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Thermal Conductivity Measurement and Analysis of Fully Ceramic Microencapsulated fuel

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1. Introduction

Fully ceramic microencapsulated (FCM) fuel is recently proposed nuclear fuel concept for improving the accident tolerance of light water reactor (LWR)[1-2]. Nuclear fuel enhancing the accident tolerance is satisfied two parts. First, the performance has to be retained compared to the existing UO_2 nuclear fuel and zircaloy cladding system under the normal operation condition. Second, under the severe accident condition, the high temperature structural integrity has to be kept and the generation rate of hydrogen has to be reduced largely. FCM nuclear fuel is composed of tristructural isotropic(TRISO) fuel particle and SiC ceramic matrix. SiC ceramic matrix play an essential part in protecting fission product. In the FCM fuel concept, fission product is doubly protected by TRISO coating layer and SiC ceramic matrix in comparison with the current commercial UO_2 fuel system of LWR. In addition to a safety enhancement of FCM fuel, thermal conductivity of SiC ceramic matrix is better than that of UO_2 fuel. Because the centerline temperature of FCM fuel is lower than that of the current UO_2 fuel due to the difference of thermal conductivity of fuel, an operational release of fission products from the fuel can be reduced[1].

SiC ceramic has attracted for nuclear fuel application due to its high thermal conductivity properties with good radiation tolerant properties, a low neutron absorption cross-section and a high corrosion resistance[3-5]. Thermal conductivity of ceramic matrix composite depends on the thermal conductivity of each component and the morphology of reinforcement materials such as fibers and particles. There are many results about thermal conductivity of fiber-reinforced composite like as SiCf/SiC composite[6-8]. Various study have been reported to analyze the thermal conductivity of fiber-reinforced composite by analytical model taking into account effects of components of composite and morphology on their thermal conductivity[9,10]. However, the thermal conductivity of FCM fuel pellets and its analytical thermal conductivity model have not been reported yet.

In this study, we focused on the thermal conductivity of FCM pellets fabricated by hot pressing. Several thermal conductivity influence factor for FCM pellets were considered. The simple Maxwell's equation of the thermal conductivity for FCM fuel were discussed.

2. Methods and Results

TRISO particle deposited by chemical vapor deposition method were used to fabricate FCM pellets. Al_2O_3 and Y_2O_3 sintering additives was used to liquid phase sintering of pellet. Nano sized SiC powder was added in order to enhance the sintering of SiC matrix. The starting powders were ball-milled with ethanol. TRISO particles were overcoated with the mixed powder in order to place TRISO particles uniformly embedded in SiC matrix and prevent direct contact between TRISO particles. The weight ratio of TRISO particles and overcoating powder was 2:3. A spherical mixed body with about diameter of about 1.5 mm consists of the core of TRISO particle and outer mixed powder overcoating layer. Both of the mixed powder and the overcoated TRISO particles were inserted into a graphite sleeve. Hot press sintering was carried out under a pressure of 15 MPa at 1850 ^oC for 1 hr in an Ar atmosphere.

The density of the sintered body was measured by Archimedes method. The as-sintered specimens were observed by scanning electron microcopy after mirror-polishing and subsequent etching with CF_4 plasma containing 10% oxygen.

Thermal diffusivity (α) was measured by the laser flash method using a Xenon Flash instrument (LFA427 Nanoflash, Netzsch Instruments Inc., Burlington, USA). Differential scanning calorimetry (DSC 200, Netzsch Instruments Inc., Burlington, USA) was used for the heat capacity (C_p) measurement. Samples were cut and polished as 10 mm diameter and 2 mm thickness for measuring thermal diffusivity and 5 mm diameter and 2 mm thickness for measuring heat capacity. Thermal conductivity, k, was calculated using the following equation:

$$k = \alpha \cdot \rho \cdot C_p \quad (1)$$

where ρ is the density of the sample.

Polycrystalline SiC ceramics without TRISO coated particles were successfully fabricated at 1850 ^oC for 1 hr via liquid phase hot press sintering adding SiC nanopowder feedstock. Image of powder overcoating TRISO particles and microstructure of FCM pellets was shown in Fig. 1. SiC ceramics with different contents of sintering additives were successfully densified and show nearly no pores in microstructure investigation. Thermal conductivity of SiC ceramics with 3, 6, 9 wt.% (2.3, 4.6, 7.0 vol.%) sintering additives were 68.4, 71.3 and 74.1 W/mK, respectively. Thermal conductivity of SiC ceramics slightly increases with an increase in contents of sintering additives in spite of an increase in low thermally conductive oxide phase which is derived from sintering additives.

