

The Effects of a Simulated Fission Product on the Thermal Diffusivities of (U_{0.924}Ce_{0.076})O₂ Pellet

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1. Introduction

The importance of cerium and cerium oxide is emphasized as one of the major fission products produced in a nuclear fuel under a nuclear reactor operation. Further, cerium oxide has often been used as a simulating material for plutonium oxide. Although cerium oxide cannot duplicate the behaviors of plutonium oxide exactly, it has been used owing to its similar chemical/thermodynamic behaviors, and a convenience in handling [1-3]. Especially, (U, Ce)O₂ properties data for a low content (below 10 mol%) are required in the relevant research of the MOX fuel for a pressurized water reactor (PWR).

The thermo-physical properties – thermal diffusivity, thermal conductivity, thermal expansion, specific heat, etc. – of nuclear fuel material are the most important properties to control the fuel performance in a nuclear reactor. Among these properties, the thermal diffusivity of a fuel pellet affects the thermal conductivity, and the thermal conductivity affects the fuel centerline temperature, operating power efficiency, safety, release of the fission product, etc [4-9]. The thermal diffusivity is affected by the fission product as well as the fuel material itself. In this regard, the thermal diffusivities of UO₂ and various element doped-UO₂ have been intensively studied by many investigators.

In a nuclear reactor, the fission products have several kinds of chemical state [10]. The chemical state of the numerous fission products have been classified into four groups, (a) fission products dissolved as oxides in the fuel matrix, (b) fission products forming metallic precipitates, (c) fission products forming oxide precipitates, and (d) fission gases and other volatile fission products. This classification can be applied to the case of (U, Pu)O₂ fuel as well as UO₂ fuel.

In the present work, the thermal diffusivities for the (U_{0.924}Ce_{0.076})O₂ pellet, with an addition of neodymium oxide or ruthenium as a simulated fission product, were measured using a Laser Flash Apparatus (LFA). Nd₂O₃ and Ru represent the chemical state of a fission product, a dissolved oxide and a metallic precipitate, respectively. It was intended to show the effects of the chemical states of a fission product on the thermal diffusivity of

(U_{0.924}Ce_{0.076})O₂, a simulated composition of a PWR-MOX fuel.

2. Methods and Results

2.1 Specimen preparation

(U, Ce, Nd)O₂ and (U_{0.924}Ce_{0.076})O₂+Ru sintered pellets were prepared as follows. Table 1 shows the used sample composition.

UO₂ (BNFL, IDR-UO₂), CeO₂ (Aldrich, 99.9%) and Nd₂O₃ (Aldrich, 99.99%), RuO₂ (Aldrich, 99.99%), powders were mixed with various contents using a Turbula™ mixer for 1h. Then the powder mixtures were milled using an attrition milling for 4h. Milled powder mixture was compacted with a compaction pressure of 300 MPa and sintered at 2023 K for 4h in a flowing H₂ atmosphere.

The attrition milling affected the homogenized distribution of additives in the UO₂ matrix. Therefore, the density of the samples was between 94 and 96 %TD. That is to say, it was absolutely reduced uncertainties in the density correction using 95 %TD.

Table 1. The measured lattice parameter and the calculated theoretical density of the samples used for the experiments.

Sample composition *	Lattice parameter (nm)	Theoretical density (g/cm ³ , 100%TD)
(U _{0.924} Ce _{0.076})O ₂	0.54651	10.6829
(U _{0.911} Ce _{0.075} Nd _{0.013})O ₂	0.54643	10.6149
(U _{0.874} Ce _{0.072} Nd _{0.053})O ₂	0.54618	10.4112
(U _{0.862} Ce _{0.071} Nd _{0.067})O ₂	0.54610	10.3431
(U _{0.801} Ce _{0.066} Nd _{0.133})O ₂	0.54568	10.0020
(U _{0.924} Ce _{0.076})O ₂ +2 mol%Ru	0.54651	10.6999
(U _{0.924} Ce _{0.076})O ₂ +5 mol%Ru	0.54651	10.7261
(U _{0.924} Ce _{0.076})O ₂ +7 mol%Ru	0.54651	10.7440

* It was ultimately intended to observe the effect of additive on the (U, Ce)O₂ system. So, in the ternary composition, the ratio of U/Ce was fixed to 12.1091 (from (U_{0.924}Ce_{0.076})O₂).

