

Thermal conductivity of CeO₂-Ce₃Si₂ hybrid ceramic

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1. Introduction

About 200 °C lower fuel centerline temperature than that of conventional light water reactors has to be one of many unique features of an innovative small modular reactor (SMR) named ATOM (Autonomous, Transportable, On-demand reactor-Module) to enable unmanned load-following operation. Since the coolant temperature needs to be maintained at the same level with typical PWRs, UO₂-based hybrid ceramic fuel including high thermal conductivity additives may be the key to resolve this issue.

Uranium sesquisilicide (U₃Si₂) - among a few considerable additive materials - is selected as the first candidate to be tested due to slightly increasing fissile density with increasing additive composition and acceptable corrosion resistance [1]. In this study, we used CeO₂-Ce₃Si₂ as surrogate material and measured thermal conductivity of various compositions of the hybrid ceramic up to 1000 °C as a preliminary study on the fabricability and thermal conductivity behavior of UO₂-U₃Si₂.

2. Experimental

2.1 Powder preparation

To synthesize Ce₃Si₂ powder, as-received cerium (99.99%, Avention) were mixed with silicon powder in a 3:2 molar ratio under an argon atmosphere and high energy ball milled for 10 h with 10-mmD ZrO₂ media at 500 rpm in 80 mL ZrO₂ vessel [2]. The mass ratio of starting material to media was 1:10. The stoichiometry of resultant material was characterized using X-ray diffraction (XRD, D/MAX2500V).

2.2 Pellet fabrication

As-received CeO₂ powder and synthesized Ce₃Si₂ powder were blended in a turbula mixer at 150 rpm for 0.5 h. The blended power was cold compacted at 350 MPa for 3 min into a 12.7 mmD green pellet in a stainless steel die. The green pellets were sintered at 1500 °C for 1 h in air atmosphere with constant power of 2 kW and 2.45 GHz microwave.

The sintered density and thermal conductivity of CeO₂-xCe₃Si₂ (x = 10, 20, 30 wt%) pellets were measured using Archimedes immersion method and laser flash analysis (LFA, NETZSCH LFA467HT), respectively.

3. Results

Figure 1 shows the scanning electron microscopy (SEM) image of high energy ball milling (HEBM) synthesized Ce₃Si₂ powder. Particle showed arbitrary shapes with ~2 μmD. Figure 2 is the XRD 2θ spectra of the synthesized Ce₃Si₂, which shows good match with Ce₃Si₂ database, however not exact, suggesting the formation of nonstoichiometric cerium silicide compound. Also, energy dispersive spectroscopy (EDS) confirmed ~1.44wt% Zr impurity perhaps due to ZrO₂ infiltrated from the milling vessel and media during HEBM process.

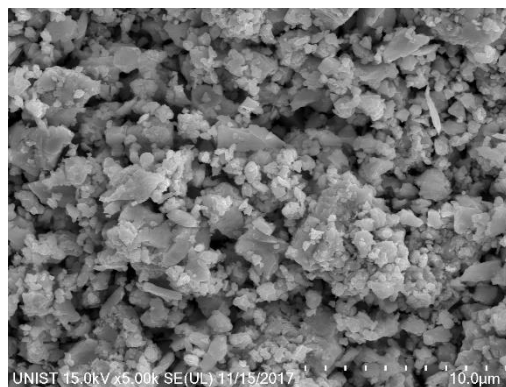


Fig. 1. Morphology of Ce₃Si₂ powder obtained by HEBM for 10 h milling at 500 rpm.

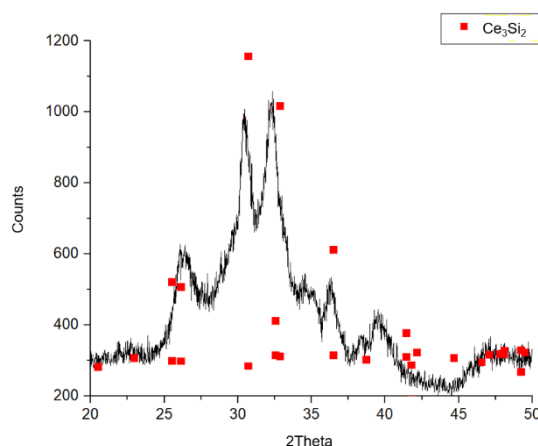


Fig. 2. XRD pattern of Ce₃Si₂ powder obtained by HEBM for 10 h milling at 500 rpm.

The microwave-sintered CeO₂-Ce₃Si₂ pellets are shown in Fig. 3 and measured average pellet density was ~93%TD. Figure 4 shows the LFA-measured thermal conductivity of three compositions of the hybrid

ceramics. Mainly two significant observations can be made from this measurement: firstly, distinctly enhanced thermal conductivity with increasing Ce_3Si_2 composition for entire temperature range up to 1000 °C; secondly, increasing thermal conductivity of the silicide-rich ceramic (CeO_2 -30wt% Ce_3Si_2) for high temperature region (> 700 °C), which is in contrast to the monotonic decrease of the other ceramics thermal conductivity. This positive thermal conductivity gradient with respect to temperature increase implies the silicide hybrid ceramic fuel deserves serious consideration to be used in the ATOM core, since effective thermal conductivity can be doubled in the central region of the fuel with increasing fissile density.



Fig. 3. CeO_2 - Ce_3Si_2 composite pellets fabricated by microwave sintering at 1500 °C for 1 h.

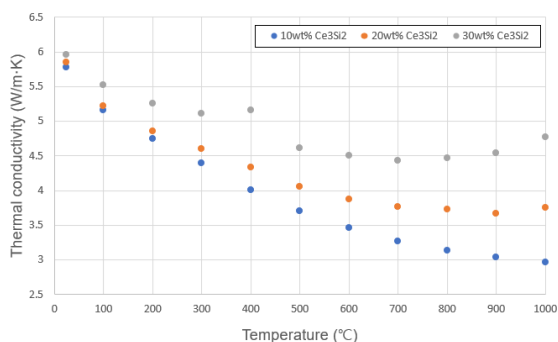


Fig. 4. Thermal conductivity of CeO_2 - $x\text{Ce}_3\text{Si}_2$ ($x = 10, 20, 30$ wt%) composite pellets.

4. Summary

For an innovative SMR named ATOM (Autonomous, Transportable, On-demand reactor-Module) that aims unmanned load-following operation, UO_2 - U_3Si_2 hybrid fuel concept was developed to lower the centerline temperature by ~200 °C. To test the feasibility of the concept, CeO_2 - $x\text{Ce}_3\text{Si}_2$ ($x = 10, 20, 30$ wt%) pellets were fabricated as the surrogate and the thermal conductivity of the hybrid ceramics was measured up to 1000 °C using LFA. Enhanced thermal conductivity over entire temperature range was confirmed with increasing Ce_3Si_2 composition. In particular, CeO_2 -30wt% Ce_3Si_2 exhibited increasing thermal conductivity with increasing temperature for high temperature region above 700 °C, which provokes the further study on silicide-rich UO_2 - U_3Si_2 hybrid ceramic fuel.

ACKNOWLEDGEMENT

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