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Reaction sintering of two-dimensional silicon carbide fiber-reinforced silicon carbide composite by sheet stacking method

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Abstract

Two-dimensionally plain woven SiC fiber-reinforced SiC composite has been developed by reaction sintering using a sheet stacking method in order to further increase mechanical and thermal properties of the composite and to obtain flexibility of manufacturing process of 2D woven SiC/SiC composites which can be applied to the fabrication of larger parts. In addition, sinterability and mechanical properties of the composite were investigated. In this study, relative density of the composites was about 90–93% and a dense composite could be obtained by reaction sintering using the sheet stacking method. The bulk density and maximum bending strength of SiC/SiC composite with a C/SiC weight ratio of 0.6 were higher than that of the composite with C/SiC ratios of 0.5 or 0.7. The values were 2.9 g/cm^3 and 200 MPa, respectively. However, the composites obtained in this study fractured in almost brittle manner due to the lower fiber volume fraction.

1. Introduction

Continuous two-dimensionally (2D) woven SiC fiber-reinforced SiC matrix composite (SiC/SiC) is one of the most attractive materials for future fusion reactors since it has low induced radioactivity, quick decay of activity, low after heat, low atomic number, good fracture resistance, excellent high-temperature mechanical and thermal properties and corrosion resistance [1–6]. Furthermore, SiC shows good resistance to high energy neutron irradiation up to very high neutron fluences [7–11]. Future fusion power reactor concepts based on the use of SiC/SiC composite have been designed by JAERI (DREAM reactor) [12], ARIES team (ARIES-I, IV and AT) [13–15] and CEA (TAURO) [16].

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There are several processes to obtain SiC/SiC composites such as chemical vapor infiltration (CVI) and polymer infiltration and pyrolysis (PIP) techniques. These processes have some advantages such as a relatively low processing temperature and formation of relatively pure SiC matrix. However, these processes require a long manufacturing time and introduce about 10–20 vol.% of voids in the composites [4,17–19]. In order to simplify the fabrication process and to fabricate dense SiC/SiC composite with high mechanical and thermal properties, the authors have explored a new fabrication process using a sheet stacking method and hot-pressing [20–23]. Dense composites with non-brittle fracture behavior could be obtained by this process, and their bending strength and thermal conductivity at room temperature were 240 MPa and 14 W/mK, respectively.

On the basis of this process, we have paid attention to reaction sintering [24–26] which has an acceptable processing temperature and the possibility of densification and near-net shape formation of components with complex shape. Reaction sintering was used in the fabrication of 2D woven SiC/SiC composite using a sheet stacking method in order to further increase mechanical and thermal properties of the composite and to obtain flexibility of the manufacturing process of 2D woven SiC/SiC composites for the fabrication of larger parts. In this study, a 2D woven SiC/SiC composite was produced by reaction sintering using a sheet stacking method, and the mechanical properties were investigated.

2. Experimental procedures

2.1. Fabrication of green sheet

Powder of submicron β-SiC (Ultrafine, average particle size: 0.32 μm, Ibiden, Japan), carbon black (average particle size: 80 nm, Asahi carbon, Japan) and a dispersing agent (SN-7347C, 20%-quaternary ammonium salt solution, San Nopco, Japan) were mixed by ball-milling for 24 h in ethanol with SiC balls (15 mm diameter). The weight ratio of C/SiC was 0.5–0.7. Polyvinyl butyral (PVB, Sekisui Chemical, Japan), dioctyl adipate (Wako Pure Chemical Industries, Japan), an organic binder and a plasticizer, were dissolved in ethanol at 40 °C for 24 h. A well-stirred solution of binder/plasticizer system was added to the above solution. This solution was further mixed by ball-milling for 3 h, followed by de-airing using a rotary pump for 30 min to obtain the final slurry for tape casting. The composition of raw materials and organics in slurry for is listed in Table 1. The green sheet was prepared using laboratory-scale tape casting equipment (DP-150, Tsugawa Seiki, Japan). The sheet was dried at room temperature and cut into pieces of 35 mm × 35 mm with a thickness of 500–700 μm.