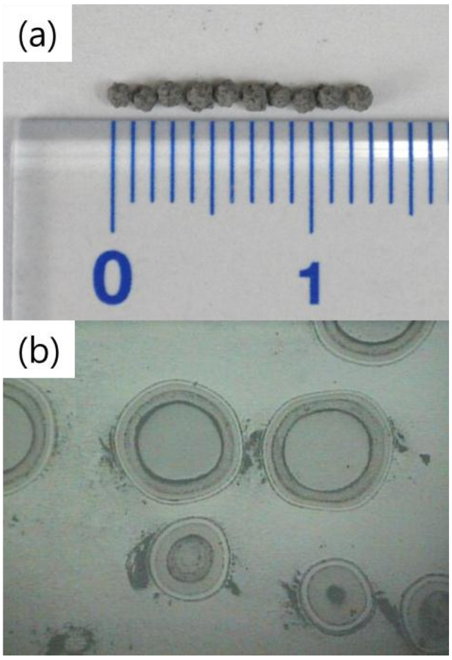

Fig. 1. (a) Image of powder overcoating TRISO particles, (b) Optical microscope image of FCM pellets.

FCM pellets which have various volume fraction of TRISO coated particles and different contents of sintering additives were prepared at same hot press sintering condition. Sintering condition, bulk density, thermal diffusivity, and thermal conductivity of SiC ceramics and FCM pellets were shown in Table 1.

Table 1. Bulk density and thermal properties of SiC ceramics and FCM pellets with different volume fraction of TRISO particles.

Amounts of sintering additives (wt.%), (vol.%)	V_f (%)	ρ_b (g/cm^3)	α (mm^2/s)	k (W/mK)
3, 2.3	0	3.17	32.2	68.4
6, 4.6	0	3.19	33.3	71.3
9, 7.0	0	3.22	34.2	74.1
3, 2.3	10	3.05	25.2	52.3
3, 2.3	20	3.00	23.6	46.8
3, 2.3	30	2.92	21.9	43.0
3, 2.3	40	2.82	18.5	34.5

In the FCM pellets, bulk density decreased with an increase in the volume fraction of TRISO particles. SEM micrographs of microstructure of FCM pellets with 3 wt.% (2.3 vol.%) sintering additives and different volume fraction of TRISO particles were shown in Fig. 2. With an increase in the volume fraction of TRISO particles, pores increased. Thermal conductivity of the FCM pellets with TRISO particles of 10 vol.%, 20 vol.%, 30 vol.% and 40 vol.% show 52.3, 46.8, 43.0 and 34.5 W/mK, respectively. The thermal conductivity of the FCM pellets decreased with an increase in the volume fraction of TRISO particles embedded in the sintered pellets. Thermal diffusivity also decreases considerably with an increase in the volume fraction of TRISO particles. According to increase the volume fraction of TRISO particles from 10 to 40%, the bulk density of FCM pellets decreased by 3.8, 5.4, 7.9, and 11.0%, respectively, and the thermal diffusivity of FCM pellets declined by 21.7, 26.7, 31.2, 42.6%, respectively. Therefore, the thermal conductivity reduction is caused by not decrease in the density of FCM pellets but the addition of low thermally conductive TRISO particles. A significant decrease in the thermal conductivity of the FCM pellet was observed with the addition of small quantity of 10 vol.% TRISO particles to SiC ceramic matrix. The FCM pellet with TRISO particles of 40 vol.% shows a large decrease in thermal conductivity and thermal diffusivity because of particularly low density and many pores, as it is shown in Fig. 2 (c)

