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Spark plasma sintering of diamond-reinforced uranium dioxide composite fuel pellets

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HIGHLIGHTS

- Fabricated UO_2–diamond composite fuel pellets using spark plasma sintering (SPS).
- Increased thermal conductivity by up to 41.6% compared to standard pure UO_2 fuel.
- Investigated the effect of diamond particle size on properties of composites.

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C. Material properties

ABSTRACT

In an effort to increase the thermal conductivity of UO_2 fuel pellets, high density UO_2–5 vol% diamond composite pellets were fabricated using the spark plasma sintering (SPS) technique. Diamond particles with nano-size (0.25 μm) and several micro-sizes (3 μm, 12 μm and 25 μm) were mixed with UO_2 powder and sintered using SPS at 1300–1600 °C with a hold time of 5 min. The resultant density, chemical reaction, microstructure, thermal conductivity and Young's modulus of the sintered pellets were investigated. The pellets with 3 μm diamond particles had a uniform distribution of particles as well as better thermal and mechanical properties compared to others. An increase in thermal conductivity of up to 41.6%, 38.3% and 34.2% at 100 °C, 500 °C and 900 °C, respectively, was measured in the UO_2–diamond composite pellets compared to the pure UO_2 fuel pellets.

1. Introduction

Uranium dioxide (UO_2) is the most common fuel material used in commercial nuclear power reactors. Compared to other fuel materials, UO_2 has the advantage of high melting point (Carbajo et al., 2001), good high-temperature stability, good chemical compatibility with both cladding and coolant and resistance to radiation (Glasstone and Sesonske, 1982). However, the main drawback of a UO_2 fuel pellet is its extremely low thermal conductivity, which results in a fairly high centerline temperature during operation in a reactor and a steep radial temperature gradient, typically around a 500° K drop within a pellet radial distance of 5 mm. Thermal stress induced by this large temperature gradient may lead to cracking within the fuel pellets and cause accelerated release of fission gases, both of which limit the operational life of UO_2 fuel in a reactor. Therefore, the importance of increasing the thermal conductivity of UO_2 fuel cannot be over emphasized.

The idea of incorporating high thermal conductivity material into a UO_2 matrix has been gaining popularity in recent years. However, initial attempts to sinter UO_2 powder with second phase high conductivity materials such as SiC have been met with limited success, especially in terms of reaching the desired 96.5% of the theoretical density (TD) in the final pellet (Wang, 2008). Ishimoto et al. (1996) considered BeO as a second phase and sintered UO_2–BeO composite in a reducing atmosphere successfully. Most recently, numerous other second phase materials have been proposed in the literature. Yeo et al. (2013a, 2013b) and Cartas et al. (2015) utilized spark plasma sintering and fabricated UO_2–SiC and UO_2–carbon nanotube (CNT) composite fuel pellets, respectively. Lee et al. (2013) proposed the idea of UO_2–graphene composite and studied the thermal properties through numerical simulation. Among all the potential second phase materials, diamond has extremely high thermal conductivity (about 22 W/cm K at room temperature), which is the highest of any solid material (Slack, 1964). Due to its sp^3 hybridization bond structure, diamond is found to be resistant to the radiation environment (Dilawar et al., 1997). Therefore, diamond makes an ideal second phase for incorporation into UO_2 pellets to achieve a higher thermal conductivity in the composite.

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From the economics perspective, the price of synthetically produced diamond powder is much lower (~\$2.5/g) than that of CNT (~\$270/g) or graphene (~\$125/g), which is beneficial for commercialization.

Fabrication of UO_2 fuel using conventional oxidative sintering methods requires elevated temperatures up to 1700 °C for 2–4 h. The entire temperature cycle (heating time, hold time, and cooling time) may take 10–15 h due to slow heating rate of only around 5 °C/min (Carrea, 1963; Williams et al., 1959; Stuart and Adams, 1975). Ishimoto et al. (1996) sintered UO_2–BeO composite at 2073 K for 2 h using a conventional sintering method. However, using spark plasma sintering (SPS) technique, Yeo et al. (2013a) achieved high density UO_2–SiC pellets at 1350 °C with a hold time as low as 5 min. SPS is a promising approach for processing difficult-to-sinter powders such as UO_2. Advantages of using SPS over traditional sintering of UO_2 can be found in the work of Ge et al. (2014). The objective of this work is to fabricate UO_2–diamond fuel pellets using SPS and investigate the microstructure and properties of this novel composite fuel.

