MICROSTRUCTURAL CONTROL OF TWO-DIMENSIONAL SIC FIBER-REINFORCED SIC COMPOSITES FOR IMPROVEMENT OF THEIR THERMAL CONDUCTIVITY

Katsumi Yoshida, Satoshi Kajikawa, Masamitsu Imai and Toyohiko Yano

Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, 2-12-1, Ookayama, Meguro-ku, Tokyo 152-8550, Japan

Introduction

Continuous SiC fiber-reinforced SiC composites (SiC_f/SiC) are expected to be used as components for gas turbine, spacecrafts and future fusion nuclear reactors. In the future fusion power reactor, the reactor concepts based on the use of SiC_f/SiC composite have been designed by JAEA, ARIES-team and CEA (TAURO). For these applications, higher thermal conductivity of the SiC_f/SiC composite is needed in addition to its higher mechanical strength and fracture toughness. Thermal conductivity of the SiC_f/SiC composite would depend on not only the thermal conductivity of each component such as SiC matrix and SiC fibers but also microstructure of the composite. Polycrystalline SiC fiber (TyrannoSA) cloth with high thermal conductivity has been commercially produced by Ube Industries, Japan. Present authors have improved the thermal conductivity of SiC matrix by the microstructural control using coarse SiC grains [1]. In addition, the authors have developed a novel process of SiC_f/SiC composite based on microstructural control using sheet stacking, hot-pressing and electrophoretic deposition methods [1-4]. In this study, we focused on microstructural control of the composite using our fabrication process and the simple model of thermal conductivity of composite, and improvement of its thermal conductivity was investigated.

Experimental

Submicron-sized α-SiC powder (average particle size; 0.40μm, Showa Denko, Japan) containing 20mass% of coarse α-SiC powder (average particle size; 2-3μm, Kojundo Chemical Laboratory, Japan) was used as the raw materials. Al_2O_3-Y_2O_3-CaO system was used as the sintering additives of SiC, and the amount of sintering additives was 20mass% in total. Green sheet of SiC with sintering additives was prepared by laboratory-scale tape casting equipment (DP-150, Tsugawa Seiki, Japan). Two-dimensionally plain-woven SiC fiber cloth (Tyranno SA, Ube Industries, Japan) was used as the reinforcement. Carbon coating on SiC fibers was formed by electrophoretic deposition (EPD) method reported in our previous paper [4]. SiC matrix between each filament was formed by EPD process using SiC powder suspension or polycarbosilane (PCS, Nippon Carbon, Japan) impregnation. These SiC cloths and SiC green sheets were stacked alternately, and then heat-treated at 300^{o}C in air. The compact was hot-pressed at 1750^{o}C for 1h in Ar flow under a uniaxial pressure of 40MPa. The composites using the SiC cloth treated by EPD and PCS were presented as EPD-composite and PCS-composite, respectively. For comparison, the composite was fabricated by hot-pressing using untreated TyrannoSA fiber cloth and SiC green sheet (Untreated composite). Thermal conductivity of the composite was measured perpendicular to the cloth layers at room temperature by laser-flash method. Microstructure of the composite was observed by scanning electron microscope (SEM).

Results and Discussion

Monolithic α-SiC containing 20mass% of coarse α-SiC grains and Al_2O_3-Y_2O_3-CaO sintering additives was hot-pressed under the same condition as the SiC_f/SiC composite described above, and its thermal conductivity was 54W/m•K at room temperature. Thermal conductivity of monolithic submicron-sized α-SiC ceramics was 47W/m•K. From this result, the addition of coarse α-SiC grains to submicron-sized α-SiC, i.e. microstructural control of SiC matrix, was effective to increase the thermal conductivity of SiC matrix.

Table 1 shows fiber volume fraction, bulk density, open porosity and thermal conductivity of the SiC_f/SiC composites fabricated in this study. The thermal conductivity of PCS-composite was 18W/m•K.

Table 1 Fiber volume fraction (V_f), bulk density (B.D.), open porosity (O.P.) and thermal conductivity (κ) of the SiC_f/SiC composites fabricated in this study.

Sample	V_f (vol%)	B.D. (g/cm^3)	O.P. (%)	κ (W/m•K)
(a) EPD-composite	41.9	3.12	1.43	44.5
(b) PCS-composite	38.4	2.97	1.06	18.3
(c) Untreated-composite	50.9	3.14	1.36	56.1

This value was higher than that of the composite fabricated in our previous study [1], but a significant increase in thermal conductivity could not be achieved. On the other hand, the thermal conductivity of EPD-composite and Untreated-composite was 45W/m•K and 56W/m · K, respectively, and these values were much higher than that of the composite reported in our previous paper and PCS-composite.

