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Thermal Expansion of a Simulated Fuel with Fission Products Forming Solid Solutions[1]

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Thermal expansions of UO_2 and a simulated fuel with fission products forming a solid solution were studied using a dilatometer in the temperature range from 298 to 1800 K. The densities of the UO_2 and the simulated fuel used in the measurements were $10.43\,\mathrm{g\cdot cm^{-3}}$ (95.2% of theoretical density (TD)) and $10.35\,\mathrm{g\cdot cm^{-3}}$ (95.6% of TD), respectively. The linear thermal expansion of the simulated fuel is higher than that of UO_2, and the difference between this fuel and UO_2 increases monotonically with temperature. The average linear thermal expansion coefficients of UO_2 and the simulated fuel are 1.09×10^{-5} and $1.23 \times 10^{-5}\,\mathrm{K^{-1}}$, respectively. As the temperature increases to 1800 K, the relative densities of UO_2 and the simulated fuel decrease to 95.1 and 94.7% of their initial densities at 298 K.

KEY WORDS: density; dilatometer; fission products; simulated fuel; thermal expansion.

1. INTRODUCTION

The concept of the direct use of spent PWR fuel in CANDU reactors (DUPIC) is a dry processing technology to manufacture CANDU fuel

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from spent PWR fuel material without separating the fissile materials and fission products in the fuel. Spent PWR fuel typically contains 0.9 wt% fissile uranium and 0.6 wt% fissile plutonium, which exceeds the natural uranium fissile content of 0.71 wt%. 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. The concept was proposed and termed the DUPIC fuel cycle by the Korea Atomic Energy Research Institute (KAERI) and Atomic Energy Canada Limited (AECL) in participation with USA in 1991 [1, 2]. The DUPIC fuel cycle offers several benefits to countries with both PWR and CANDU reactors: no need for a spent PWR fuel disposal, savings on natural uranium resources for the fabrication of CANDU fuel, and the extended burnup of CANDU fuel by utilizing the DUPIC fuel.

The main characteristic of DUPIC fuel is its initial content of fission products as impurities. The thermal properties of DUPIC fuel are expected to be different from CANDU fuel because of the fission products. The thermal properties of this fuel material should be known to assess the behavior of the fuel elements at high temperatures in reactors. The thermal expansion of nuclear fuel is one of the most important properties because it affects the gap conductance and the interaction with the cladding. It also causes a density variation with temperature, which is used for the calculation of other properties such as the thermal conductivity.

The importance of the thermal expansion of nuclear fuel has led to numerous experimental studies using high temperature X-ray and neutron diffraction techniques [3–10] and the dilatometer [11, 12]. X-ray and neutron diffraction provide lattice parameters, and the dilatometer provides macroscopic length changes including the effects of the Schottky defect. Gronvold [3], Baldock et al. [4], and Albinati [5] measured the lattice parameters of uranium oxides with temperature and found the coefficients of the linear thermal expansion. Hutchings [6] and Tyagi and Mathews [10] investigated thermal expansions by measuring the lattice parameters with the temperature of $(Th, U)O_2$. Momin et al. [7] studied the lattice thermal expansion behavior of UO_2, ThO_2, and $(U_{0.2}Th_{0.8})O_2$ doped with fission product oxides. Yamashita et al. [8, 9] studied the thermal expansions of NpO_2 and some other actinide dioxides and $(Np, U)O_2$. Tokar et al. [11] and Lorenzelli and El Sayed Ali [12] measured the thermal expansion of PuO_2 and $(U, Pu)O_{2-x}$, respectively, using a dilatometer. Martin [13] reviewed the available expansion data related to UO_2 and (U, Pu) mixed oxides and recommended fitting equations and coefficients for the thermal expansion of these materials.

In the case of DUPIC fuel, the direct measurement of the thermal properties is very difficult in a laboratory due to its high level of radioactivity. As a part of a DUPIC fuel development program, the thermal properties have been investigated using simulated DUPIC fuel. Simulated fuels provide a convenient way to investigate the intrinsic fuel thermal properties.

