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Thermal Diffusivity of $(U,Er)O_2$ Solid Solutions As a Function of $ErO_{1.5}$ Content

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1. Introduction

Thermal conductivity is one of the most important properties of nuclear reactor fuel pellets, as it directly influences the fuel operating temperatures, and the fuel operating temperature directly affects the fuel performance and behaviors such as fission gas release and swelling. A number of studies related to the effects of the addition of burnable poison such as $GdO_{1.5}$ to the thermal conductivity of UO_2 have been published [1]. However, there is no report on the thermal conductivity for an Er-doped UO_2 pellet.

In this work, thermal diffusivities of near-stoichiometric $(U_{1-y}Er_y)O_2$ solid solutions, $0 \le y \le 0.1$, were determined from room temperature to 1673K.

2. Methods and Results

The UO_2 powder was mixed with weighed amounts of $ErO_{1.5}$ powder, at concentrations of 1, 3, 5, 7, 9, 10, 20mol%, by a Turbula® mixer for 1 hour and then successively milled by a dynamic ball mill for 1 ~ 6 hours to prepare specimens of a similar density. The green pellet specimens were sintered at 2023K in flowing H_2 for 6 hours. The X-ray diffraction patterns were recorded in the range of $20° < 2\theta < 120°$ by using a monochromatic Cu-Kα radiation on an X-ray diffractometer(MXP 3A-HF, MacScience). The lattice parameters of the $(U_{1-y}Er_y)O_2$ solid solutions, $0 \le y \le 0.2$, were calculated from all the reflections by employing the least-squares method for the Nelson-Riley extrapolation and the theoretical density of each sample was evaluated from its lattice parameter.

In the temperature range from room temperature to 1673K, the measurements of the thermal diffusivity were carried out three times at every test temperature step in a vacuum at a pressure of less than 10^{-5} Pa(Netzsch LFA-417). The thermal diffusivity (α) was calculated from the following relationship.

$$\alpha = \frac{W}{\pi}\frac{L^2}{t_{1/2}} \quad (1)$$

where $t_{1/2}$ is the time in seconds to one-half of the maximum temperature rise at the rear surface of the sample and L is the sample thickness in mm. W is a dimensionless parameter which is a function of the relative heat loss from the sample during the measurement.

2.1. Lattice Parameter

Table 1 shows the disc thickness, lattice parameter, bulk density and relative density of each sample.

Table 1. Sample characteristics of the UO_2 and $(U,Er)O_2$ pellets.

$ErO_{1.5}$ content (mol%)	Thickness (mm)	Lattice parameter (nm)	Sintered density (g/cm^3)	Relative density (%T.D.)
0	1.104	0.5470	10.66	97.3
1	0.996	0.5468	10.52	96.2
3	1.041	0.5465	10.40	95.4
5	1.058	0.5461	10.41	95.8
7	1.092	0.5455	10.40	95.8
10	1.094	0.5444	10.44	96.5
20	1.000	0.5419	10.39	97.3

Fukushima et al. [1] measured the lattice parameter and O/M ratios of $(U_{1-y}Nd_y)O_2$, $(U_{1-y}Sm_y)O_2$, $(U_{1-y}Eu_y)O_2$ and $(U_{1-y}Y_y)O_2$ solid solutions, containing up to about 15mol% of rare earth elements, sintered at 1973K in an Ar-8%H_2 mixture for 3h. Their results showed that the O/M ratio of all the samples were very close to 2.000, in the range of 1.995 to 2.003, depending on the rare earth or yttrium content.

According to Tagawa and Fujino [2], hypostoichiometric $U_{1-y}LaO_{2-x}$ has been reported to oxidize easily in air, even at room temperature, to a near-stoichiometric composition.

Although the O/M ratio of the samples was not measured chemically in this study, the deviation from the stoichiometry is assumed to be very small up to 10 mol%$ErO_{1.5}$ based on the near stoichiometric behavior of other substitutional impurities as discussed above. Therefore, we will indicate the chemical formulae of the Er-doped UO_2 solid solutions as approximately $(U_{1-y}Er_y)O_2$. The thermal diffusivity was measured for UO_2 with up to 10 mol% $ErO_{1.5}$ in this study.

