

Thermal Conductivity of Simulated Fuels with Dissolved Fission Products

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Abstract The thermal diffusivity of simulated fuels with dissolved fission products was measured by using the laser-flash method in the temperature range from room temperature to 1,473 K. Three kinds of simulated fuels with an equivalent burn-up of 3, 6, and 12 at% were used in the measurement. The thermal diffusivity and the thermal conductivity of the simulated fuels with the dissolved fission products decreased, as the temperature and the equivalent burn-up increased. The thermal conductivities of simulated fuels with equivalent burn-ups of 3, 6, and 12 at% were lower than that of UO_2 by 84.70, 67.17, and 44.97% at 300 K and 99.17, 89.88, and 80.56% of UO_2 at 1,473 K, respectively. The difference in the thermal conductivity between the simulated fuel and UO_2 was large at room temperature, and it decreased as the temperature increased. The thermal resistivity of the simulated fuels increased linearly with temperature up to 1,473 K.

Keywords Laser-flash method · Simulated spent fuel · Solid solution · Thermal conductivity

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1 Introduction

The concept of the direct use of spent pressurized water reactor fuel in CANDU reactors (DUPIC) is a dry processing technology to manufacture CANDU fuel from spent PWR fuel material without separating the fissile materials and fission products in the fuel. Spent PWR fuel typically contains 0.9 mass% fissile uranium and 0.6 mass% fissile plutonium, which exceeds the natural uranium fissile content of 0.71 mass%. The neutron economy of a CANDU reactor is sufficient to allow DUPIC fuel to be used in a CANDU reactor, which was originally designed for natural uranium fuel [1,2].

The thermal properties of a nuclear fuel should be known to assess the behavior of the fuel elements at high temperature in a reactor. The main characteristic of a DUPIC fuel is its initial content of fission products as impurities. The thermal properties of a DUPIC fuel are expected to be different from a CANDU fuel because of its fission products. This causes adverse effects on the in-reactor behavior of a fuel 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 the maximum power of a nuclear power plant. The thermal conductivity, k , can be obtained from the thermal diffusivity, α , measured under transient conditions because it is very difficult to measure the thermal conductivity under steady-state conditions at high temperatures (above 1,500 K). The relation between the two properties is

$$k = \rho c_p \alpha, \quad (1)$$

where ρ is the density and c_p is the specific heat at a constant pressure.

The importance of the thermal conductivity of UO_2 fuel in determining a fuel's operating temperature has led to numerous experimental and theoretical studies. The thermal conductivity, k , of irradiated UO_2 depends on the deviation from its stoichiometry, x , its burn-up, b , its fractional porosity, p , and its temperature, T :

$$k = k(x, b, p, T), \quad (2)$$

Changes in its thermal conductivity occur during irradiation because of fission-gas bubble formation, pores, cracks, fission product build-up, and possible changes in the oxygen-to-uranium ratio (O/U). Its dependence on temperature and porosity has been studied extensively [3–5] and incorporated into computer codes used for in-pile fuel behavior analysis [6]. There are several studies on the effect of an impurity on the thermal conductivity of UO_2 [7,8]. However, little work has been done on the effect of burn-up on the thermal conductivity because of the difficulty in dealing with a highly-radioactive material. Therefore, a simulated spent fuel has usually been used to estimate the thermal diffusivity of an irradiated fuel. In the case of a DUPIC fuel, a direct measurement of its thermal properties is also very difficult in a laboratory setting due to its high level of radioactivity. As part of the DUPIC fuel development program, the thermal properties were investigated by using a simulated fuel. Simulated fuel provides a convenient way to investigate the intrinsic fuel thermal properties. There are several

studies on the thermal diffusivity of a simulated spent fuel. Lucuta et al. [9, 10] studied the thermal conductivity of a stoichiometric and a hyper-stoichiometric simulated spent fuel with equivalent burn-ups of 1.5, 3, and 8 at%. They established the effects of the fission products of a simulated fuel. They reported that the thermal conductivity of a simulated spent fuel was lower than that of UO_2 and that a small increase in the O/U ratio (2.001) resulted in a slight decrease in the thermal conductivity. Each 1 at% burn-up increase corresponds to a decrease in the thermal conductivity of about 6–9% at low temperatures (300 K) and 1–2% at high temperatures (1,770 K). The thermal resistivity (the inverse of the thermal conductivity) increased linearly with temperature and burn-up. 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 thermal conductivity because they coexist in a simulated fuel. The dissolved fission products in UO_2 fuel reduce its thermal conductivity, and the precipitated fission products increase it.