In this study the thermal expansions of UO_2 and a simulated fuel with fission products forming solid solutions are measured using a dilatometer in the temperature range of 298–1800 K to conform the effect of solid solutions on the thermal expansion of a simulated fuel. Also, the thermal expansion coefficient and density variations with temperature are calculated from the thermal expansion data.

2. EXPERIMENTAL

2.1. Sample Preparation and Characterization

Simulated fuel pellets with an equivalent burnup of 60 GWd/tU were used in this study. The specimens were fabricated by compaction and sintering the powder prepared by adding stable oxides as surrogates for the fission products into 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 [14] was used to calculate the compositions of the fission products, which were added into the UO_2 powder. In this study the fission products only forming solid solutions were added into UO_2 in order to confirm the effect of solid solutions on the thermal expansion of a simulated fuel. Table I shows the contents of the fission products added into the UO_2 powder.

To prepare a simulated fuel, the mixed powder of UO_2 and the additives were pressed at $300\,\mathrm{MN\cdot m^{-2}}$ into green pellets, and sintered at 1973 K for 4 h in a 100% H_2 flowing gas stream. Complete descriptions of the fabrication methods and characterization results have been provided in a previous publication [15]. The theoretical density of the simulated fuel was calculated by assuming that the fission products added to the UO_2 were fully formed solid solutions with UO_2. The densities of UO_2 and the simulated fuel were calculated to be $10.43\,\mathrm{g\cdot cm^{-3}}$ (95.2% of TD) and $10.35\,\mathrm{g\cdot cm^{-3}}$ (95.6% of TD), respectively. It is also assumed that the specimens used in the experiment are stoichiometric because they are sintered in conditions of 100% H_2 at a high temperature. The microstructures of the pellets are shown in Fig. 1. UO_2 and the simulated fuel have almost the same microstructure and grain size. The average grain sizes of UO_2 and the simulated fuel are measured to be 12.5 and 13.0 μm, respectively.

Table I. Contents of the Surrogates for the Fission Products Added to the UO_2 Powder

Fission Products	Simulated Fuel (60 GWd/tU)	Reference Simulated Fuel (35 GWd/tU) [17]	Forms in UO_2
Sr (SrO)	0.173	0.101	Solid solution
Y (Y_2O_3)	0.144	0.084	Solid solution
Zr (ZrO_2)	0.723	0.422	Solid solution
La (La_2O_3)	0.245	0.143	Solid solution
Ce (CeO_2)	0.477	0.278	Solid solution
Nd (Nd_2O_3)	0.816	0.476	Solid solution
Mo (MoO_3)	–	0.392	Metallic precipitate
Ru (RuO_2)	–	0.269	Metallic precipitate
Pd (PdO)	–	0.187	Metallic precipitate
Rh (Rh_2O_3)	–	0.052	Metallic precipitate
Ba ($BaCO_3$)	–	0.218	Oxide precipitate
Te (TeO_2)	–	0.049	Oxide precipitate
Total	2.578	2.671	

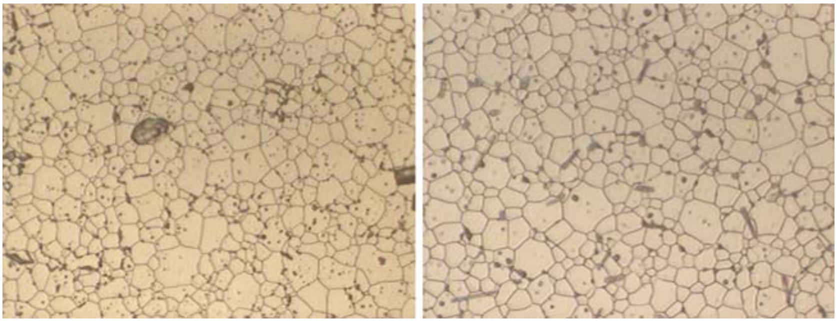
(a) UO_2 **(b)** simulated fuel

Fig. 1. Optical microscope image of (a) UO_2 and (b) simulated fuel (500×).

