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Letter to the Editors

Thermal expansion of UO_2 and simulated DUPIC fuel

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Abstract

The lattice parameters of simulated DUPIC fuel and UO_2 were measured from room temperature to 1273 K using neutron diffraction to investigate the thermal expansion and density variation with temperature. The lattice parameter of simulated DUPIC fuel is lower than that of UO_2, and the linear thermal expansion of simulated DUPIC fuel is higher than that of UO_2. For the temperature range from 298 to 1273 K, the average linear thermal expansion coefficients for UO_2 and simulated DUPIC fuel are 10.471×10^{-6} and 10.751×10^{-6} K^{-1}, respectively.

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 from spent PWR fuel material without any separation of fissile elements and fission products. 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 concept was proposed and termed DUPIC fuel cycle by the Korea Atomic Energy Research Institute (KAERI) and Atomic Energy Canada Limited (AECL) in participation with the US Department of State (DOS) in 1991 [1,2]. A comprehensive research and development program is being implemented at KAERI to demonstrate the DUPIC fuel cycle concept. 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 natural uranium based CANDU fuel because of the fission products. The direct measurement of DUPIC fuel properties is very difficult in a laboratory due to its high level of radiation. As a part of the DUPIC fuel development program, the thermal properties have been investigated using simulated DUPIC fuel pellets. In this study the lattice parameters of simulated DUPIC fuel and UO_2 are measured by neutron diffraction in the temperature range of 298 to 1273 K to investigate the thermal expansion and density variation with temperature.

2. Experimental

Simulated DUPIC fuel pellets were fabricated by compacting and sintering powder made by the oxidation and reduction of oxide fuel (OREOX) process of simulated spent PWR fuel [3]. The OREOX-treated powder was pressed at 300 MPa into green pellets and sintered at 1973 K for 4 h in a flowing H_2 atmosphere. The diffraction data were obtained from the 32-detector high resolution powder diffractometer (HRPD) of KAERI, using 0.18339 nm wave length neutrons from a Ge(3 3 1) monochromator, and 0.12193 nm neutrons from a Ge(3 3 5) monochromator, both with 90° take off angles. Collimations used were: $\alpha_1 = 6'$, $10'$ or $20'$ for the in-pile collimator, $\alpha_2 = 30'$ for the collimator in front of the sample, and $\alpha_3 = 10'$ for collimator in front of the detectors. The diffraction patterns were measured from 5° to 155°.

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3. Results and discussion

The compositions and contents of precipitates were investigated using electron probe microanalysis (EPMA) in a previous study [3]. The minor components of simulated DUPIC fuel pellets added as surrogates for fission products consisted of Zr (0.537 wt%), Rh (0.047 wt%), Y (0.03 wt%), La (0.123 wt%), Ce (0.154 wt%), Sr (0.08 wt%), Ba (0.358 wt%), Mo (0.319 wt%), Pd (0.006 wt%), Nd (0.59 wt%) and O (0.48 wt%). It was observed from EPMA that the oxide precipitates mainly contained Zr and Ba, which showed a barium zirconate-type perovskite phase. The spherical metallic precipitates, which were distributed on the grain boundaries, contained Mo, Ru, Pd and Rh. The other additives such as Sr, Zr, Y, La, Nd and Ce were dissolved in the matrix. These results match well with those of Lucuta et al. [4].

The lattice parameters and cell volumes of UO_2 and simulated DUPIC fuel determined in this study are given in Table 1.

Fig. 1 shows the measured lattice parameters of UO_2 and simulated DUPIC fuel together with the results from other studies [5–7].

The lattice parameters of UO_2 obtained in our study are very similar to other results. The lattice parameters of simulated DUPIC fuel pellets, however, are lower than those of UO_2. This result is in agreement with Momin et al. [8]. He found that the cell parameters of UO_2ThO_2 and $(U_{0.2}Th_{0.8})O_2$ were all decreased with the additions of oxides of rare earths, alkaline earths and zirconium. The variations of lattice parameters, a, with temperature measured in this study are given by the following equations:

For UO_2,

$$a_T = (0.54608 \pm 0.00011) + (2.79613 \pm 0.52332)10^{-6}\ T + (3.18639 \pm 0.72476) \times 10^{-9}\ T^2 - (1.00848 \pm 0.30533) \times 10^{-12}\ T^3. \quad (1)$$

Table 1
Lattice parameter and cell volume for UO_2 and simulated DUPIC fuel

Temperature (K)	UO_2		Simulated DUPIC fuel	
	a (nm)	Volume (nm^3)	a (nm)	Volume (nm^3)
298	0.547176	0.163825	0.546832	0.163517
473	0.547993	0.164560	0.547664	0.164264
673	0.549116	0.165574	0.548825	0.165311
873	0.550276	0.166626	0.550064	0.166433
1073	0.551499	0.167739	0.551297	0.167555
1273	0.552762	0.168894	0.552564	0.168713

