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Fabrication and characterization of dual phase magnesia–zirconia ceramics doped with plutonia

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Abstract

Dual phase magnesia–zirconia ceramics doped with plutonia are being studied as an inert matrix fuel (IMF) for light water reactors. The motivation of this work is to develop an IMF with a thermal conductivity superior to that of the fuels based on yttria stabilized zirconia. The concept uses the MgO phase as an efficient heat conductor to increase thermal conductivity of the composite. In this paper ceramic fabrication and characterization by scanning electron microscopy, energy and wavelength dispersive X-ray spectroscopy is discussed. Characterization shows that the ceramics consist of the two-phase matrix and PuO_2-rich inclusions. The matrix is comprised of pure MgO phase and $MgO–ZrO_2–PuO_2$ solid solution. The PuO_2-rich inclusion contained dissolved MgO and ZrO_2.
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1. Introduction

Advanced fuel cycle initiative (AFCI) is being pursued by the United States Department of Energy with a four-prong objective: recovery of energy from spent nuclear fuel (SNF), reduction of the inventory of civilian plutonium, reduction of the toxicity and heat load of stored SNF, and more effective use of the SNF repository. AFCI's mission is to develop and demonstrate technologies that enable the transition to a stable, long-term, environmentally, economically and politically acceptable fuel cycle.

Use of existing light water reactors (LWR) as a neutron source to fission plutonium and transmute minor actinides provides a near term opportunity for very effective in-reactor disposition of these surplus nuclear materials [1]. A need for a non-fertile matrix material that can be safely used in LWR fuels as plutonium and minor actinide dilutant drives material development research in this field.

Yttria stabilized zirconia (YSZ) is the most evolved candidate for use in IMF. Both steady-state [2] and transient [3] irradiations of YSZ-based IMF have been performed. Material properties of YSZ have been closely examined [4,5]. Out-of pile irradiation studies designed to understand the mechanism of radiation damage have been completed [6]. Neutronic feasibility of YSZ-based IMF has been also assessed

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[7] complemented by core burnup calculations and accident analyses [8]. Despite its excellent radiation resistance, compatibility with reactor materials and good neutronic properties, low thermal conductivity is the main disadvantage of YSZ. According to a recent analysis [7] fuel centerline temperature of the YSZ-based IMF may be 100 K higher than the limit specified for LWRs. Safe use of YSZ as a matrix in LWR fuel is only possible if a reactor is operated at a lower power or if fuel pellets feature central voids. Both measures increase the cost and decrease the feasibility of plutonium disposition.

Use of composites containing a phase with a higher thermal conductivity could improve performance characteristics of zirconia-based IMF. Use of MgO as such a phase has been proposed [9,10]. It has been shown [10] that dual phase MgO–ZrO_2 ceramics have the thermal conductivity superior to that of UO_2 and have notable resistance to the water at the temperature of 573 K and pressure 8.6 MPa, making them attractive for use as an IMF matrix. Development of IMF based on MgO–ZrO_2 ceramics continues at Idaho National Laboratory with the support of the AFCI. The present paper describes results of the first experiment to fabricate dual phase magnesia–zirconia ceramics doped with plutonia as a potential IMF form for use in LWRs.

2. Fabrication

Dual phase magnesia–zirconia ceramics doped with plutonia were fabricated from the oxide powder mixture using conventional pressing and sintering techniques. The fabrication process was based on the earlier work [10] that dealt with non-radioactive materials. The flow diagram of the fabrication process is shown in Fig. 1.

Pre-weighed amounts of magnesia and magnesium zirconium oxide powders were combined with water in a beaker. The weight of water was approximately three times greater than the weight of the powders combined. The water and powder mixture was stirred using a magnetic stirring bar for 6 h. The slurry was dried in air at 353 K for 5 h. The resulting powder was transferred into an alumina crucible and heat-treated at 1273 K for 5 h in a high temperature tube furnace. The ramp-up and ramp-down rates were 10 K min^{-1}. Upon cool-down, zinc stearate (Fisher Scientific Fair Lawn, NJ Z-78–4, lot 871095, UPS grade) in the amount of 1% by weight was mixed into the powder using a mortar and a pestle. The powder was then pressed into pellets with a force of 13.34 kN using a cylindrical die of 12.72 mm diameter. Resulting MgO–ZrO_2 pellets weighing 3.8986 g and 3.3571 g were transferred into a glovebox. Once in the glovebox, the pellets were ground and 1.0265 g of PuO_2 powder was mixed in using a mortar and pestle. The powder was then pressed into pellets with a force of 44.45 kN using a cylindrical die of 12.72 mm diameter. Resulting pellets were ground into powder using a mortar and pestle. The powder was passed through a sieve with an aperture size of 250 μm. The mixture was pressed again into pellets with a force of 13.34 kN using a cylindrical die of 12.72 mm diameter. The pellets were placed into an alumina crucible and sintered in air for 7.5 h at 1973 K in a high temperature box furnace. The ramp-up rate was 10 K min^{-1} up to 1273 and 5 K min^{-1} from 1273 to 1973 K. The ramp-down rate was 10 K min^{-1}. The pellets were cooled in the furnace after sintering.

Magnesium oxide was procured from Cerac Incorporated (Milwakee, WI, item M-1017, lot X25111, typically 99.95% pure). Magnesium zirconium oxide supplied by Alfa Aesar (Ward Hill, MA, stock 12343, lot C01E, 99.7% metals basis) was used as a source of zirconia. Use of magnesium zirconium oxide facilitated obtaining a homogeneous distribution of MgO and ZrO_2 in the final product.

3. Characterization of dual phase magnesia–zirconia ceramics doped with plutonia

3.1. Visual inspection, dimensions, weight and density

Two ceramic pellets were fabricated. Photographs of the pellets are shown in Fig. 2. As-sintered pellets are shown in Fig. 2(a). Pellet cross-section exposed by cutting a pellet with a diamond saw is shown in Fig. 2(b). Each pellet featured one crack near one face as illustrated in Fig. 2(c). The crack was probably caused by density gradients developed during pressing.

The pellets were weighed and measured with a caliper. Results of the measurements are shown in Table 1. Pellet diameter was measured three times: in the mid-pellet and near each face. Pellet hourglassing developed during sintering was manifested by diameter decrease in the mid-pellet region.

Pellet density values derived from weight and volume and measured by water immersion technique are included in Table 1. The pellet density was