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Thermal conductivity of a simulated fuel with dissolved fission products

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1. Introduction

The thermal properties of a nuclear fuel should be known to assess the behavior of the fuel elements at a high temperature in a reactor. The thermal properties of a DUPIC fuel are expected to be different from a CANDU fuel because of the fission product. This causes adverse effects on the in-reactor fuel behaviors such as the thermal conductivity, thermal expansion, creep, fission gas release and the swelling of the pellets. The thermal conductivity of a nuclear fuel is one of the most important properties because it affects the fuel operating temperature and maximum power of the nuclear power plant. The thermal conductivity, k, of irradiated UO_2 depends on the deviation from a stoichiometry, x, the burnup, b, and the fractional porosity, p, as well as the temperature, T:

$$k = k(x, b, p, T), \quad (1)$$

Changes in the thermal conductivity occur during an irradiation because of a fission-gas bubble formation, pores, cracks, fission product build-up and possible changes in the oxygen to uranium ratio (O/U). Each 1 at% burnup increase corresponds to a decrease in the thermal conductivity of about 6-9 % at low temperatures (300 K) and 1-2 % at high temperatures (1770 K), respectively. However, it is difficult to distinguish between the effects of a solid precipitated fission product and the effects of a dissolved fission product on the conductivity because they coexist in a simulated fuel. The dissolved fission products in UO_2 fuel reduce the thermal conductivity, and thus the precipitated fission products increase it.

In this paper, the thermal diffusivity of a simulated fuel with fission products forming a solid solution with UO_2 has been measured using the laser flash apparatus in the temperature range from room temperature to 1673 K in order to investigate the effects of the dissolved fission products in UO_2 on the thermal diffusivity.

2. Experimental

Simulated spent fuel pellets with an equivalent burnup of 6 at% were used in this study. The specimens were fabricated by a compaction and sintering of the powder prepared by adding stable oxides as surrogates for the dissolved fission products into UO_2.

To prepare a simulated fuel, the mixed powder of UO_2 and the additives were pressed with 300 MN/m^2 into green pellets, and sintered at 1973 K for 4 hours in a flowing 100 % H_2 gas stream. The density and the grain size of the simulated fuel with the solid solutions used in the measurement were 10.49 g/cm^3 (96.9 % of theoretical density) and 9.5 μm, respectively.

The thermal diffusivity of the simulated fuel with the dissolved solid solution in UO_2 was measured by the laser flash method over the temperature range of 300 to 1673 K in a vacuum using a laser flash apparatus (LFA 427, Netzsch). Disk samples with a 10mm diameter and ~1mm thickness were taken from the pellets for the thermal diffusivity measurements.

3. Results

The thermal diffusivity of the simulated fuel is shown in Table 2 and Fig. 2 with that of UO_2 as a function of the temperature for the purpose of a comparison. From the figure it is observed that the thermal conductivity of the simulated fuel and UO_2 decreases progressively with a temperature increase. The thermal diffusivity of the simulated fuel with the solid solution decreases from 2.108 m^2/s at 300 K to 0.626 m^2/s at 1673 K. The effect of the additives is obvious as the results show a significant degradation of the thermal diffusivity of the simulated fuel with the dissolved fission products when compared to that of UO_2. The difference of the thermal diffusivity between the simulated fuel with the dissolved fission products and the UO_2 is large at room temperature, and it decreases as the temperature increases.

The thermal resistivity of the simulated fuel with the dissolved fission product increases linearly with the temperature up to 1673 K. This linearity indicates that the thermal conductivity can be expressed as a function of the temperature by using the following equation,

$$R = 1/k = A + BT = R_l + R_p, \quad (2)$$

where T is the absolute temperature, A and B are constants, R_l is the thermal resistivity caused by phonon-lattice defect interactions, or the lattice defect thermal resistivity, and R_p is the thermal resistivity caused by phonon-phonon interactions based on the Umklapp process, or the intrinsic lattice thermal resistivity.

The value of A and B were determined by fitting straight lines to the data. For the simulated fuel with the dissolved fission products, we found that A was 0.128 (m · K/W) and B was 0.226×10^{-3}. A is 0.053 and B is 0.22×10^{-3} for UO_2, A is 0.101 and B is 0.219×10^{-3} for the 3 at% burnup simulated fuel and A is 0.181 and B is 0.216×10^{-3} for the 8 at% burnup simulated fuel. The value of A of the simulated fuel with the dissolved fission products is higher than that of UO_2. It shows a similar tendency of an increase with the burnup of the simulated fuel. The value of B shows that the slope of the resistivity graph slightly decreases as the burnup of the simulated fuel increases. However, the value of B of the simulated fuel with the dissolved fission products is higher than that of UO_2. It is thought that since the simulated fuel used in this study has no metallic precipitates to increase the thermal conductivity, the slope of the thermal resistivity is higher than that of UO_2.

Table I. The specific heat, the density and the thermal diffusivity of the simulated fuel.

Temp. (K)	Specific heat (J/kg K)	Density (kg/m^3)	Diffusivity (m^2/s)
300	236.57	10490.8	2.108
473	277.56	10437.7	1.585
673	298.91	10373.2	1.213
873	308.4	10305.4	0.997
1073	312.62	10234.4	0.857
1273	316.03	10160.2	0.762
1473	322.3	10082.8	0.692
1673	334.59	10002.2	0.626

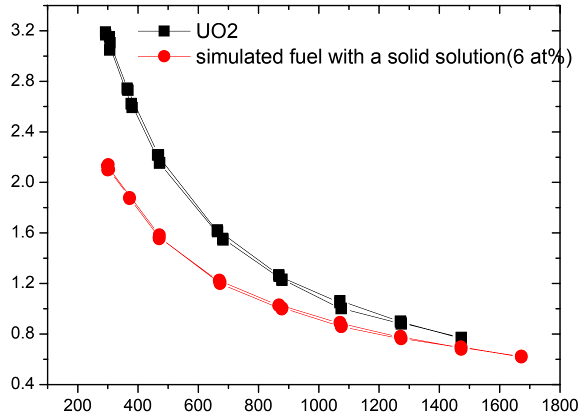

Fig.1. Thermal diffusivities as a function of the temperature for the UO_2 and the 6 at% burnup simulated fuel with the dissolved fission products.

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