2.2. Measurement of the Thermal Expansion

The thermal expansions of the simulated fuel pellets were measured in the axial direction with a linear variable differential transformer (LVDT) transducer in the temperature range of 298–1800 K by a push-rod-type dilatometer (DIL 402 C, Netzsch). The measurements were carried out at a constant heating rate of $5\,K \cdot min^{-1}$ in a vacuum. The maximum error of the dilatometer used in the experiment is estimated to be within 2% for a standard material of Al_2O_3.

3. RESULTS AND DISCUSSION

The linear thermal expansion which represents the ratio of the length change to the initial length is calculated using the following expression [16]:

$$\text{Expansion, } \frac{\Delta L}{L_0}, \% = \frac{L_T - L_{298}}{L_{298}} \times 100, \tag{1}$$

where L_T and L_{298} represent the lengths of the specimens at temperature T and at 298 K, respectively. The linear thermal expansions of UO_2 and the simulated fuel determined in this study are plotted as a function of temperature in Fig. 2. The linear thermal expansion of a reference simulated fuel with an equivalent burnup of 35,000 MWd/tU reported by Kang et al. [17] is also shown in Fig. 2 for the purpose of comparison. Martin [13] reviewed the available data of UO_2 thermal expansions and recommended a third-degree polynomial equation as a function of temperature for the linear thermal expansion of UO_2. The computational result from this equation is also shown in Fig. 2.

From the figure, as expected, it is observed that the linear thermal expansions of all specimens increase monotonically with temperature. The linear thermal expansion of UO_2 obtained in this study is in good agreement with Martin's result. For the simulated fuel, it is slightly higher than that of UO_2, and the difference between this fuel and UO_2

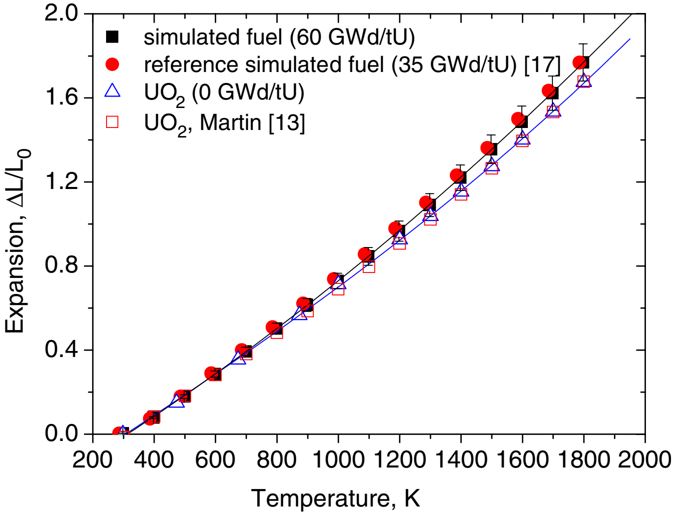

Fig. 2. Linear thermal expansions of UO_2, the simulated fuel and the reference simulated fuel as a function of temperature.

increases progressively with temperature. However, the difference between the simulated fuel and the reference simulated fuel can hardly be observed. Simulated fuel of a 60 GWd/tU burnup incorporates fission products that form solid solutions, while the reference simulated fuel has fission products that form metallic and oxide precipitates as well as solid solutions. However, they have a similar total amount of additives. The thermal expansion of the simulated fuels was influenced by total amount of additives rather than forms of those in the UO_2 matrix. The thermal expansions obtained in this study are fitted by the following equations:

For the simulated fuel with solid solution fission products,

$$\begin{aligned}\Delta L/L_0(\%) = {} & -0.2849 + 8.6831\times 10^{-4}T + 1.4094\times 10^{-7}T^2 \\ & + 6.1311\times 10^{-12}T^3 \pm 0.0028. \end{aligned} \tag{2}$$

For UO_2,

$$\begin{aligned}\Delta L/L_0(\%) = {} & -0.2885 + 9.3345\times 10^{-4}T + 1.7696\times 10^{-8}T^2 \\ & + 3.8034\times 10^{-11}T^3 \pm 0.0077. \end{aligned} \tag{3}$$

In the above equations, the last terms represent the standard deviations.

