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Enhanced Thermal Conductivities of Mo wire reinforced Zirconium Oxide Pellets

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1. Introduction

Research to improve low thermal conductivity of UO_2 pellets has been reported. Previously, the fabrication of thin tungsten or molybdenum channels at the grain boundaries of UO_2 pellets via sintering WO_3/UO_2 or MoO_3/UO_2 mixture compacts under a reducing atmosphere has been reported [1-3].

Considering the reported results, a promising concept for increasing the thermal conductivity of UO_2 is forming 2D or 3D networks of thermally conducting phases inside the oxide pellets. But there is no reported results using high-thermal-conductivity 2D or 3D networks to enhance the thermal conductivity of oxide pellets. Theoretically high porosity metal foams and nanowire networks can be used as the 3D network preforms in the powder processing of UO_2-based pellet fabrication. In this study, ZrO_2 which has a thermal conductivity as low as UO_2 is used for experiment, while chemical and microstructural evolution might be different. Since UO_2 pellets are fabricated at high temperatures above 1700°C, one of the selection criteria for the reinforcement network metal should be high temperature stability. Among the refractory metals, Mo has significantly high thermal conductivity (138 W/mK).

2. Experimental Procedures

Zirconium oxide powder and Molybdenum powder were purchased from Sigma - Aldrich. Mo wire mesh was purchased from Goodfellow (UK) and a microstructure image of the Mo wire mesh taken by SEM is presented in Fig. 1. The diameter of Mo wire was 70 μm and the aperture of the mesh was 440 μm. Mixing ZrO_2 with Mo mesh was conducted by conventional mixing in a graphite mold with a Mo fraction of 7 vol.% (10 wt%). In addition, Mo powder added ZrO_2 was prepared by conventional hand mixing with a Mo fraction of 10 vol.%(16 wt%).

To fabricate the composite pellets, spark plasma sintering (SPS) was used. Spark plasma sintering is very efficient to sinter composite pellets at high temperatures when compared with conventional sintering processes. Dr. SINTER LAB (Model: SPS-515S) was used for the SPS process. With uniform distribution of Mo mesh in ZrO_2 powder in a 13mm-diameter graphite mold, the axial pressure of 50 MPa was applied. The maximum sintering temperature was 1500°C and the samples are held for 10 minutes at 1500°C.

The sintered densities of composite pellets were measured using the Archimedes method and scanning electron microscopy (SEM) was used to observe the cross sections of polished composite pellets. The chemical interaction of ZrO_2 with Mo mesh and Mo powder was analyzed with energy dispersive X-ray spectroscopy (EDS). The phase formation of composite pellets was analyzed with X-ray diffractometry (XRD). The thermal conductivities of the composite pellets were obtained through measuring the thermal diffusivities, specific heat capacities, and thermal expansion coefficients at 1100°C. The laser flash method was used to measure the thermal diffusivities; the specific heat capacities of composite pellets were calculated using differential scanning calorimetry (DSC); and the densities of the sintered samples were calculated by using the thermal expansion coefficients and the density measured at room temperature.

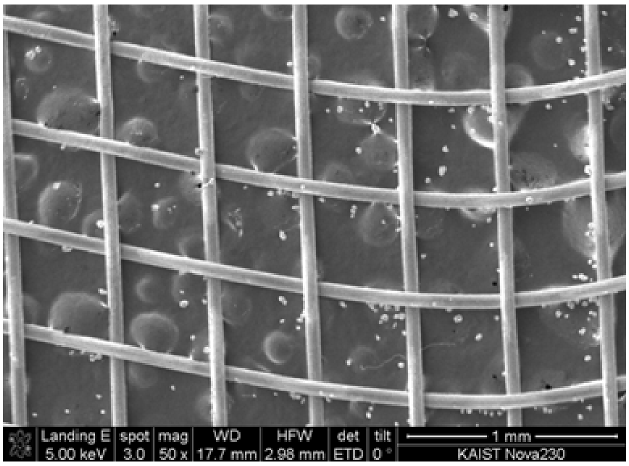

Fig. 1. An SEM image of Mo wire mesh

3. Results

According to the axial displacement of the SPS press punch, Mo-wired ZrO_2 and Mo-powdered ZrO_2 compacts began densification at approximately 970°C and the densification continued up to 1200°C. After 1200°C, thermal expansion dominated the axial displacement of the press punch up to 1500°C. Fig. 2 presents the fractured microstructure of the Mo-wired ZrO_2 composite pellet after sintering and Fig. 3 presents the XRD result of the Mo powder added ZrO_2 composite pellet after sintering. The spark plasma sintered Mo-wired ZrO_2 composite pellet is composed of grains with diameters of 20–30 μm as shown in the Figure 2 and the diffraction patterns of the Mo-powdered ZrO_2 composite pellet shows there was not severe chemical interaction between the two components.

