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# Characterization of simulated burnup fuel by nanoindentation

Ken Kurosaki *, Yoshiyuki Saito, Masayoshi Uno, Shinsuke Yamanaka

*Division of Sustainable Energy and Environmental Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan*

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**Abstract**

A simulated burnup $UO_2$ based fuel (150 GWd/t) was prepared by solid-state reactions. The phase equilibria of the simulated fuel were evaluated by XRD and SEM/EDX analysis. Nanoindentation tests were performed for the simulated fuel at room temperature in air. The modulus and hardness of the matrix phase and oxide precipitates that exit in the simulated fuel were directly evaluated by the nanoindentation.


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## 1. Introduction

An actual irradiated fuel is in extreme situations such as a high radiation field and a large temperature gradient, which leads to the complicated phase relation and microstructure. In addition, a lot of fission product (FP) elements are produced and accumulated under irradiation, and they have a great influence on the thermal and mechanical properties of an irradiated fuel. In order to evaluate the effect of FP elements on the physical properties of the fuel, a concept of simulated burnup fuel that contains non-radioactive solid-state FP elements has been proposed.

The simulated burnup fuel is a material that replicates the chemical states and phase relation of the irradiated fuel. Ordinarily, the simulated fuel is composed of the matrix phase and precipitates [1–3]. In the matrix phase, the actinide and rare earth elements are incorporated as solid solutions. The majority of Ba and Sr appears to be the oxide precipitates as a perovskite-type oxide with the chemical form of $(Ba,Sr)(U,Zr,Mo)O_3$. The metallic precipitates mainly consist of Mo, Tc, Ru, Rh, and Pd. The composition of the phases is affected by the circumstance in the fuel, especially the oxygen potential.

Previous investigations on the simulated fuel have examined the phase equilibria [1–3], the effect of the oxygen potential on the crystalline structure [4], and the thermal conductivity [5], etc. Recently, Lucuta et al. [5] and Matzke et al. [6] have studied the effect of the fission product (FP) elements on the thermophysical and thermochemical properties

* Corresponding author. Tel.: +81 6 6879 7905; fax: +81 6 6879 7889.
*E-mail address:* kurosaki@nucl.eng.osaka-u.ac.jp (K. Kurosaki).

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of the simulated fuel with equivalent simulated burnups of 3 at.% (corresponding to 28 GWd/t) and 8 at.% (corresponding to 75 GWd/t).

On the other hand, the nanoindentation technique [7–11] has been developed in some decades, and the mechanical properties within a sub-micron or nanoscale are widely discussed. The techniques are expected to be useful for measurement of the mechanical properties of thin films or local structure of various materials. However, there are few attempts of applying the nanoindentation to evaluate the mechanical properties of the nuclear fuels and materials. In our previous studies [12,13], we have performed the nanoindentation tests for $UO_2$ and some other oxide ceramics, and evaluated the nanoscale mechanical properties.

In the present study, the nanoindentation tests are performed for the simulated fuel. The nanoindentation test enables us to evaluate directly the modulus and hardness of the matrix phase and oxide precipitates that exist in the simulated fuel.

## 2. Experimental

The simulated fuel with a burnup of 150 GWd/t was prepared. The composition of the simulated fuel was determined using ORIGEN-2 code [14] as shown in Table 1. Appropriate amounts of high-purity oxides of the representative FP elements were mechanically mixed with $UO_2$ powder and calcined at 2023 K in reduction atmosphere (under $H_2 + N_2$ gas flow condition). The powder thus obtained was pressed into a pellet under a uniaxial pressure. The pellet was annealed at 1400 K for 18 h under an oxygen potential of −390 kJ/mol.

Table 1
Composition of the simulated fuel with the burnup of 150 GWd/t

| 6.2 wt%-U235 | Simulated burnup: 150 GWd/t (15.47 at.%) | |
|---|---|---|
| Representative elements | mol% | wt% |
| U | 78.90 | 88.48 |
| Ba | 1.944 | 1.257 |
| Zr | 3.227 | 1.387 |
| Mo | 3.655 | 1.652 |
| Ru | 2.688 | 1.280 |
| Rh | 0.140 | 0.068 |
| Pd | 2.108 | 1.057 |
| Y | 0.392 | 0.164 |
| La | 1.147 | 0.751 |
| Ce | 1.797 | 1.186 |
| Nd | 3.998 | 2.716 |
| Total | 100.00 | 100.00 |

The phase equilibria of the simulated fuel were examined by XRD (Cu-Kα radiation) and SEM/EDX analysis. The oxygen and metal (O/M) ratio of the sample was identified to be 2.001, which was evaluated using an oxygen analyzer HORIBA EMGA-550. The sample bulk density was evaluated to be 8.58 g/cm$^3$, which corresponds to 88% of the theoretical density (12% porosity). The average grain size of the sample was several μm.

For the nanoindentation tests, the sample was formed to a disc-shape with the diameter of 10 mm and thickness of 1.0 mm. The surface of the sample was polished with 0.3 μm-diamond powders to a mirror-like surface.

The nanoindentation tests were performed at room temperature in air using an atomic force microscope (AFM) supplied by JEOL (JSPM-4200) with a nanoindenter named TriboScope (Hysitron Inc.). From load–displacement curves obtained by the nanoindentation tests, the reduced modulus $E_r$ and nanohardness $H_n$ can be evaluated by a method suggested by Oliver and Pharr [7]. The reduced modulus $E_r$ can be combined with the modulus of a specimen and an indenter by the following relationship [7]:

$$\frac{1}{E_r} = \frac{1 - v_s^2}{E_s} + \frac{1 - v_i^2}{E_i}, \quad (1)$$

where $E$ and $v$ are the Young's modulus and Poisson's ratio, and the subscripts s and i represent the sample and indenter, respectively. Assuming that the value of $E_i = 1140$ GPa and $v_i = 0.07$ for the diamond indenter, and $v_s = 0.25$ for the sample, $E_s$ is about 15% higher than $E_r$.

In order to obtain the accurate relationship between the indentation depth and projected contact area, it is necessary to perform the tip shape calibration based on a method suggested by Oliver and Pharr [7]. In the present study, to determine the area function, a series of the indentations at the various contact depths corresponding to the load of 2500–3500 μN were performed on the standard sample (fused $SiO_2$). The load of 3000 μN was applied for the nanoindentation tests for the simulated fuel.

## 3. Results and discussion

Fig. 1(a) shows the XRD pattern of the simulated fuel for the burnup of 150 GWd/t annealed under the oxygen potential of −390 kJ/mol. There are