The corresponding instantaneous coefficient of the thermal expansion, α, is defined by the following expression [16]:

$$\alpha = \frac{1}{L_{298}}\frac{dL}{dT}, \tag{4}$$

The instantaneous thermal expansion coefficients of UO_2 and the simulated fuel determined in this study are given in Table II and plotted against temperature in Fig. 3.

From the figure it is observed that the instantaneous thermal expansion coefficient of the simulated fuel is slightly higher than that of UO_2. The instantaneous coefficients of the thermal expansions in the temperature range of 300–1800 K derived in this study are fitted by the following equations:

For the simulated fuel with solid solution fission products,

$$\begin{aligned}\alpha = {} & 6.714\times 10^{-6} + 9.779\times 10^{-9}T - 7.212\times 10^{-12}T^2 \\ & + 2.408\times 10^{-15}T^3 \pm 4.335\times 10^{-7}. \end{aligned} \tag{5}$$

For UO_2,

$$\begin{aligned}\alpha = {} & 4.336\times 10^{-6} + 1.305\times 10^{-8}T - 8.211\times 10^{-12}T^2 \\ & - 2.139\times 10^{-15}T^3 \pm 4.379\times 10^{-7}. \end{aligned} \tag{6}$$

Table II. Instantaneous Thermal Expansion Coefficients of UO_2 and the Simulated Fuel Determined in this Study

	$\alpha \times 10^{-5}$ (K^{-1})	
Temperature (K)	UO_2	Simulated Fuel
400	0.820	0.903
500	0.974	1.06
600	1.03	1.07
700	1.05	1.09
800	1.06	1.10
900	1.08	1.14
1000	1.12	1.15
1100	1.14	1.20
1200	1.18	1.20
1300	1.25	1.29
1400	1.33	1.38
1500	1.36	1.25
1600	1.31	1.34
1700	1.36	1.44
1800	1.45	1.49

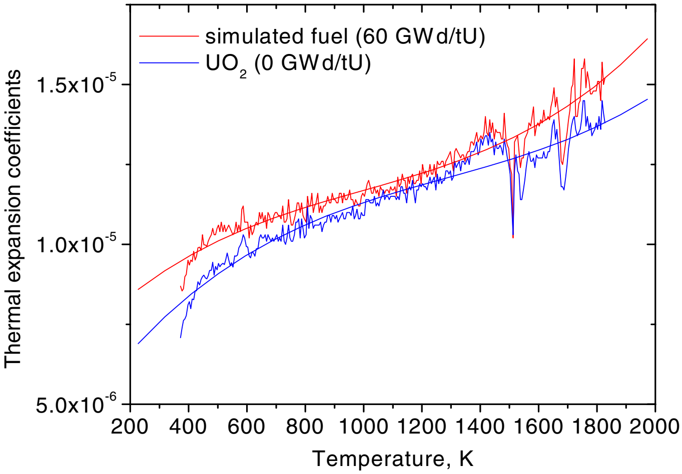

Fig. 3. Instantaneous thermal expansion coefficients of UO_2 and the simulated fuel.

In the above equations, the last terms represent the standard deviations.

The coefficient of the average linear thermal expansion, $\overline{\alpha}$, is defined by the following equation:

$$\overline{\alpha} = \frac{1}{L_{298}} \left(\frac{L_T - L_{298}}{T - 298} \right). \tag{7}$$

For the temperature range from 400 to 1800 K, the linear thermal expansion coefficients of UO_2 and the simulated fuel are 1.09×10^{-5} and $1.13 \times 10^{-5}\,K^{-1}$, respectively. These results are in agreement with those from Momin et al. [7]. Momin et al. found that the coefficients of the average linear thermal expansions for UO_2 and the solid solution of UO_2 with 20 wt% Ln_2O_3 (Ln = La, Nd, Ce, Y, Sm, Gd and Eu) are 1.08×10^{-5} and $1.43 \times 10^{-5}\,K^{-1}$, respectively. As Momin et al. [7] mentioned, the higher values for the simulated fuels indicate that the partial substitution of U^{4+} with (Y, La, Nd, and Ce)$^{3+}$ added to the simulated fuels results in a weakening of the interatomic bonding in the solid solution matrix.