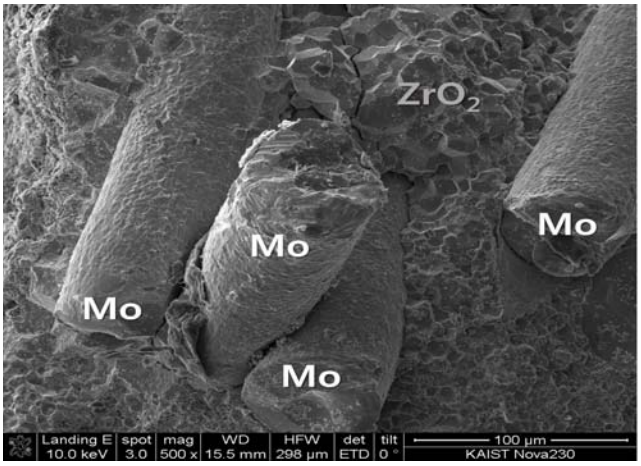

Fig. 2. An SEM image of fractured surface of a Mo wire reinforced ZrO_2 composite pellet

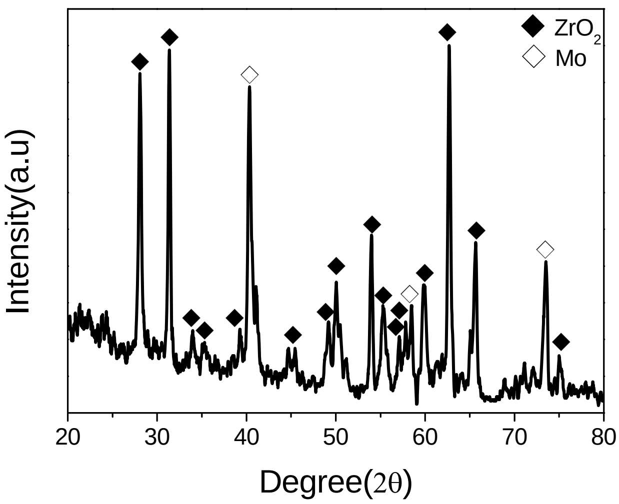

Fig. 3. An XRD pattern of a Mo powder added ZrO_2 composite pellet

The distribution of the Mo reinforcement is important to improve the thermal properties of the ZrO_2 pellets. Unlike the Mo-powder added ZrO_2 sample, the Mo wire is connected and uniformly distributed in the Mo wire-added ZrO_2 pellet as shown in in Fig. 2.

The thermal conductivity of the solid materials was obtained using the laser flash method that measures the thermal diffusivity of the materials. The measured thermal diffusivity was converted to thermal conductivity using the following equation:

$$k(T) = a(T)\cdot c_p(T)\cdot \rho(T) \qquad (1)$$

where k is the thermal conductivity, a is the thermal diffusivity measured using the laser flash method, c_p is the specific heat measured using differential scanning calorimetry, and ρ is the density at the measuring temperature.

The thermal conductivities of the ZrO_2 and Mo-wire added ZrO_2 and Mo-powder added composite pellets at 1100°C were 1.6 W/m-K, 2.5 W/m-K and 3.3 W/m-K respectively. The thermal conductivity of the Mo-wired ZrO_2 was increased by 56% and thermal conductivity of the Mo-powdered ZrO_2 increased by 106%.

Table 1. Thermal Conductivity of Composite Pellets

	Thermal conductivity (W/m-K)	Mo Volume % (%)
Pure	1.6	0
Mo Mesh	2.5	7
Mo Powder	3.3	10

To improve thermal conductivity of the Mo-wired ZrO_2 sample without increasing the volume percent Mo, the distribution of continuous reinforcement might be critical. Because the Mo-wired mesh sheets are layered vertically to the axis of the cylindrical ZrO_2 pellet without connected each other and the thermal conductivity of the Mo-wired ZrO_2 pellet measured in this study is the through-plane thermal conductivity, the Mo wire mesh might not affect thermal conductivity significantly. Therefore, fabrication of composite pellets with Mo wires parallel to the pellet axis will improve the thermal conductivity.

4. Conclusions

In order to increase the thermal conductivity of ZrO_2 pellets, spark plasma sintering of Mo wire added ZrO_2 and Mo powder added ZrO_2 was conducted. Although an enhanced thermal conductivity was measured for Mo-wire-added ZrO_2, more improvement might be attainable by adjusting the direction of the 2D reinforcement network or by formation of a 3D network.

REFERENCES

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