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Development of CAP process for fabrication of ThO_2–UO_2 fuels Part II: Characterization and property evaluation

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Abstract

Coated agglomerate pelletization (CAP) process is being developed for the fabrication of ThO_2–UO_2 mixed oxide fuel pellets. The procedure for the fabrication of ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets by this process has been described in detail in part I of this series of papers. The present part II deals with the characterization of the above pellets by optical microscopy, scanning electron microscopy (SEM) and electron probe microanalysis (EPMA). The microstructure of the ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets showed a duplex grain structure. Thermal expansion and thermal conductivity were measured in the temperature range from room temperature to 1500 °C. It was found that the thermal conductivity decreased by the addition of UO_2 to ThO_2 at any temperature. Addition of 20%UO_2 to ThO_2 caused very large decrease in thermal conductivity. The thermal expansion of ThO_2–20%UO_2 pellet was different from that of ThO_2 and ThO_2–4%UO_2, e.g. it increased more rapidly with increasing temperature in the temperature range of 1000–1500 °C.

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1. Introduction

Fabrication of ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets by a novel technique (CAP process) has been described in detail in part I of this series of papers. This part (part II) deals with the microstructural evaluation and determination of thermophysical properties of the above pellets. The microstructure is important since it is deeply related to the irradiation behaviour. It controls the in-pile fuel behaviour like plasticity, in-pile creep and swelling [1]. The microstructure of the fuel is intimately related to the behavior of the fission gases. The improvement in plasticity and fission gas release can be attained by modifying the microstructures during fabrication [2–6]. The conventional process for nuclear ceramics fabrication consists of a number of stages including calcination, milling, incorporating additives, pressing, drying and densification. Since each of these process steps affects the microstructure of the fuel pellets they must all be understood. It is possible to obtain a wide range of microstructures for ThO_2–UO_2 system if the CAP is chosen. This process can tailor the microstructure for better performance during irradiation.

The behaviour of nuclear fuel during irradiation is largely dependent on its physico-chemical properties and their change with temperature and burn-up [7]. Thermal conductivity is an important parameter to understand the performance of the fuel pins under irradiation [1]. If the thermal conductivity is low, the temperature gradient in the radial

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direction of the fuel pellet is large causing to give high temperature at the central part of the fuel pin [1,7]. The thermal conductivity of nuclear fuel influences almost all important processes such as fission gas release, swelling, grain growth etc. and limits the linear power [8,9]. The changes in thermal conductivity occurs during irradiation by the formation of fission gas bubbles, build-up of fission products, and by the change of oxygen-to-metal ratio (O/M) [10]. Hence the knowledge of thermal conductivity is needed to evaluate its thermal performance. The coefficient of thermal expansion (CTE) values are needed to calculate stresses occurring in the fuel and cladding on change in temperature. If the thermal expansion varies considerably between the fuel and cladding, stresses will be accumulated during the thermal cycling [11]. This can lead to the deformation of the clad and eventually may result in the breakage of the clad. Hence, precise evaluation of CTE data of the fuel is needed.

This paper deals with characterization of ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets made by CAP process with the help of optical microscopy, XRD, SEM and EPMA. The microstructures are evaluated in terms of grain size, pore size and its distribution and homogeneity of uranium. This paper also describes the evaluation of thermal conductivity and thermal expansion for the mixed oxide pellets of the above compositions at temperatures up to 1500 °C, and discusses the effect of composition, microstructure and O/M ratio on the experimental results.

2. Experimental

2.1. Sample preparation

ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets for this study was prepared by the CAP process as described in part I. The pellets were sintered in air at 1450 °C for 8 h. The sintered pellets were about 12 mm in diameter and around 10 mm in length. Table 1 gives details of the samples studied in this work. For metallography, the sintered pellet was fixed by Araldite and ground using successive grades of emery papers. The final polishing was done using diamond paste. The pellet was removed from the mount by dissolving the mount in acetone and then etched thermally by holding the sample at 1400 °C for 4 h in air. The grain size was determined by the intercept method. The microstructure was then characterized by scanning electron microscopy. The distribution of Th, U and O was determined by X-ray mapping and also by line scans with the help of EPMA. A few ThO_2 pellets were also made using the above technique as control samples under the same conditions.

2.2. Thermal conductivity

The studies were carried out using a Ulvac Sinku-Riko (model TC 3000) thermal diffusivity apparatus. For the thermal diffusivity measurement, the sintered pellet was sliced into discs of about 10 mm diameter and 2 mm thickness using a low speed cut-off wheel. A pulse of laser was projected on to the front surface of the pellet and the temperature rise on the rear side of the pellet was recorded as a transient signal by using an infrared detector. The measurements of the thermal diffusivity were carried out up to 1500 °C in vacuum at a pressure of less than 10^{-5} Pa. At each temperature, measurement was carried out thrice. The average value of these three measurements was used and the experimental uncertainty associated with these measurements was within 5%. The thermal diffusivity (α) was calculated from the following relationship:

$$\alpha = WL^2/\pi t_{1/2}, \tag{1}$$

where $t_{1/2}$ the time required in seconds to reach half of the maximum temperature rise at the rear surface of the sample and L the sample thickness in millimeter. W a dimensionless parameter which is a function of the relative heat loss from the sample during the measurement. The data was corrected for radiation heat losses by the method of Clark and Taylor [12].

The thermal conductivities of ThO_2, ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets were derived from the measured values of thermal diffusivity data determined by laser flash technique by using the relation:

$$\lambda = \alpha\rho C_p, \tag{2}$$

where λ the thermal conductivity, α the thermal diffusivity, ρ the density of the material and C_p its specific heat at constant pressure. The literature values of specific heat were used in this study and specific heat capacity of (Th,U)O_2 was estimated from those of ThO_2 [13] and UO_2 [7] using the Kopp's law.

2.3. Thermal expansion

The thermal expansion of ThO_2, ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets was measured by a Netzsch (model 402E) horizontal dilatometer in Ar atmosphere with a heating rate of 6 K/min. The accuracy of the measurement of

Table 1
Density, pore fraction and O/M ratio of ThO_2, ThO_2–4%UO_2 and ThO_2–20%UO_2 pellets fabricated by CAP process

Sample	O/M ratio	Bulk density g/cm^3	Theoretical density (T.D.)	% T.D.	% Pore fraction
ThO_2	2.00	9.203	10.00	92.03	7.97
ThO_2–4%UO_2	2.01	9.451	10.038	94.15	5.85
ThO_2–20%UO_2	2.07	9.211	10.192	90.37	9.63