Figure 1(a) shows the schematic illustration of a rule of mixtures in multilayered structures. Thermal conductivity of the multilayer structures in the directions parallel (κ_{c1}, parallel model) and perpendicular (κ_{c2}, series model) to the layer, i.e. fiber cloth alignment, can be simply given by

$$\kappa_{c1} = \kappa_f V_f + \kappa_m V_m \quad (1)$$

$$1/\kappa_{c2} = V_f/\kappa_f + V_m/\kappa_m \quad (2)$$

where κ_m is the thermal conductivity of the matrix, κ_f the thermal conductivity of the matrix, V_m volume fraction of matrix, V_f volume fraction of fibers. Assuming that the κ_f of Tyranno SA is 60W/m•K and the κ_m is 30, 54 and 60W/m•K (54W/m•K and 60W/m•K is corresponding to the value of SiC matrix and the value of Tyranno SA fibers, respectively), the thermal conductivities of the composite calculated by Eq.(1) and Eq.(2) as a function of fiber volume fraction are shown in Fig.1(b). Untreated-composite had a multilayered structure consisting of polycrystalline SiC fiber cloths and SiC matrices derived from SiC sheet from the result of SEM observation, and its thermal conductivity well agreed with the ideal thermal conductivity calculated by series model using κ_m (54W/m•K) in Fig.1(b). In the case of PCS-composite, the SiC matrix formed in SiC fiber cloths was derived from PCS and this matrix shows lower thermal conductivity due to its low crystallinity. PCS-impregnated SiC fiber cloths would act as the layer with low thermal conductivity since the low thermal conductivity of PCS-derived SiC matrix would be dominant to that of the cloth layers. As a result, the thermal conductivity of PCS-composite would show the low thermal conductivity as shown in thermal conductivity of the composite using SiC matrix with low thermal conductivity calculated by series model. EPD-composite had a thermal conductivity of 45W/m•K, and this value was slightly lower than the value calculated by series model. This difference in thermal conductivity would be caused by the interfacial condition between SiC matrix and fibers. From these results, higher thermal conductivity of SiC_f/SiC composite could be successfully achieved by microstructural control and the thermal conductivity of untreated-composite in maximum by the optimization of EPD process for the fabrication of SiC_f/SiC composite.

Summary

We focused on microstructural control of the composite using our fabrication process and the simple model of thermal conductivity of composite, and improvement of its thermal conductivity was investigated. Untreated-composite had a multilayered structure consisting of polycrystalline SiC fiber cloths and SiC matrices and its thermal conductivity well agreed with the ideal thermal conductivity calculated by series model. In the case of PCS-composite, PCS-impregnated fiber cloths would act as the layer with low thermal conductivity since the low thermal conductivity of PCS-derived SiC matrix would be dominant to that of the cloth layers. The SiC_f/SiC composite fabricated by electrophoretic deposition and sheet stacking method showed high thermal conductivity of 45W/mK.

References

1. Yoshida, K., Imai, M., Yano, T., Mechanical and Thermal Properties of Hot-Pressed SiC/SiC Composite Using SiC Matrix Containing Coarse SiC Grains. *High Temperature Ceramic Matrix Composites*, (2001) 381-387.
2. Yoshida, K., Budiyanto, Imai, M., Yano, T., Processing and Microstructure of Silicon Carbide Fiber-Reinforced Silicon Carbide Composite by Hot-Pressing. *J. Nucl. Mater.*, **258-263** (1998) 1960-1965.
3. Yoshida, K., Imai, M., Yano, T., Improvement of Mechanical Properties of Hot-Pressed Silicon Carbide Fiber-Reinforced Silicon Carbide Composites by Polycarbosilane-Impregnation Method. *Compos. Sci. Technol.*, **61**(2001) 1323-1329.
4. Yoshida, K., Matsukawa, K., Yano, T., Microstructure and Mechanical Properties of Silicon Carbide Fiber-Reinforced Silicon Carbide Composite Fabricated by Electrophoretic Deposition and Hot-Pressing. *J. Nucl. Mater.*, **386-388** (2009) 643-646.

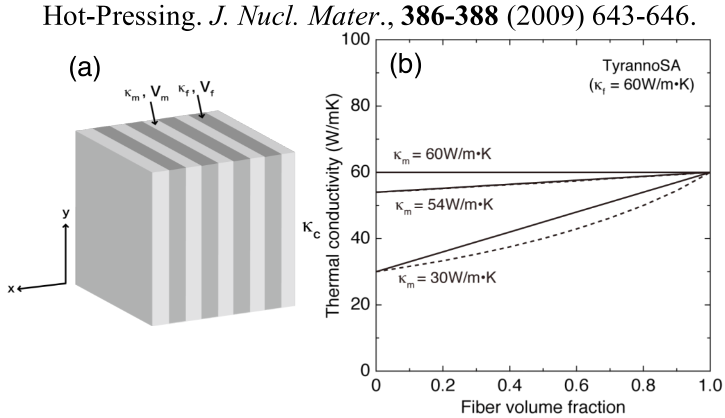

Fig.1(a) Schematic illustration of a rule of mixture in multilayered structures. (b)Relation between fiber volume fraction and thermal conductivity of the multilayered structure calculated by Eqs.(1)(Straight line) and (2) (broken line)