2.2 Thermal diffusivities measurement using laser flash method

Samples for the thermal diffusivity measurement were cut to 0.9-1.1 mm in thickness, 6 mm in diameter from a sintered pellet and polished. In the temperature range between room temperature and 1673 K, the thermal diffusivity was measured using a Laser Flash Apparatus (Netzsch, LFA-427) in a vacuum.

The thermal diffusivity data of (U, Ce, Nd)O₂ are shown in Figure 1. It shows that the thermal diffusivities gradually decreased with increasing Nd contents. It is thought that the Nd content is primarily responsible for the thermal diffusivity of (U_{0.924}Ce_{0.076})O₂, because the O/M ratio of these samples is near-stoichiometric state. The increasing content of dissolved Nd affects the phonon-lattice defect interaction. That is to say, it can be considered that the lattice anharmonicity increased with the increasing mass difference between the additive (Nd) and the host (U) atoms.

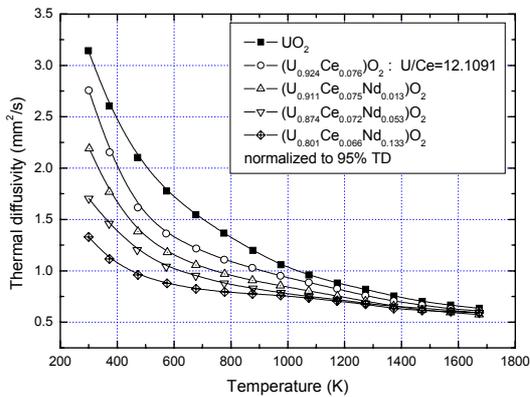


Figure 1. Thermal diffusivities of (U, Ce, Nd)O₂ samples with a various Nd content (Dissolved oxide).

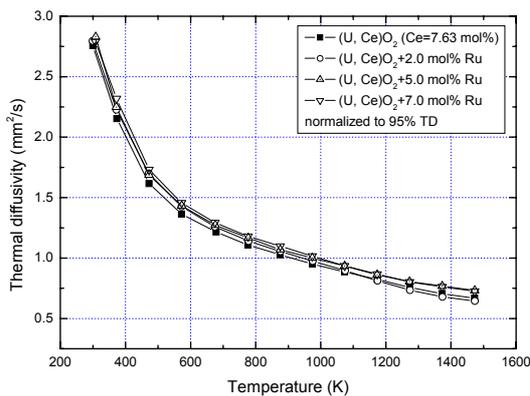


Figure 2. Thermal diffusivities of (U_{0.924}Ce_{0.076})O₂+Ru samples with a various Ru content (Metallic precipitate).

Figure 2 shows the results of a measured thermal diffusivity for (U_{0.924}Ce_{0.076})O₂+Ru sintered pellet. The

measured thermal diffusivities slightly increased with increasing Ru content. It can be thought that metallic component in the matrix is responsible for the thermal diffusivity. Generally, thermal diffusivity of metallic materials is much larger than that of ceramic materials. On the other hand, the effect of metallic precipitate is not very large. It is supposed that an isolated metallic precipitate act as an obstacle for lattice wave, while the free electron in the metallic component is increased the thermal diffusivity.

3. Conclusion

The thermal diffusivities of (U, Ce, Nd)O₂ and (U_{0.924}Ce_{0.076})O₂+Ru sintered pellets were measured by using Laser Flash Method, and the 95%TD-normalized data were compared. As results, a simulated FP forming a dissolved oxide gradually affected to decrease the thermal diffusivity, while a simulated FP forming a metallic precipitate was slightly increased the thermal diffusivity.

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