The density variations with temperature can be obtained from the thermal expansion data using the following equation [18]:

$$\rho(T) = \rho(298) \left(\frac{L_{298}}{L_T} \right)^3, \tag{8}$$

where $\rho(T)$ and $\rho(298)$ represent the densities of the specimens at temperature T and at 298 K, respectively.

The relative density ($\rho(T)/\rho(298) \times 100$) variations to the initial density of the simulated fuel and UO_2 determined in this study are plotted against temperature in Fig. 4. From the figure it is observed that the relative density variations of all the specimens decrease monotonically with temperature, as expected. For UO_2, as the temperature increases to 1800 K, the relative density decreases to 95.1% of the initial density at 298 K. For the simulated fuel, the relative density is lower than that of UO_2. At a low temperature to 600 K, the difference of the relative densities of the simulated fuel and UO_2 is small and increases with temperature. As temperature increases to 1800 K, the relative density of the simulated fuel decreases to 94.7% of the initial density at 298 K.

The relative density variations with temperature calculated using Eq. (8) are fitted by the following equations:

For the simulated fuel with solid solution fission products,

$$\begin{aligned}\rho(T)/\rho(298) \times 100, \% = {} & 100.86 - 0.0026T - 3.9453 \times 10^{-7} T^2 \\ & + 3.3158 \times 10^{-12} T^3 \pm 0.0083.\end{aligned} \tag{9}$$

For UO_2 fuel,

$$\begin{aligned}\rho(T)/\rho(298) \times 100, \% = {} & 100.96 - 0.0032T + 4.3327 \times 10^{-7} T^2 \\ & - 2.6065 \times 10^{-10} T^3 \pm 0.0175.\end{aligned} \tag{10}$$

In the above equations, the last terms represent the standard deviations.

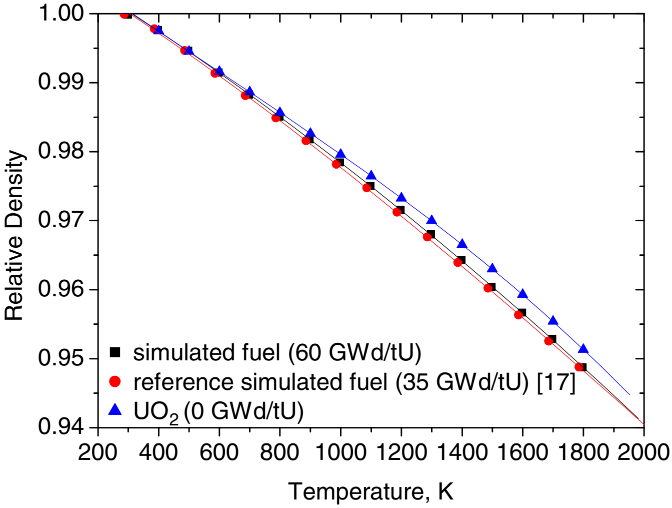

Fig. 4. Density variations of UO_2, the simulated fuel, and the reference simulated fuel as a function of temperature.

4. CONCLUSIONS

The thermal expansions of a simulated fuel and UO_2 were measured using a dilatometer over the temperature range of 298–1800 K in order to estimate the thermal expansion and density variation with temperature of DUPIC fuel, and the following results were obtained.

(1) The thermal expansion of the simulated fuel with fission products forming a solid solution is higher than that of UO_2.

(2) In the temperature range of 298–1800 K, the average linear thermal expansion coefficient of the simulated fuel is $1.13 \times 10^{-5}\,K^{-1}$.

(3) The relative density of the simulated fuel with fission products forming a solid solution is lower than that of UO_2.

(4) The thermal expansions, the average linear thermal expansion coefficients, and the density variations of the simulated fuel of a 60 GWd/tU burnup and the reference simulated fuel are similar to each other.

(5) The thermal expansion of the simulated fuels was influenced by total amount of additives rather than forms of those in the UO_2 matrix.

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