In this article, the thermal diffusivity of simulated fuels (equivalent burn-up of 3, 6, and 12 at%) with dissolved fission products in UO_2 has been measured by using a laser-flash apparatus in the temperature range from room temperature to 1,473 K in order to investigate the effects of dissolved fission products in UO_2 on the thermal diffusivity. The thermal conductivity was calculated by combining the thermal diffusivity with the specific heat and density.

2 Experimental

2.1 Specimens

Simulated spent fuel pellets with equivalent burn-up of 3, 6, and 12 at% were used in this study. The specimens were fabricated by compacting and sintering the powder prepared by adding stable oxides as surrogates for the fission products in UO_2 . The fission product composition of the irradiated fuel was determined by its initial enrichment and irradiation history. The ORIGEN (Oak Ridge Isotope Generation and Depletion) code was used to calculate the compositions of the fission products that were added to the UO_2 powder. In this study, only the fission products forming solid solutions were added to the UO_2 in order to confirm the effect of solid solutions on the thermal conductivity of a simulated fuel. The contents of the fission products added to the UO_2 powder are shown in Table 1.

To prepare a simulated fuel, the UO_2 powder and the additives were dynamically milled to obtain a homogeneous mix and the resulting product was pressed at $300 \text{ MN} \cdot \text{m}^{-2}$ into green pellets, and sintered at 2,023 K for 4 h in a flowing 100% H_2 gas stream. A complete description of the fabrication methods and characterization results were provided in a previous publication [11]. The theoretical density of the simulated fuel was calculated by assuming that the fission products added to the UO_2 formed solid solutions with UO_2 . The densities of the specimens used in the measurements were $10.291 \text{ g} \cdot \text{cm}^{-3}$ (94.5% of theoretical density) for 3 at% burn-up simulated fuel, $10.251 \text{ g} \cdot \text{cm}^{-3}$ (94.7% of theoretical density) for 6 at% burn-up simulated fuel, and $10.213 \text{ g} \cdot \text{cm}^{-3}$ (95.5% of theoretical density)

Table 1 Contents of the surrogates for the fission products added to the UO₂ powder

Fission products	3 at%	6 at%	12 at%
Sr (SrO)	0.087	0.173	0.346
Y (Y ₂ O ₃)	0.072	0.144	0.288
Zr (ZrO ₂)	0.362	0.723	1.426
La (La ₂ O ₃)	0.123	0.245	0.490
Ce (CeO ₂)	0.239	0.477	0.954
Nd (Nd ₂ O ₃)	0.408	0.816	1.632
Total	1.289	2.578	5.156

for 12 at% burn-up simulated fuel. The grain sizes are 18.4 μm for 3 at% burn-up simulated fuel, 11.5 μm for 6 at% burn-up simulated fuel, and 16.5 μm for 12 at% burn-up simulated fuel. It was also assumed that the specimens used in the experiment were stoichiometric because they were sintered in conditions of 100% H₂ at a high temperature. The microstructures of the pellets are shown in Fig. 1. The UO₂ and the simulated fuel have almost the same microstructure and grain size.

2.2 Measurement of the Thermal Diffusivity

The thermal diffusivities of the simulated fuels with the dissolved fission products in UO₂ were measured by the laser-flash method over the temperature range of 300–1,473 K in a vacuum by using a laser-flash apparatus (LFA 427, Netzsch). Disk samples with a 10 mm diameter and ~1 mm thickness were taken from the pellets for the thermal-diffusivity measurements, abraded with 600 grit silicon-carbide paper, washed in acetone in an ultrasonic cleaner, and rinsed with ethyl alcohol.

3 Results and Discussion

The thermal diffusivity of the simulated fuels is shown in Fig. 2 along with that of UO₂ [10] as a function of temperature for the purpose of comparison. From the figure, it is observed that the thermal diffusivities of the simulated fuels and UO₂ decrease progressively as the temperature increases. 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 UO₂. The difference in the thermal diffusivity between the simulated fuel with the dissolved fission products and UO₂ is large at room temperature and decreases with an increase in the temperature. This is similar to the results of the simulated spent fuel measured by Lucuta et al. [10].

