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Thermal Expansion of Simulated Fuels with Dissolved Fission Products in a UO_2 Matrix

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Abstract As a part of the DUPIC (direct use of spent PWR fuel in CANDU reactors) fuel development program, the thermal expansion of simulated spent fuel pellets with dissolved fission products has been studied by using a thermo-mechanical analyzer (TMA) in the temperature range from 298 K to 1773 K to investigate the effects of fission products forming solid solutions in a UO_2 matrix on the thermal expansions. Simulated fuels with an equivalent burn-up of (30 to 120) GWd/tU were used in this study. The linear thermal expansions of the simulated fuel pellets were higher than that of UO_2, and the difference between these fuel pellets and UO_2 increased monotonically with temperature. For the temperature range from 298 K to 1773 K, the values of the average linear thermal expansion coefficients for UO_2 and simulated fuels with an equivalent burn-up of (30, 60, and 120) GWd/tU are $1.19 \times 10^{-5}\,K^{-1}$, $1.22 \times 10^{-5}\,K^{-1}$, $1.26 \times 10^{-5}\,K^{-1}$, and $1.32 \times 10^{-5}\,K^{-1}$, respectively.

Keywords Fission products · Simulated fuel · Thermal expansion · TMA

1 Introduction

The concept of the *d*irect *u*se of spent *P*WR fuel *i*n *C*ANDU reactors (DUPIC) is a dry processing technology to manufacture CANDU fuel from spent PWR fuel material

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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. 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 spent PWR fuel disposal, savings on natural uranium resources for the fabrication of CANDU fuel, and the extended burn-up of CANDU fuel by utilizing the DUPIC fuel.

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. 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 pellet-cladding mechanical interaction. 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 [3–11] and the dilatometer [12–14]. 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. studied the thermal expansions of NpO_2 and some other actinide dioxides [8] and (Np, U)O_2 [9]. Tokar et al. [12] and Lorenzelli and El Sayed Ali [13] measured the thermal expansion of PUO_2 and (U, Pu)O_{2-x}, respectively, using a dilatometer. Kang et al. [11] measured the lattice parameter of the simulated fuel pellets using neutron diffraction and calculated the thermal expansion of them using the lattice parameter. They also measured the thermal expansion of the simulated fuel pellets with the equivalent burn-up of 60 GWd/tU using a dilatometer [14]. Martin [15] 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 a fuel's intrinsic thermal properties.

In this study, the thermal expansions of the simulated fuel pellets with fission products forming solid solutions were measured using a TMA in the temperature range of 298 K to 1773 K to investigate the effects of solid solutions on the thermal expansion of the simulated fuel pellets. Also, the thermal expansion coefficients and density variations with temperature were calculated from the thermal expansion data.

2 Experimental

2.1 Sample Preparation and Characterization

Simulated fuel pellets with an equivalent burn-up of (30, 60, and 120) 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 compositions of the irradiated fuel were determined by their initial enrichment and irradiation history. The ORIGEN (Oak Ridge Isotope Generation and Depletion) code [16] 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 investigate the effects of solid solutions on the thermal expansions of the simulated fuel pellets. 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 $300\ \mathrm{MN \cdot m^{-2}}$ into green pellets, and sintered at 2023 K for 4 h in a flowing 100 % H_2 gas stream. A complete description of the fabrication methods and characterization results was provided in a previous publication [17].

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 $10.291\ \mathrm{g \cdot cm^{-3}}$ (94.5 % of theoretical density) for 30 GWd/tU burn-up simulated fuel, $10.251\ \mathrm{g \cdot cm^{-3}}$ (94.7 % of theoretical density) for 60 GWd/tU burn-up simulated fuel, and $10.213\ \mathrm{g \cdot cm^{-3}}$ (95.5 % of theoretical density) for 120 GWd/tU burn-up simulated fuel. The grain sizes were 18.4 μm for 30 GWd/tU burn-up simulated fuel, 11.5 μm for 60 GWd/tU burn-up simulated fuel, and 16.5 μm for 120 GWd/tU burn-up simulated fuel. It was also assumed that the specimens used in the experiment were stoichiometric because they were sintered

Table 1 Contents of the surrogates for the fission products added to the UO_2 powder

Fission products	Contents (mass%)		
	30 (GWd/tU)	60 (GWd/tU)	120 (GWd/tU)
Sr (SrO)	0.087	0.173	0.346
Y (Y_2O_3)	0.072	0.144	0.288
Zr (ZrO_2)	0.362	0.723	1.426
La (La_2O_3)	0.123	0.245	0.490
Ce (CeO_2)	0.239	0.477	0.954
Nd (Nd_2O_3)	0.408	0.816	1.632
Total	1.289	2.578	5.156

