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The influence of SiC particle size and volume fraction on the thermal conductivity of spark plasma sintered UO_2–SiC composites

Sunghwan Yeo [a], Ronald Baney [a], Ghatu Subhash [b,*], James Tulenko [a]

[a] Materials Science and Engineering Department, University of Florida, Gainesville, FL 32611, USA
[b] Mechanical and Aerospace Engineering Department, University of Florida, Gainesville, FL 32611, USA

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ABSTRACT

This study examines the influence of Silicon Carbide (SiC) particle addition on thermal conductivity of UO_2–SiC composite pellets. UO_2 powder and β-SiC particles of different sizes and of different volume fractions were mechanically mixed and sintered at 1350–1450 °C for 5 min by Spark Plasma Sintering (SPS). The particle size (0.6–55 μm diameter) and volume fraction (5–20%) of SiC were systematically varied to investigate their influence on the resulting UO_2–SiC composite pellet microstructure and the thermal properties. It was found that SiC particle size less than 16.9 μm with larger volume fraction is more effective for improving the thermal conductivity of the fuel pellets. Scanning Electron Microscopy examination revealed micro-cracking and interfacial debonding in the composites containing larger size SiC particles (16.9 and 55 μm) which resulted in reduced thermal conductivity. For the UO_2–SiC composite pellets containing 1 μm diameter SiC particles, the thermal conductivity increased almost linearly with volume fraction of particles. However, the addition of a larger volume fraction of SiC reduces the amount of heavy metal in the composite pellet and therefore a higher U-235 enrichment is necessary to compensate for the heavy metal loss. The experimental thermal conductivity values of the UO_2–SiC composite pellets are in good agreement with the theoretical values based on the available model in the literature.

1. Introduction

The primary ceramic fuel used in nuclear reactors, uranium dioxide (UO_2), has a low thermal conductivity (7.7 W/m K at 300 K [1]) which results in a decrease in the energy output due to low heat transfer efficiency from fuel to coolant. The introduction of high thermal conductivity fuel pellets enables a nuclear reactor to produce more thermal energy while maintaining plant safety due to lower pellet centerline temperature and thermal gradient, resulting in a lower level of fission gas release and thermal cracking.

In recent years, Spark Plasma Sintering (SPS) has evolved as a promising sintering technique for rapid fabrication of UO_2 pellets of required shape and size [2]. In our previous research [3] it has been shown that enhanced thermal conductivity of UO_2–10 vol%-SiC composite fuel pellets can be fabricated by the Spark Plasma Sintering (SPS) technique. In that study, SPS provided higher density composites, better interfacial contact, and reduced chemical reaction between UO_2 and SiC particles, compared to conventional sintering. SPS pellets also revealed a thermal conductivity increase of up to 62.1% at 900 °C compared to the literature value of UO_2 [1]. Because of its unique and superior properties such as high thermal conductivity, low neutron cross section, high melting point, and great chemical stability, Silicon Carbide (SiC) was chosen as the secondary phase in the UO_2 matrix to form heat conducting paths in the ceramic composites.

The influence of particle size and volume fraction of second phase particles on the effective thermal conductivity of a composite and its association with the interfacial thermal resistance have been well documented in literature. Hanada et al. [4] determined that diamond particle size and volume fraction had a significant influence on the effective thermal conductivity of copper–diamond composites. They found that the composites containing larger diamond particles up to 7.7 μm diameter and smaller volume fraction near 1% show higher thermal conductivity. Bai et al. [5] fabricated $MoSi_2$–SiC composites containing 10, 20, and 30 vol% of 0.5 μm and 100 nm SiC particles and found that the composite showed lower thermal conductivity with decreasing SiC particle size and increasing volume fraction due to the interfacial thermal resistance. Chu et al. [6] sintered Cu–Carbon Nanotube (CNT) composites and found that these composites had much lower thermal conductivity than calculated thermal conductivity value based on the rule of mixture which ignored the interfacial resistance. In general, it is found that ceramic composites containing larger particles (low surface to volume ratio) reduce the interfacial thermal resistance, and hence, potentially increase the effective thermal conductivity of ceramic composites.

* Corresponding author. Tel.: +1 352 392 7005; fax: +1 352 392 7303.
E-mail address: Subhash@ufl.edu (G. Subhash).

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In the current study, a series of UO_2–SiC composite fuel pellets with different sizes and volume fractions of SiC particles was fabricated using the SPS technique. The thermal conductivities of these composites were measured and compared to the values determined from theoretical formulations available in the literature. During the fabrication process, the sintering parameters such as hold time, up/down ramp rate, and pressure were kept constant so as to investigate only the effects of SiC particle size and volume fraction on the resulting thermal conductivity of the composite pellets.