The thermal conductivity of the simulated fuels with the dissolved fission products was evaluated by multiplying the thermal diffusivity by the specific heat capacity and the density. The specific heat of simulated fuels was measured by Verrall and Lucuta [12]. Their results for two kinds of simulated fuels with equivalent burn-ups of 6 and 8 at% were in close agreement with undoped UO₂. Fink [13] reviewed the

Fig. 1 Optical microscope images of (A) UO_2 and (B) the 6 at% burn-up simulated fuel with dissolved fission products ($\times 500$)

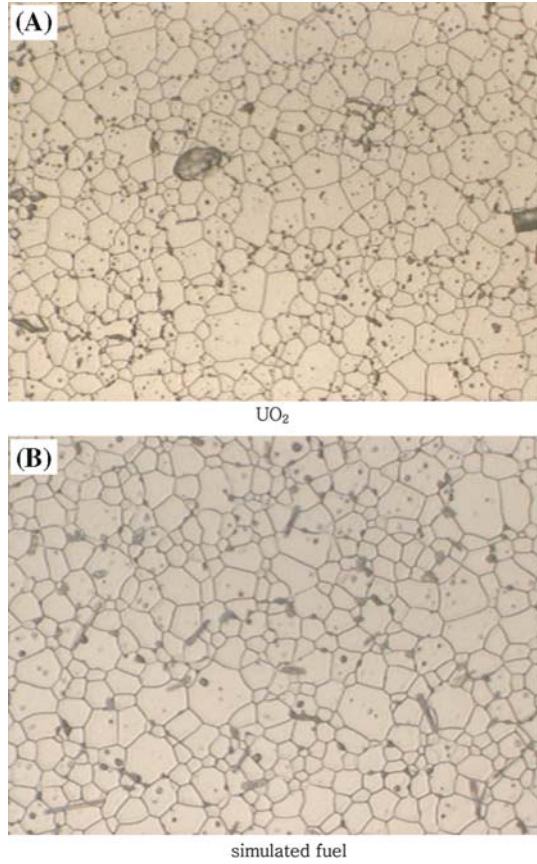


Fig. 2 Thermal diffusivity of UO_2 and the simulated fuels with dissolved fission products at equivalent burn-ups of 3, 6, and 12 at% as a function of temperature

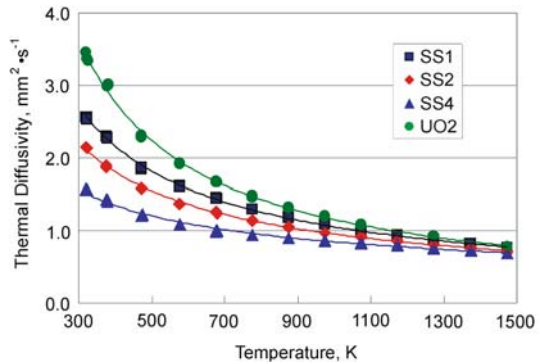
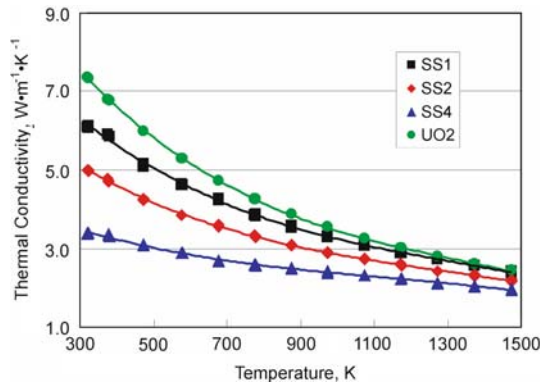


Fig. 3 Thermal conductivity of UO_2 and the simulated fuels with dissolved fission products at equivalent burn-ups of 3, 6, and 12 at% as a function of temperature



available published data on the specific heat of UO_2 and recommended a best-fit equation. The specific heat capacity recommended by Fink was used in calculating the thermal conductivity of the simulated fuel. In this study, the thermal expansion of simulated fuels with dissolved fission products was measured with a dilatometer, thereby providing the temperature-dependent densities used in the calculation of the thermal conductivity [14].