Table 1
Composition of C, SiC powder and organics in the green sheet

	C/SiC weight ratio (mass, %)		
	0.5	0.6	0.7
C	33.3	37.5	41.2
β-SiC	66.7	62.5	58.8
Subtotal	100	100	100
Dispersing agent*	4	4	4
Binder*	16	16	16
Plasticizer*	11	11	11

* Amount of dispersing agent, binder and plasticizer added to above powders.

2.2. Fabrication of the SiC/SiC composite by reaction sintering

Two-dimensionally (0°/90°) plain woven BN-coated Hi-Nicalon (Nippon Carbon, Japan) fiber cloth was used as the reinforcement. The thickness of BN-coating was about 0.4 μm. In this study, we used BN-coating on fiber due to the effectiveness for the inhibition of the reaction between the SiC matrix and fiber although BN-coating is not suitable for fusion environment. However, it is considered that this process can be applied on other coatings such as carbon.

The cloth was cut into 35 mm × 35 mm squares, followed by desizing with hot water (80 °C). The cloth was impregnated with the slurry obtained by diluting the slurry for tape casting with ethanol for 30 min. After impregnation, the cloth was dried at 60 °C for 1 h. This impregnation-drying process was repeated three times. The slurry for tape casting was applied to the green sheets and Hi-Nicalon fiber cloth, and they were stacked alternately. The green compact was dried at 60 °C under a uniaxial pressure of 34 kPa, and then heat-treated at 350 °C in air for 36 h under a uniaxial pressure of 20 kPa. The compact was set in a graphite crucible with 95 mass% of silicon (<200mesh, Hirano Seizaemon Co. Ltd., Japan) and 5 mass% of boron (Hirano

Seizaemon Co. Ltd., Japan). Afterwards reaction sintering was performed at 1450 °C for 2 h in vacuum. Fiber volume fraction of the composite was 13–15%.

2.3. Characterization

Reaction-sintered specimens were cut into rectangular bars (width: 4.0 mm, thickness: 3.0 mm, length: 34 mm). Bulk density was measured by Archimedes' method using water. Constituent crystalline phases of SiC/SiC composite were determined by X-ray diffractometry (XRD, PW-1700, Philips).

Three-point bending strength was measured at room temperature with a crosshead speed of 0.5 mm/min and a span of 30 mm. Bending strength measurement was performed using a universal testing machine (Instron 1185, USA). Fracture surface of SiC/SiC composite after bending test was observed by field emission scanning electron microscope (FE-SEM, S-4800, Hitachi, Japan).

3. Results and discussion

X-ray diffraction patterns of SiC/SiC composite fabricated by reaction sintering at 1450 °C are shown in Fig. 1. The composites obtained in this study mainly consisted of β-SiC. However, a small amount of silicon that did not react with carbon remained in the composites.

Fig. 2 shows the bulk density of SiC/SiC composites with the various weight ratio of C/SiC fabricated by reaction sintering at 1450 °C. For

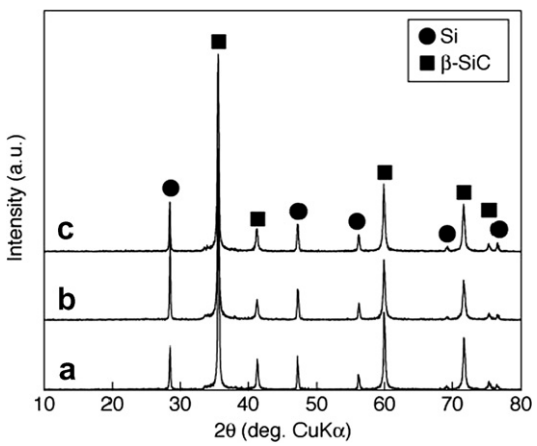

Fig. 1. X-ray diffraction patterns of SiC/SiC composite fabricated by reaction sintering at 1450 °C for 2 h in vacuum. The weight ratio of C/SiC is (a) 0.5, (b) 0.6 and (c) 0.7.