2. Experimental

2.1. Fabrication of UO_2–SiC composite pellets

UO_2–SiC pellet fabrication procedure using SPS was described in detail in our recent publication [3]. Therefore, only a brief description is provided here. The uranium dioxide ($UO_{2.11}$) powder was obtained from AREVA NP, Richland, WA and the SiC powder was obtained from Superior Graphite, Inc., Chicago, IL. The SiC powder was reported to have a high purity (>98%), a surface area from gas absorption method [7] of 1.46 m^2/g, a particle density range of 3.1–3.2 g/cm^3, and particle mean diameters of 0.6, 1.0, 9.0, 16.9, and 55 μm. Reported major impurities are listed in Table 1. The UO_2 and SiC powders were mixed in a ceramic vial with stainless steel balls and a blending aid, 2,3-dihydroperfluoropentane, and blended in a SPEX 8000 shaker for 1 h. The blended mixture was then dried up in a hood for at least 6 h to decompose the blending aid. It was assumed that the blending aid was all decomposed because the weight change between unblended powders and dried mixture was less than 0.13%. For each mixing run the SiC mean particle size and the volume fraction of SiC powder in the mixture with UO_2 were varied as shown in Table 2 to investigate their effect on the thermal conductivity of the resulting UO_2–SiC composite pellet. The SiC particle sizes used in this study are most widely used in various applications and are also available as high purity (>98%) powders, and hence were obvious choice in our study. SiC particles with 1 μm size at 5, 10, 15, and 20 vol% were chosen to fabricate UO_2–SiC composite pellets. This volume range was selected because 20 vol% is the maximum range where the available models are valid (to be described in the next section) so that we can compare the resulting thermal conductivity of the composites with model prediction. Also, excessive addition of SiC in nuclear fuel is unrealistic due to the exorbitant cost and stringent regulation of U-235 enrichment process which is necessary to compensate reduced fissile isotope in UO_2–SiC composite fuel. SiC particles dispersed in UO_2 powders were then sintered using a Dr. Sinter® SPS-1030 system at 1350 °C and 1450 °C for 5 min in a vacuum (~30 m Torr). The ramp up/down rate and mechanical pressure at the maximum sintering temperature were held constant at 100 °C/min and 36 MPa, respectively.

The heat treatment procedure described in ASTM C 1430-07 was conducted on the sintered composite pellet to reduce UO_{2+X} to stoichiometric $UO_{2.0}$, which is known to have the optimum thermal properties [8]. This process is also required to produce similar O/U ratio in all fabricated UO_2–SiC composites. The ramp up/down rate and maximum temperature were set at 2.6 °C/min and 800 °C, respectively. The heat treatment was performed in a Lindberg® alumina tube furnace using 4%H_2–N_2 gas with a dew point maintained at 35 °C.

Table 1
Reported major impurities and their fractions in SiC powder.

Element	Maximum fraction
Carbon (free)	1.0 wt%
Oxygen	1.0 wt%
Nitrogen	0.4 wt%
Silicon (metallic)	0.03 wt%
Iron	800 ppm
Calcium	400 ppm

2.2. Characterization methods

The weight of each pellet in air and water was measured and the average density was calculated from three weight measurements per pellet using the Archimedes principle. The measured density of the composite was then compared with theoretical density obtained from the rule of mixture [9].

$$\rho_c = \rho_{UO_2}(1 - V_p) + \rho_{SiC}V_p \quad (1)$$

where ρ_{UO_2}, ρ_{SiC}, and V_p are the densities of UO_2 and SiC, and the SiC volume fraction, respectively.

The microstructure of the fabricated composite pellets were observed using a scanning electron microscope (SEM, JEOL JSM-6335F). The pellets were metallographically polished with successively smaller grit SiC abrasive paper and finally with 0.06 μm colloidal silica. The surface was thermally etched at 1340 °C in Ar atmosphere for 4 h to reveal the grain boundaries of UO_2 matrix in the composite pellets.

The measurement of thermal diffusivity was carried out at 100, 500, and 900 °C using a laser flash instrument (AnterFlashline®3000) with a Xenon discharge pulse for 1 μs duration. Three measurements were performed at each temperature on each pellet and the average diffusivity was obtained. The specific heat capacity of UO_2–SiC composite pellet was calculated using the Neumann–Kopp rule [10] i.e.,

$$C_p = C_{UO_2}(1 - f_p) + C_{SiC}f_p \quad (2)$$

where C_{UO_2}, C_{SiC}, and f_p are theoretical specific heat capacities of UO_2 and SiC, and weight fraction of SiC particles, respectively, at a specific temperature. C_{UO_2} and C_{SiC} at 100 °C, 500 °C, and 900 °C are listed in Table 3. The thermal conductivity, K, of composite pellets was then determined from the relation.

$$K = DC_p\rho_c \quad (3)$$

where D and ρ_c are the thermal diffusivity and density of the composite, respectively.

3. Results and discussion

3.1. Size effect of SiC particles on UO_2–5 vol% SiC composite properties

The micro-morphologies and thermal properties of UO_2–5 vol%SiC composite fuel pellets containing SiC particles with five

Table 2
Details of SiC particle size, volume fraction, and sintering conditions in the SPS.[a]

SiC particle mean diamter (μm)	SiC volume fraction (%)	Maximum sintering temperature (°C)	% TD of the composite pellet ± SD
0.6	5	1350	95.25 ± 0.24
1	5	1350	95.27 ± 0.3
1	5	1450	96.81 ± 0.39
1	10	1450	96.63 ± 0.35
1	15	1450	95.14 ± 0.23
1	20	1450	94.41 ± 0.3
9	5	1350	95.15 ± 0.09
16.9	5	1350	94.75 ± 0.17
55	5	1350	95.1 ± 0.13

[a] Hold time = 5 min; ramp up/down rate = 100 °C/min; pressure = 36 MPa.