To consider the effect of porosity, we used the modified Loeb equation, i.e.,

$$k = k_{\text{TD}}(1 - \beta P), \quad (3)$$

where P is the pore volume fraction, the subscript TD refers to the theoretical density, and $\beta = 2.58 - 0.58 \times 10^{-3}T$. Therefore, the thermal conductivity normalized to 95% of TD, k_{95} is given by

$$k_{95} = k_{\text{M}}(1 - 0.05\beta)/(1 - \beta P), \quad (4)$$

where the subscripts M and 95 denote the measured value and the value corresponding to 95% of the TD, respectively.

The thermal conductivity of the simulated fuels, normalized to 95% of the TD, is shown in Table 2 for various temperatures between 300 and 1,500 K. It is plotted in Fig. 3 against temperature with the thermal conductivity of UO_2 measured by Lucuta et al. [10]. From the figure, as expected, it is observed that the thermal conductivity of the simulated fuels with dissolved fission products is lower than that of UO_2 and the difference decreases progressively with temperature. The thermal conductivity of the simulated fuels decreases progressively with the burn-up. They are 84.70, 67.17, and 44.97% that of UO_2 at 300 K and 98.53, 90.96, and 79.88% that of UO_2 at 1,473 K. Most of the difference in the thermal conductivity of the simulated fuels when compared with that of fresh UO_2 is due to the difference in the thermal diffusivity; the differences in the density and specific heat had a small effect.

It is well known that the heat in UO_2 is transferred by lattice vibrations at low temperature and by electrons at high temperature. The electronic mobility in UO_2 is too slow to provide a significant contribution at temperatures lower than about 1,800 K,

Table 2 Thermal conductivity of UO₂ and the simulated fuels with dissolved fission products at equivalent burn-ups of 3, 6, and 12 at%

Temperature (K)	Thermal Conductivity (W · m ⁻¹ · K ⁻¹)			
	UO ₂	SS1	SS2	SS4
300	7.591	6.428	5.099	3.414
400	6.581	5.635	4.583	3.212
500	5.784	5.017	4.161	3.032
600	5.140	4.521	3.811	2.871
700	4.609	4.114	3.515	2.726
800	4.166	3.775	3.262	2.595
900	3.790	3.487	3.042	2.476
1,000	3.467	3.240	2.850	2.368
1,100	3.189	3.025	2.682	2.269
1,200	2.948	2.838	2.532	2.177
1,300	2.740	2.672	2.398	2.093
1,400	2.562	2.524	2.277	2.015
1,473	2.412	2.392	2.168	1.943

Note: SS1, SS2, and SS4 designate simulated fuel with dissolved fission products at equivalent burn-ups of 3, 6, and 12 at%, respectively

and consequently, heat transfer by lattice vibrations is dominant at low temperature. However, the heat transferred by phonons decreases since the defects that cause phonon scattering increase with increasing temperature, whereas that transferred by electrons increases. The simulated fuel has a similar mechanism for heat transfer as UO₂. To observe the effect of the amount of dissolved fission products (burn-up) on the thermal conductivity of the simulated fuels, it is plotted in Fig. 4 against various burn-ups. As expected, it is observed that the thermal conductivity of the simulated fuels with dissolved fission products decreases with an increase in the burn-up. The decrease in the thermal conductivity of the simulated fuel with burn-up is due to increased scattering of the phonons by the additives. At low temperatures, the burn-up effect on the thermal conductivity is obvious; however, it decreases as the temperature increases. The thermal conductivity of the simulated fuel with dissolved fission products can be expressed as a function of the burn-up by using the following equation,

$$k = C_1 + C_2 B \quad (5)$$

where B is the burn-up, and C_1 and C_2 are constants. The values of C_1 and C_2 of the simulated fuel with dissolved fission products were determined by fitting straight lines to the data, and they are shown in Table 3.

The temperature dependence of the thermal resistivity ($R = 1/k$) of simulated fuels and UO₂ is shown in Fig. 5 as a function of temperature. The thermal resistivity of simulated fuels with the dissolved fission products increases linearly with temperature up to 1,473 K. This linear behavior is similar to the results obtained by Fukushima et al.