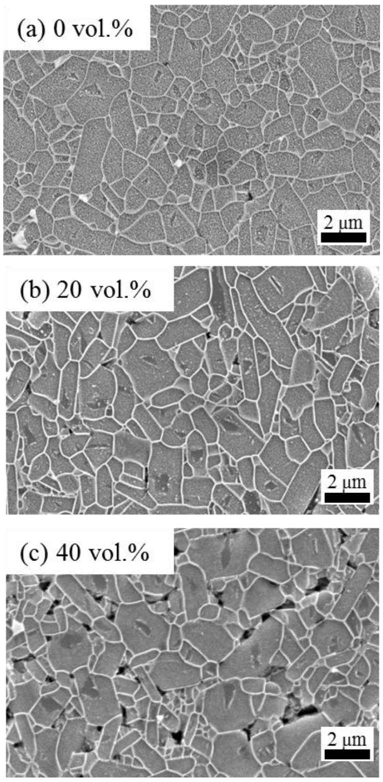

Fig. 2. SEM micrographs of etched surface of FCM pellets with TRISO particles of (a) 0 vol.%, (b) 20 vol.% (c) 40 vol.%

Based on the experimental results of the thermal conductivity and the microstructure of SiC ceramics and FCM pellets, the thermal conductivity of FCM pellets was discussed with simple Maxwell's equation. Maxwell's equation is one of the oldest, most famous equations that explain thermal conductivity of heterogeneous solids as materials made of sphere of k_0 embedded in a continuous phase of k_1. Maxwell's model assumes spheres do not interact thermally and it means that sphere is existed in matrix as small volume fraction. Maxwell's equation is following as

$$\frac{k}{k_1} = \frac{1+2\beta\phi}{1-\beta\phi}\ , \quad \kappa = \frac{k_0}{k_1}\ , \quad \beta = \frac{\kappa-1}{\kappa+2} \qquad (2)$$

where κ is the ratio of the thermal conductivity of the dispersed particles to the continuous phase used in effective conductivity model, β is the reduced polarizability from the analogy with potential theory, ϕ is the particle volume fraction.

Measured thermal conductivity of FCM pellets with different volume fraction of TRISO particles and analytical thermal conductivity models of FCM pellets were shown in Fig. 3. In the analytical models, thermal conductivity of TRISO particles was applied as 4.13 W/mK which was calculated from Maxwell's equation by Stains by et al. study about TRISO fuel for HTGR[11]. As the volume fraction of TRISO particles increased, the measured thermal conductivity values closely follow the prediction of Maxwell's equation. In the assumption of Maxwell's equation, there is no thermal interference between particles because the particles are far enough apart. In this study, a powder overcoating technique on TRISO particles was used in order to not only prevent direct contacts between the particles and but also maintain a certain distance between the particles. The thermal conductivity of the FCM pellets with high volume fraction of TRISO particles can be analogized by Maxwell's equation since TRISO particles maintain an enough distance by powder overcoating thickness. Low thermal conductivity of FCM pellets with low volume fraction of TRISO particles is thought to be due to an extremely non-uniform distribution or a locating to one side caused by experimental difficulties of TRISO particles arrangement.

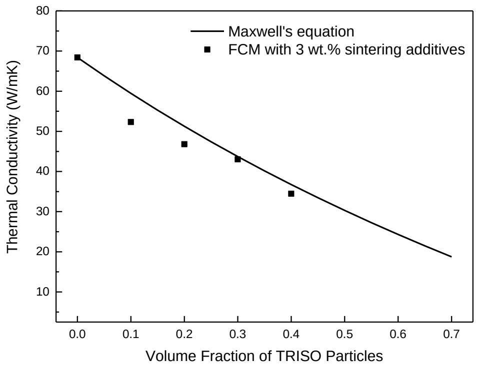

Fig. 3. Measured thermal conductivity of FCM pellets with different volume fraction of TRISO particles and the prediction of Maxwell's equation

3. Conclusions

Thermal conductivity of SiC ceramics and FCM pellets with the volume fraction of TRISO particles were measured and analyzed by analytical models. Polycrystalline SiC ceramics and FCM pellets with TRISO particles were fabricated by hot press sintering with sintering additives. Thermal conductivity of the FCM pellets with TRISO particles of 0 vol.%, 10 vol.%, 20 vol.%, 30 vol.% and 40 vol.% show 68.4, 52.3, 46.8, 43.0 and 34.5 W/mK, respectively. As the volume fraction of TRISO particles increased, the measured thermal conductivity values closely followed the prediction of Maxwell's equation. In high volume fraction of TRISO particles, the thermal conductivity of the FCM pellets could be explained by Maxwell's equation.

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