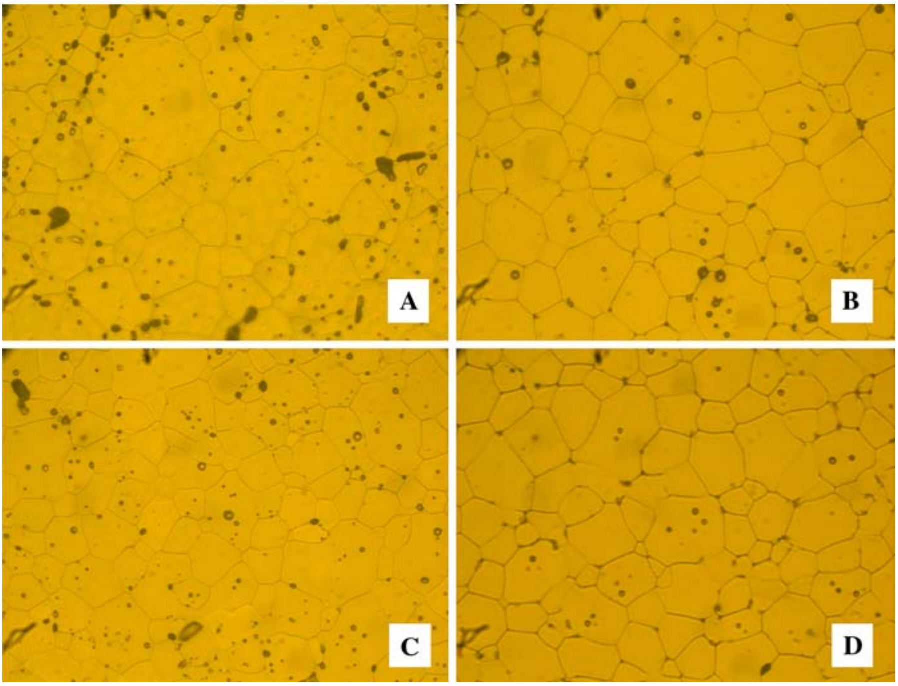

Fig. 1 Optical microscope images of (**a**) UO_2, (**b**) 30 MWd/tU, (**c**) 60 MWd/tU, and (**d**) 120 MWd/tU burn-up simulated fuel with dissolved fission products (500×)

in conditions of 100 % H_2 at 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.

2.2 Experimental Procedure

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 K to 1773 K by a push-rod-type TMA (Setaram). The measurements were carried out at a constant heating rate of $5\,\mathrm{K \cdot min^{-1}}$ in a flow of Ar. The maximum error of the TMA 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, was calculated using the following expression [18]:

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

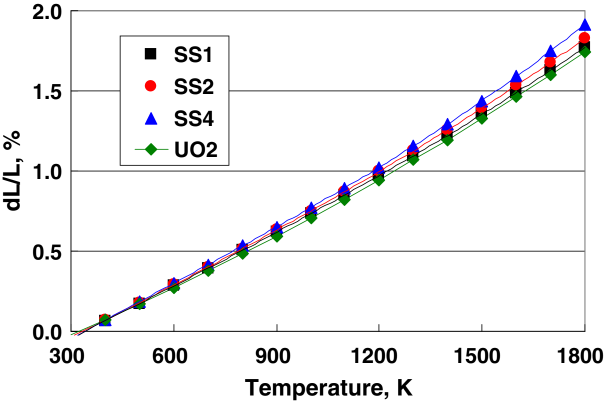

Fig. 2 Linear thermal expansions of UO_2 and simulated fuels with dissolved fission products at equivalent burn-ups of (30 (SS1), 60 (SS2), and 120 (SS4)) GWd/tU as a function of temperature

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 simulated fuels determined in this study are plotted with temperature in Fig. 2. From the figure, as expected, it is observed that the linear thermal expansions of all the specimens increase monotonically as the temperature increased. For simulated fuels, they are slightly higher than that of UO_2, and the difference between this fuel and UO_2 increases progressively with temperature. It also increases with an increase in the burn-up which represents the content of dissolved fission products in UO_2. As Momin et al. [7] noted, 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.

To confirm the effect of dissolved fission products on the thermal expansion of simulated fuels, the ratio of thermal expansions between UO_2 and simulated fuels, *re*, is introduced and defined by the following expression:

$$re = \frac{\text{Thermal expansion of simulated fuel}}{\text{Thermal expansion of UO}_2} \tag{2}$$

The ratios of thermal expansions calculated from the thermal expansion data of simulated fuels are plotted in Fig. 3. From the figure, the average *re* of simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups are 1.03, 1.06, and 1.09, respectively. They are approximately constant with temperature. We could conclude that dissolved fission products enhanced the thermal expansion of simulated fuels, and the effect was independent of temperature.