Fig. 4 Thermal conductivity of the simulated fuels vs. burn-ups of 3, 6, and 12 at%

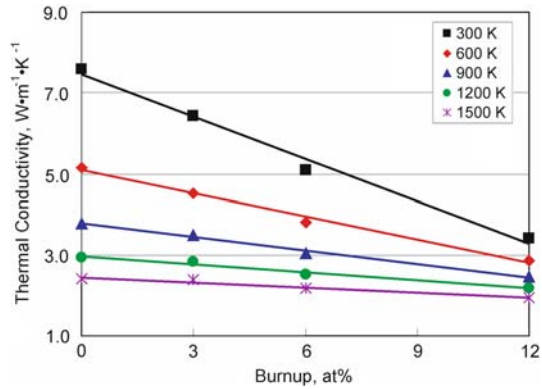


Table 3 Constants of the linear fit, $k = C_1 + C_2B$ for UO_2 and the simulated fuel with dissolved fission products at 3, 6, and 12 at% equivalent burn-ups

Temperature (K)	C_1 ($W \cdot m^{-1} \cdot K^{-1}$)	C_2 ($W \cdot m^{-1} \cdot K^{-1} \cdot at\%^{-1}$)
300	7.4628	-0.3485
600	5.0803	-0.1895
900	3.7816	-0.1110
1,200	2.9749	-0.0669
1,473	2.4492	-0.0420

Table 4 Constants of the linear fit, $R = 1/k = C_3 + C_4T$ for the simulated fuels with dissolved fission products at 3, 6, and 12 at% equivalent burn-ups

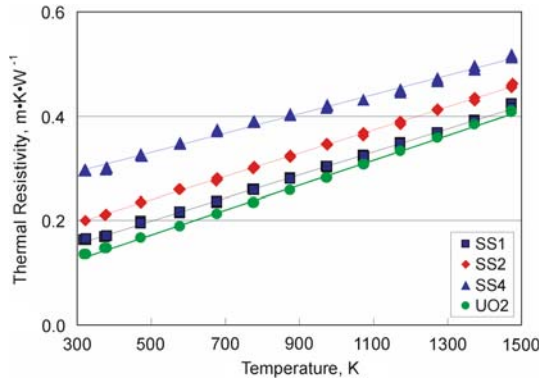
Material	C_3 ($m \cdot K \cdot W^{-1}$)	C_4 ($m \cdot W^{-1}$)
UO_2	0.054	0.218×10^{-3}
3 at% burn-up simulated fuel	0.090	0.219×10^{-3}
6 at% burn-up simulated fuel	0.130	0.221×10^{-3}
12 at% burn-up simulated fuel	0.237	0.185×10^{-3}

[8] for single additive tests and by Lucuta et al. [9, 10] for simulated high-burn-up fuel. This linearity indicates that the thermal conductivity can be expressed as a function of temperature by using the following equation,

$$R = 1/k = C_3 + C_4T = R_1 + R_p, \tag{6}$$

where T is the absolute temperature, C_3 and C_4 are constants, R_1 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 Umklapp processes, or the intrinsic lattice thermal resistivity. The values of C_3 and C_4 of the simulated fuel with dissolved fission products were determined by fitting straight lines to the data, and they are shown in Table 4.

Fig. 5 Thermal resistivity of UO_2 and the simulated fuels with dissolved fission products at 3, 6, and 12 at% equivalent burn-ups as a function of temperature



The thermal conductivity of the simulated fuels with dissolved fission products of 3, 6, and 12 at% equivalent burn-up can be expressed as a function of temperature by using the following equations:

for 3 at% equivalent burn-up simulated fuel

$$k = \frac{1}{0.090 + 0.219 \times 10^{-3}T} \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}, \tag{7}$$

for 6 at% equivalent burn-up simulated fuel

$$k = \frac{1}{0.130 + 0.221 \times 10^{-3}T} \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}, \tag{8}$$

for 12 at% equivalent burn-up simulated fuel

$$k = \frac{1}{0.237 + 0.185 \times 10^{-3}T} \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}. \tag{9}$$

4 Conclusions

The thermal diffusivities of simulated fuels with dissolved fission products in UO_2 , were measured by using the laser-flash method from room temperature to 1,473 K. Based on the work reported in this article, we can draw the following conclusions:

1. The thermal conductivities of the simulated fuels decrease as the temperature increases and as the equivalent burn-up increases.
2. The thermal conductivities of simulated fuels are lower than that of UO_2 . The differences in the thermal conductivities between simulated fuels and UO_2 are large at room temperature, and decrease as the temperature increases.
3. The thermal resistivities of the simulated fuels increase linearly with temperature up to 1,473 K.
4. The measured and calculated data in this study will be useful for a performance evaluation of the in-reactor behavior of DUPIC fuel.

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