Figure 1 shows the variation of the lattice parameter of the $(U_{1-y}Er_y)O_2$ solid solutions as a function of the Er content. The lattice parameter of the $(U_{1-y}Er_y)O_2$ linearly decreases as a function of the Er content and follows Vegard's law, indicating the formation of a complete solid solution between the UO_2 and $ErO_{1.5}$ phases. A regression was performed on the measured lattice parameters of $(U,Er)O_2$ to express the variation of the lattice parameters(L.P.) as a linear equation. It can be expressed as :

$$L.P. = 0.5471 - 0.0264y \ (0 \le y \le 0.2) \quad (2)$$

where y denotes the Er content.

2.2. Thermal Diffusivity

Figure 2 shows the thermal diffusivities of the $(U,Er)O_2$ solid solutions as a function of the temperature. The data of all the samples were normalized to 95% of the theoretical density by using the following equation[3].

$$\alpha_{95} = \frac{\alpha_M [(1-0.05\eta)(1-P)]}{[1-\eta P)(1-0.05)]} \quad (3)$$

where α_M, η and P are, respectively, the measured thermal diffusivity, the experimentally determined fit parameter including its temperature dependence and the porosity of the sample. Here, for the value of η, which is a function of T, the following equation suggested by Brandt and Neuer [4] was used :

$\eta = 2.6\text{-}5 \times 10^{-4}$ (T-273.15) (4)

where T is the temperature in Kelvin.

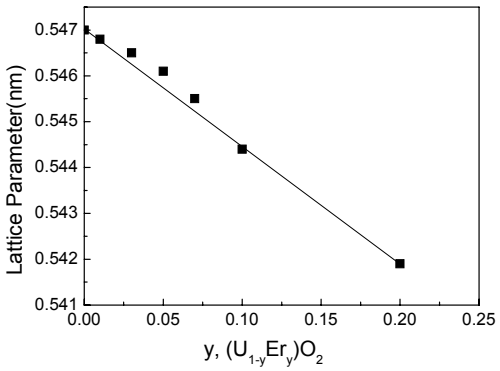

Figure 1. Lattice parameters of the $(U_{1-y}Er_y)O_2$ solid solutions as a function of the $ErO_{1.5}$ content.

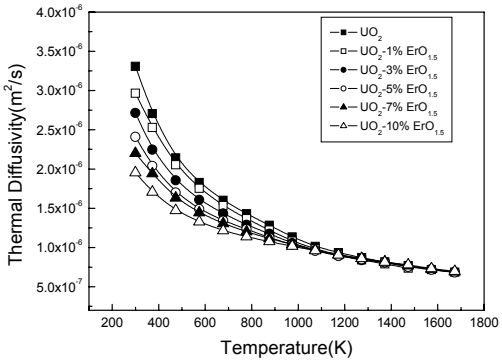

Figure 2. Variation of the thermal diffusivity of the $(U,Er)O_2$ solid solutions with different $ErO_{1.5}$ content as a function of the temperature.

The thermal diffusivities of UO_2 and $(U,Er)O_2$ gradually decreased with the test temperature as shown in Figure 2. Figure 2 also shows the dependence of the thermal diffusivity as a function of $ErO_{1.5}$ content. The thermal diffusivity of $(U,Er)O_2$ decreased with an increase of the $ErO_{1.5}$ content at low temperatures while it was independent of the $ErO_{1.5}$ content at higher temperatures, above approximately 1473K. These phenomena were also observed by Yang et al. [5] and Hirai and Ishimoto [3]. According to Yang et al. [5], the thermal diffusivities of $UO_{2.14}$ and $(U_{1-y}Gd_y)O_{2.14}$ with y = 0.09 and 0.17, were nearly the same above 1473K. In the case of Hirai and Ishimoto [3], they measured the thermal diffusivity of $(U_{1-y}Gd_y)O_2$ with y = 0.04, 0.07, 0.1, 0.15, and the aforementioned trend was observed above 1700K. However, Fukushima et al. [4] found that the thermal diffusivity curves of the UO_2 and $(U,R)O_2$ solid solutions, where R is Gd, Nd, Sm, Eu or Y, exhibited a dependence on R up to 2000K.

3. Summary

Thermal diffusivities of UO_2 and $(U, Er)O_2$ solid solutions were measured from room temperature to 1673K by a laser flash method. The thermal diffusivities of each sample decreased with increasing the temperature. The thermal diffusivity of $(U,Er)O_2$ decreased with an increase of the $ErO_{1.5}$ content at low temperatures while it was independent of the $ErO_{1.5}$ content at higher temperatures, above approximately 1473K.

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