The thermal expansions of UO_2 and simulated fuels with dissolved fission products can be expressed as a function of temperature by using the following equation:

$$\Delta L/L_0(\%) = A + BT + CT^2 + DT^3 \tag{3}$$

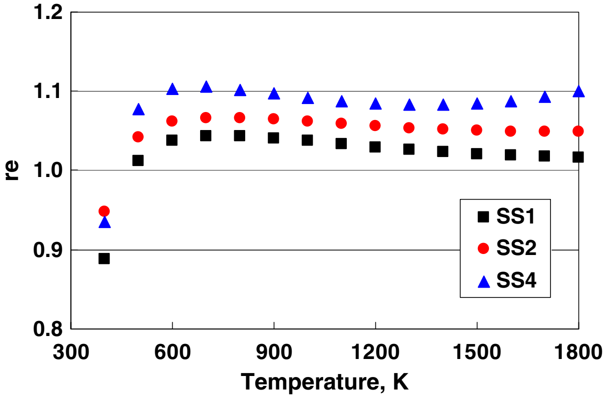

Fig. 3 Ratios of thermal expansion between UO_2 and simulated fuels with dissolved fission products at equivalent burn-ups of (30 (SS1), 60 (SS2), and 120 (SS4)) GWd/tU

Table 2 Constants of the third-degree polynomial fit of the thermal expansion, $\Delta L/L_0(\%) = A + BT + CT^2 + DT^3$, for UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups

Burn-up (GWd/tU)	*A*	*B*	*C*	*D*
30 (SS1)	-3.643×10^{-1}	1.082×10^{-3}	-3.684×10^{-8}	5.287×10^{-11}
60 (SS2)	-3.562×10^{-1}	1.058×10^{-3}	2.725×10^{-9}	4.658×10^{-11}
120 (SS4)	-4.157×10^{-1}	1.280×10^{-3}	-2.182×10^{-7}	1.262×10^{-10}
0 (UO_2)	-2.886×10^{-1}	8.529×10^{-4}	1.306×10^{-7}	1.255×10^{-11}

where ΔL represents the length variation with temperature and L_0 represents the initial length at room temperature. The values of A, B, C, and D were determined by fitting this third-degree polynomial expression to the data, and they are shown in Table 2.

The corresponding instantaneous coefficient of the thermal expansion, α, is defined by the following expression [18]:

$$\alpha = \frac{1}{L_{298}} \frac{\Delta L}{\Delta T}. \tag{4}$$

The instantaneous thermal expansion coefficients of UO_2 and simulated fuels determined in this study are plotted against temperature in Fig. 4.

The instantaneous thermal expansion coefficients of the simulated fuel pellets with the dissolved fission products can be expressed as a function of the temperature by using the following equation,

$$\alpha = E + FT + GT^2. \tag{5}$$

The values of E, F, and G were determined by fitting this second-degree polynomial expression to the data, and they are shown in Table 3.

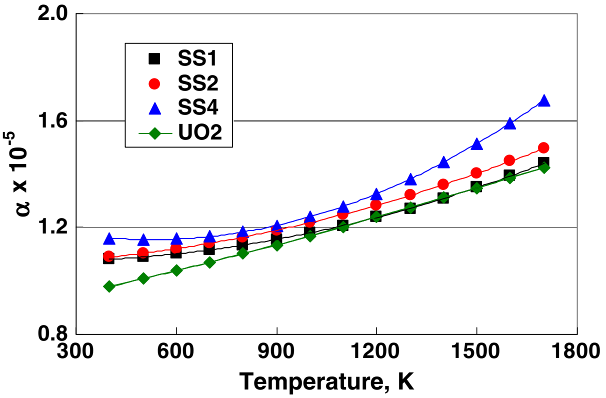

Fig. 4 Instantaneous thermal expansion coefficients of UO_2 and simulated fuels with dissolved fission products at equivalent burn-ups of (30 (SS1), 60 (SS2), and 120 (SS4)) GWd/tU as a function of temperature

Table 3 Constants of the second-degree polynomial fit of the instantaneous thermal expansion coefficients, $\alpha = E + FT + GT^2$, for UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups

Burn-up (GWd/tU)	*E*	*F*	*G*
30 (SS1)	1.079×10^{-5}	-5.782×10^{-10}	1.586×10^{-12}
60 (SS2)	1.059×10^{-5}	1.942×10^{-10}	1.397×10^{-12}
120 (SS4)	1.259×10^{-5}	-3.985×10^{-9}	3.786×10^{-12}
0 (UO_2)	8.661×10^{-6}	2.650×10^{-9}	3.765×10^{-13}

From the figure it is observed that the instantaneous thermal expansion coefficients of the simulated fuels are slightly higher than that of UO_2.

