Thermal Diffusivity Measurements for UO_2 and (U, M)O_2 Pellet (M=Ce and/or Nd)

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1. Introduction

The thermal conductivity of nuclear fuel materials is the most important property to evaluate the fuel performance in a nuclear reactor. This property affects the fuel centerline temperature, operating power efficiency, safety, and release of the fission product. In this regard, the thermal conductivities of UO_2 and various doped-UO_2 have been intensively studied by many investigators [1-3].

The importance of cerium and cerium oxide is emphasized as one of the major fission products produced in a nuclear fuel. Further, cerium oxide has often been used as a simulating material for plutonium oxide [4-6].

In the present work, the thermal diffusivities were measured using a laser flash method as a function of the Ce and/or Nd contents. Neodymium is one of the major fission products, as well. And neodymium oxide forms a solid solution in the uranium dioxide matrix, like cerium oxide. For the comparison of the measured data, the density of samples was normalized to 95 %TD (Theoretical density), which was obtained by the calculation using the lattice parameters from XRD (X-ray diffractometry). Finally, the thermal diffusivities of the UO_2 and (U, M)O_2 (M=Ce and/or Nd) were compared.

2. Methods and Results

2.1 Specimen preparation

UO_2+x mol%CeO_2 (x=0, 7.63, 14.84, 21.68 and 28.17) and UO_2+5 wt%CeO_2+y mol%Nd_2O_3 (y=0, 1, 3.94, 5 and 10) sintered pellets were prepared as follows.

UO_2 (BNFL, IDR-UO_2), CeO_2 (Aldrich, 99.9%) and Nd_2O_3 (Aldrich, 99.99%) powders were mixed with various Ce and/or Nd contents using a Turbula^{TM} 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 H_2 atmosphere.

The attrition milling affected the homogenized distribution of additives in the UO_2 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.

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.

To the compositional comparison, whole data were normalized to 95 %TD. The theoretical densities of each composition are shown in Table 1, which the TD was calculated using the measured lattice parameter. The lattice parameter was measured using XRD (Mac Science, MAC-M03XHF), and calculated by using a Nelson-Riley method from XPRESS^{TM} program (X-ray powder research software).

Table 1. The calculated theoretical density from the measured lattice parameter.

Sample	Ce or Nd content (x, y, mol%)	Theoretical density (g/cm^3, 100%TD)
UO_2 +x mol%CeO_2	0	10.96005
	7.63	10.68293
	14.84	10.41944
	21.68	10.16860
	28.17	9.92952
UO_2+5wt%CeO_2 +y mol%Nd_2O_3	1	10.61489
	3.94	10.41525
	5	10.34314
	10	10.00202

The thermal diffusivity data of UO_2+x mol%CeO_2 (x=0, 7.63, 14.84, 21.68 and 28.17) are shown in Figure 1. It shows that the thermal diffusivities gradually decreased with increasing Ce contents. It is thought that the Ce content is primarily responsible for the thermal diffusivity of (U, Ce)O_2, because the O/M ratio of these samples is near-stoichiometric state. That is to say, it can be concluded that the dissolved Ce atoms substituting U atom in U sub-lattice act as the point defect, which interrupt the transport of heat energy. At last, the thermal diffusivity decreases. The increasing content of dissolved CeO_2 affects the phonon-lattice defect interaction. Also,

the content of the dissolved CeO_2 affects the phonon-phonon interaction. It can be considered that the lattice anharmonicity increased with the increasing mass difference between the additive (Ce) and the host (U) atoms, and this anharmonicity on the lattice vibration affected phonon-phonon scattering [7].

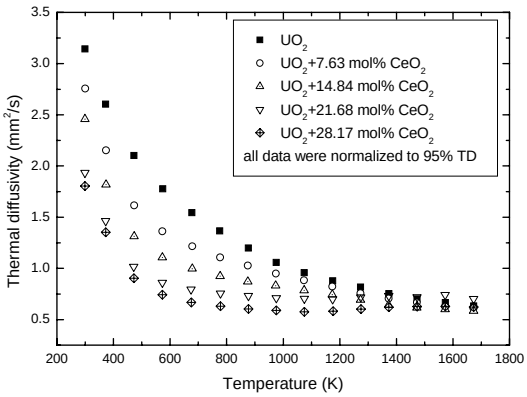

Figure 1. Thermal diffusivities of the UO_2+x mol%CeO_3 samples with the Ce content: (a) x=0, (b) 7.63, (c) 14.84, (d) 21.68 and (e) 28.17.

It is also shown that the thermal diffusivity of samples in higher Ce content decreased with increasing temperature above ~1000 K, however, slightly increased in the higher temperature region. Similar result was published by K. Kurosaki [8].

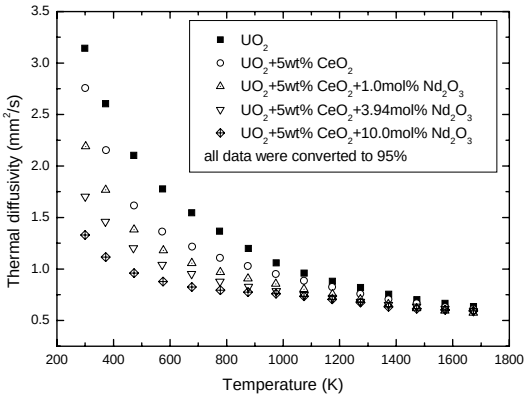

Figure 2. The measured lattice parameters of (U, Ce)O_2 and (U, Ce)O_2+Nd_2O_3 as a fuction of the Ce and Nd contents.

It can be supposed that the electron contribution caused by the higher Ce content affects the thermal diffusivity increase at higher temperature, because the concentrations of the electron are increased by the existence of trivalent Ce ions substituted for U ions [9]. And these electrons act as a carrier for heat conduction.

Figure 2 shows the results of a measured thermal diffusivity for UO_2+5wt%CeO_2+y mol%Nd_2O_3 sintered pellet (y=0, 1, 3.94, 5 and 10). Because, in the case of this experimental composition range, neodymium does also form a solid solution in the uranium dioxide matrix, like as cerium, it has a similar tendency with the results of UO_2+x mol%CeO_2. That is to say, the thermal diffusivities decreased with increasing Nd contents.

However, the influence of Nd content for the thermal diffusivity is larger than Ce content. It can be supposed that the effect of Nd content on the electron contribution is larger than by Ce, because of the increasing probability for the existence of trivalent ions.

3. Conclusion

The thermal diffusivities of UO_2 and (U, M)O_2 sintered pellets (M=Ce and/or Nd) were measured by using Laser Flash Method, and the 95%TD-normalized data were compared. As results, the cerium oxide and neodymium oxide formed solid solution, and it affected to decrease the thermal diffusivity. And the effect of Nd content is larger than Ce.

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