameters of spent UO_2 fuel pellets (average burn-up: 41 GWd/tU) along the radius from the fuel/cladding interface to the center

During irradiation, many fission products are generated with typical yields. Among these products, rare earth elements are dissolved as a solid solution in UO_2 matrices[8,9]. Gadolinium has often been chosen as a burnable absorber for urania-gadolinia fuels to extend the cycle length. The lattice parameters of the poisoned fuel after irradiation are shown in Fig. 5. This poisoned spent fuel (UO_2-5.98 wt % Gd_2O_3) has a burn-up of 41 GWd/tU, as measured by the Nd-148 method. As reported by other researchers[18], the lattice parameter changes linearly with increasing doped cation content depending on the ion radii, following Vegard's law. The obtained lattice parameters from the surface to the center were larger than the corresponding values for fresh UO_2-5.98 wt % Gd_2O_3 (a = 545.5 pm[11]) fuel due to radiation damage, similarly to the case of UO_2 fuel. However, the lattice parameters across the radius of the poisoned spent fuel were smaller than those of the fresh UO_2 fuel. When

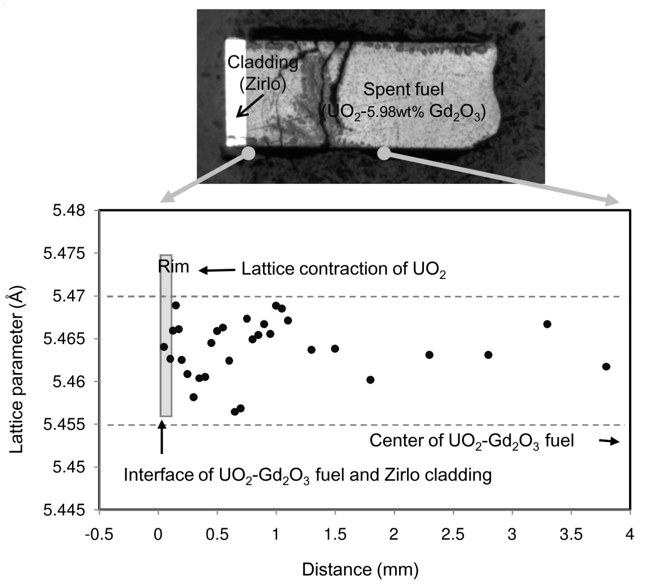

Fig. 5. Variations of the lattice parameters of spent 5.98 wt % Gd-bearing UO_2 fuel (average burn-up: 41 GWd/tU) along the radius from the surface to the center of the fuel

compared with 41 GWd/tU spent UO_2 fuel, it is very clear that this lattice contraction is caused by the formation of a solid solution with Gd that dissolved into the UO_2 lattice.

As shown in Fig. 5, the measured values were scattered between the fresh Gd-bearing fuel and fresh UO_2 fuel. Therefore, it is not possible to observe any remarkable change in the rim region of Gd-bearing fuel, but a slight lattice contraction can be observed and the rim thickness for this poisoned sample was estimated as approximately 100 μm. This lattice contraction in the rim region was attributed to the release of lattice strain by the release of the fission gas from the UO_2 matrix into the many pores and grain boundaries of the rim region[1,4,8].

Conclusion

The lattice parameter changes in nuclear fuels after irradiation (41 GWd/tU) were measured by a shielded μ-XRD. Lattice expansion after irradiation was observed for the UO_2 fuel. A small contraction in the rim region of the fuel pellet was also observed, which indicated the thickness of the rim. The burnable poison fuel (5.98 wt % Gd_2O_3-UO_2) contracted more than the fresh UO_2 fuel. It was concluded that the rim region has thickness of *ca.* 100 μm for the 41 GWd/tU spent UO_2 and poisoned fuel.

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