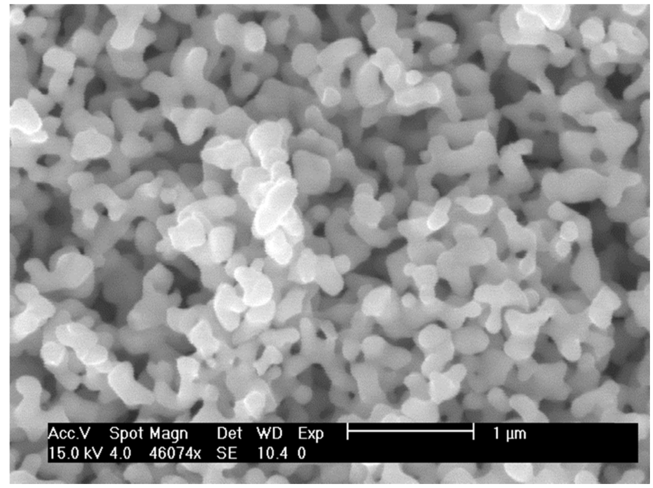

Fig. 1. SEM image of as-received UO_2 powder.

2. Experimental

2.1. Powder preparation

Figs. 1 and 2 show SEM images of the as-received UO_2 and diamond powders, respectively. The uranium dioxide powder was obtained from AREVA Federal Services, Hanford, Washington, USA. The powder was reported to have a bulk density of 2.3 g/cm^3, mean particle diameter of 2.4 µm and a BET surface area of 3.11 m^2/g. The grain size was determined to be 200–400 nm, see Fig. 1. The O/U ratio was determined to be 2.11 using ASTM equilibration method (C1430-07). The diamond powder was obtained from Advanced Abrasives, Pennsauken, NJ. Four sizes of diamond particles with a mean particle size of 0.25 µm, 3 µm, 12 µm and 25 µm, were used in this study.

A SPEX-8000 shaker was used to blend UO_2 and a 5 vol% diamond powder for 1 h with a blending aid 2,3-dihydroperfluoropentane. The use of a blending aid was proven to be effective and non-contaminative to the final powder (Chen et al., 2014).

2.2. SPS

A graphite die with inner diameter of 12.7 mm and outer diameter of 31.5 mm was used to sinter the mixed powder. A thin graphite foil was inserted into a graphite die and then 4 g of UO_2 + diamond mixture was poured into the die. Graphite punches were used on either side of the die to hold the powder. The graphite foil prevents reaction between the powders and the die. A Dr. Sinter® SPS-1030 spark plasma system (SPS) machine was used to sinter the blended powder. The entire die assembly with powder and punches was placed in the SPS chamber. During sintering, a vacuum of less than 10 Pa was maintained in the chamber and the temperature was measured using a pyrometer that focused on the surface of the graphite die. A temperature ramp rate of 100 °C/min was employed and the maximum sintering temperature was varied from 1300 °C to 1600 °C with a hold time of 5 min for various powder mixtures. An axial pressure of 36 MPa was applied when the maximum sintering temperature was reached, and the pressure was released when the cooling process started. More detailed description of this method can be found in recent publications (Yeo et al., 2013a, 2013b; Chen et al., 2014; Ge et al., 2013). It is important to note that

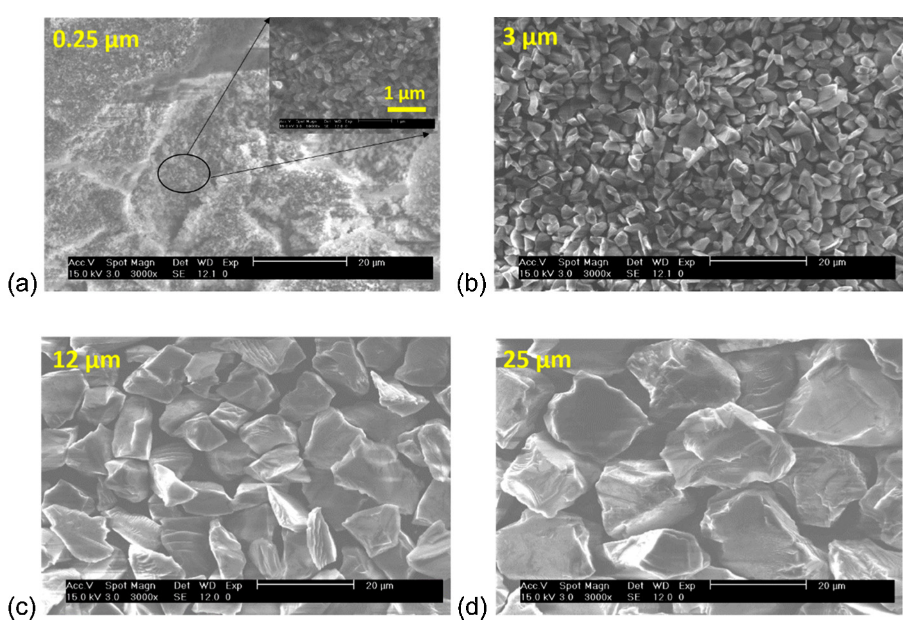

Fig. 2. SEM images showing morphologies of diamond powder with a mean particle size of (a) 0.25 µm, (b) 3 µm, (c) 12 µm, and (d) 25 µm.