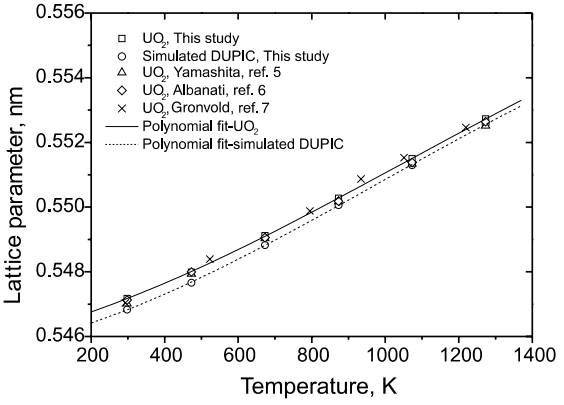

Fig. 1. Lattice parameters of UO_2 and simulated DUPIC fuel as a function of temperature.

For the simulated DUPIC fuel pellet,

$$a_T = (0.54578 \pm 0.00015) + (2.45728 \pm 0.70622)10^{-6}\ T + (3.96164 \pm 0.97806) \times 10^{-9}\ T^2 - (1.34114 \pm 0.41204) \times 10^{-12}\ T^3. \quad (2)$$

The percentage linear thermal expansion in the temperature range from 298 to 1273 K was calculated using the following expression:

$$\text{Expansion } \% = \{(a_T - a_{298})/a_{298}\} \times 100, \quad (3)$$

where a_T and a_{298} represent the lattice parameters at temperature T and room temperature, respectively. These values were fit to third order polynomials and the resultant equations are given as follow:

For UO_2,

$$\text{Expansion } \% = (-0.20012 \pm 0.02037) + (5.1101 \pm 0.95640) \times 10^{-4}\ T + (5.82334 \pm 1.32454) \times 10^{-7}\ T^2 - (1.84307 \pm 0.5580) \times 10^{-10}\ T^3. \quad (4)$$

For simulated DUPIC fuel,

$$\text{Expansion } \% = (-0.19309 \pm 0.02751) + (4.49366 \pm 1.29148) \times 10^{-4}\ T + (7.24472 \pm 1.78859) \times 10^{-7}\ T^2 - (2.45256 \pm 0.75350) \times 10^{-10}\ T^3. \quad (5)$$

The percent linear thermal expansions calculated from the above equations are shown in Fig. 2 along with the values measured in this study and the data available in the literature.

As mentioned above, the lattice parameter of simulated DUPIC fuel is lower than that of UO_2. However,

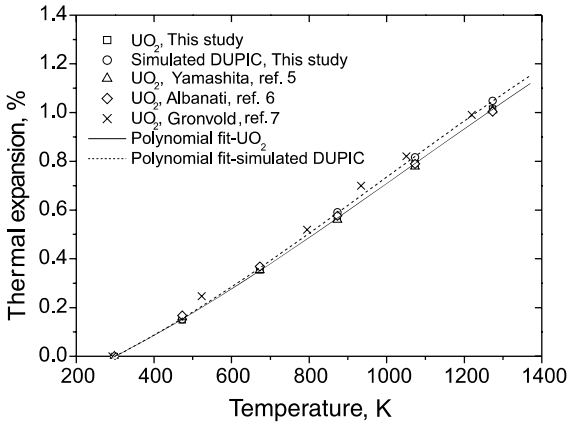

Fig. 2. Percent linear thermal expansion of UO_2 and simulated DUPIC fuel as a function of temperature.

the thermal expansion of the simulated DUPIC fuel pellet is higher than that of UO_2. Linear thermal expansion coefficient values α_T can be calculated by differentiating the expansion curve a_T with respect to temperature T:

$$\alpha_T = \frac{1}{a_{293}}\left(\frac{\partial a_T}{\partial T}\right), \tag{6}$$

where a_{293} is the lattice parameter at 293 K. These values were fit to third order polynomials and the resultant equations are given as follow:

For UO_2,

$$\alpha_T = (6.38733)\times 10^{-6} + (8.05647)\times 10^{-9}\ T - (3.12511)\times 10^{-12}\ T^2 - (1.82125)10^{-16}\ T^3. \tag{7}$$

For simulated DUPIC fuel,

$$\alpha_T = (5.41076)\times 10^{-6} + (1.34729)10^{-8}\ T - (9.68337)\times 10^{-12}\ T^2 - (2.26817)\times 10^{-15}\ T^3. \tag{8}$$

For the temperature range from 298 to 1273 K, the values of the average linear thermal expansion coefficients for UO_2 and the simulated DUPIC fuel pellet are 10.471×10^{-6} and $10.751\times 10^{-6}\ K^{-1}$, respectively.

4. Conclusions

The lattice parameters of simulated DUPIC fuel and UO_2 were measured using neutron diffraction from room temperature to 1273 K to investigate thermal expansion and density variation with temperature. The lattice parameter of simulated DUPIC fuel is lower than that of UO_2, and the linear thermal expansion of simulated DUPIC fuel is higher than that of UO_2. The data measured and calculated in this study will be useful in the evaluation of in-reactor DUPIC fuel behavior.

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