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

$$\bar{\alpha} = \frac{1}{L_{298}} \left(\frac{L_T - L_{298}}{T - 298} \right) \tag{6}$$

For the temperature range from 298 K to 1773 K, the values of the average linear thermal expansion coefficients for UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups are $1.19 \times 10^{-5}\,K^{-1}$, $1.22 \times 10^{-5}\,K^{-1}$, $1.26 \times 10^{-5}\,K^{-1}$, and $1.32 \times 10^{-5}\,K^{-1}$, respectively. These results are in agreement with those from Momin et al. [7]. They found that the coefficients of the average linear thermal expansions for UO_2 and the solid solution of UO_2 with 20 mass% Ln_2O_3 (Ln = La, Nd, Ce, Y, Sm, Gd, and Eu) are $1.08 \times 10^{-5}\,K^{-1}$ and $1.43 \times 10^{-5}\,K^{-1}$, respectively.

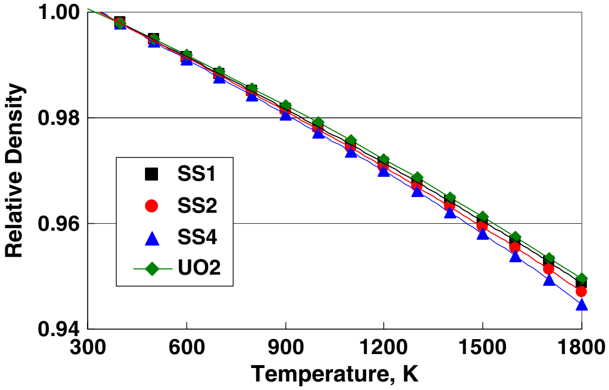

Fig. 5 Relative densities of simulated fuels with dissolved fission products at equivalent burn-ups of (30 (SS1), 60 (SS2), and 120 (SS4)) GWd/tU and UO_2 as a function of temperature

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

$$\rho(T) = \rho(298)\left(\frac{L_{298}}{L_T}\right)^3 \tag{7}$$

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 from the initial density of UO_2 and simulated fuels determined in this study are plotted against temperature in Fig. 5. From the figure it is observed that the relative density variations of all the specimens decreased monotonically with temperature, as expected. For the simulated fuels, the relative densities are lower than that of UO_2 and the difference of the relative densities of the simulated fuel and UO_2 increases with temperature.

As the temperature increased to 1773 K, the relative densities for UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups decrease to 94.5 %, 94.9 %, 94.7 %, and 94.5 % of the initial density at 298 K, respectively. The relative density variations of UO_2 and simulated fuels with the dissolved fission products can be expressed as a function of temperature by using the following equation,

$$\rho(T)/\rho(298) = H + IT + JT^2 + KT^3 \tag{8}$$

where $\rho(T)$ represents the density at temperature T, and $\rho(298)$represents the initial density at room temperature.

The values of H, J, J, and K were determined by fitting a third-degree polynomial expression to data, and they are shown in Table 4.

Table 4 Constants of the third-degree polynomial fit of the density, $\rho(T)/\rho(298) = H + IT + JT^2 + KT^3$, for UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups

Burn-up (GWd/tU)	H	I	J	K
30 (SS1)	1.011	-3.268×10^{-5}	1.397×10^{-9}	-1.372×10^{-12}
60 (SS2)	1.011	-3.196×10^{-5}	1.964×10^{-9}	-1.157×10^{-12}
120 (SS4)	1.012	-3.839×10^{-5}	6.531×10^{-9}	-3.4117×10^{-12}
0 (UO_2)	1.009	-2.579×10^{-5}	-3.675×10^{-9}	-1.500×10^{-13}

4 Conclusions

The thermal expansions of UO_2 and simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups were measured using a TMA over the temperature range of 298 K to 1773 K in order to estimate the effects of fission products forming solid solutions in UO_2 matrix on the thermal expansion of DUPIC fuel and density variation with temperature, and the following results were obtained.

(1) The thermal expansions of the simulated fuels with fission products forming a solid solution are higher than that of UO_2.
(2) In the temperature range of 298 K to 1773 K, the average linear thermal expansion coefficients of simulated fuels with dissolved fission products at (30, 60, and 120) GWd/tU equivalent burn-ups are $1.22 \times 10^{-5}\,K^{-1}$, $1.26 \times 10^{-5}\,K^{-1}$ and $1.32 \times 10^{-5}\,K^{-1}$, respectively.
(3) Dissolved fission products enhanced the thermal expansion of simulated fuels, and the effect was independent of temperature.
(4) The relative density of the simulated fuel with fission products forming a solid solution is lower than that of UO_2.
(5) The data measured and calculated in this study will be useful for performance evaluation of DUPIC fuel.

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