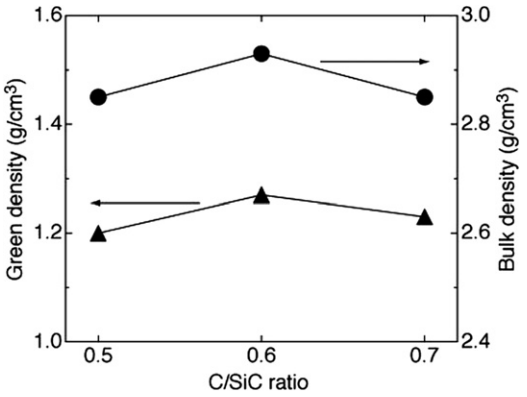

Fig. 2. Bulk density of SiC/SiC composite with the various weight ratio of C/SiC fabricated by reaction sintering at 1450 °C for 2 h in vacuum. Composite in green density of the composite was also indicated in this figure.

comparison, the green density of SiC/SiC composite is also shown in Fig. 2. The green density of the composites was 1.20–1.27 g/cm^3, and the composite with C/SiC weight ratio of 0.6 showed higher green density compared to the composite with C/SiC weight ratio of 0.5 or 0.7. The difference in bulk density of the composites corresponded to the difference in green density. The values were 2.85–2.93 g/cm^3. Based on the fiber volume fraction of the composite (13–15%) and the theoretical density of Hi-Nicalon fiber (2.74 g/cm^3) and SiC matrix (3.21 g/cm^3), the theoretical density of SiC/SiC composite fabricated in this study can be estimated to be approximately 3.15 g/cm^3. In this study, the relative density of the composites was about 90–93% and dense composite were obtained.

The maximum bending strength of SiC/SiC composites with the various weight ratio of C/SiC fabricated by reaction-sintering at 1450 °C is shown in Fig. 3. In this paper, bending strength was calculated from the maximum load for fracture in a load–displacement curve and we defined the value as maximum bending strength. The difference in maximum bending strength corresponds to the difference in bulk density with values of 110–190 MPa. The composite with a C/SiC weight ratio of 0.6 showed a higher maximum bending strength than the composites with a C/SiC weight ratio of 0.5 or 0.7. From the results of bulk density and maximum bending strength, it was found that the optimum C/SiC weight ratio for the composite was 0.6.

Typical load–displacement curves for SiC/SiC composite with the various C/SiC weight ratios from three-point bending tests at room temperature

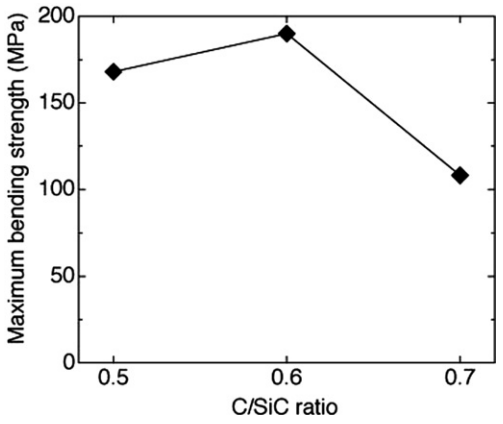

Fig. 3. Maximum bending strength of SiC/SiC composite with the various weight ratio of C/SiC fabricated by reaction sintering at 1450 °C for 2 h in vacuum.

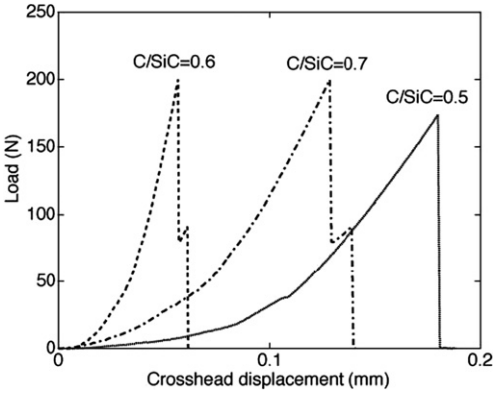

Fig. 4. Typical load–displacement curves of SiC/SiC composite with the various weight ratio of C/SiC in three-point bending test at room temperature.

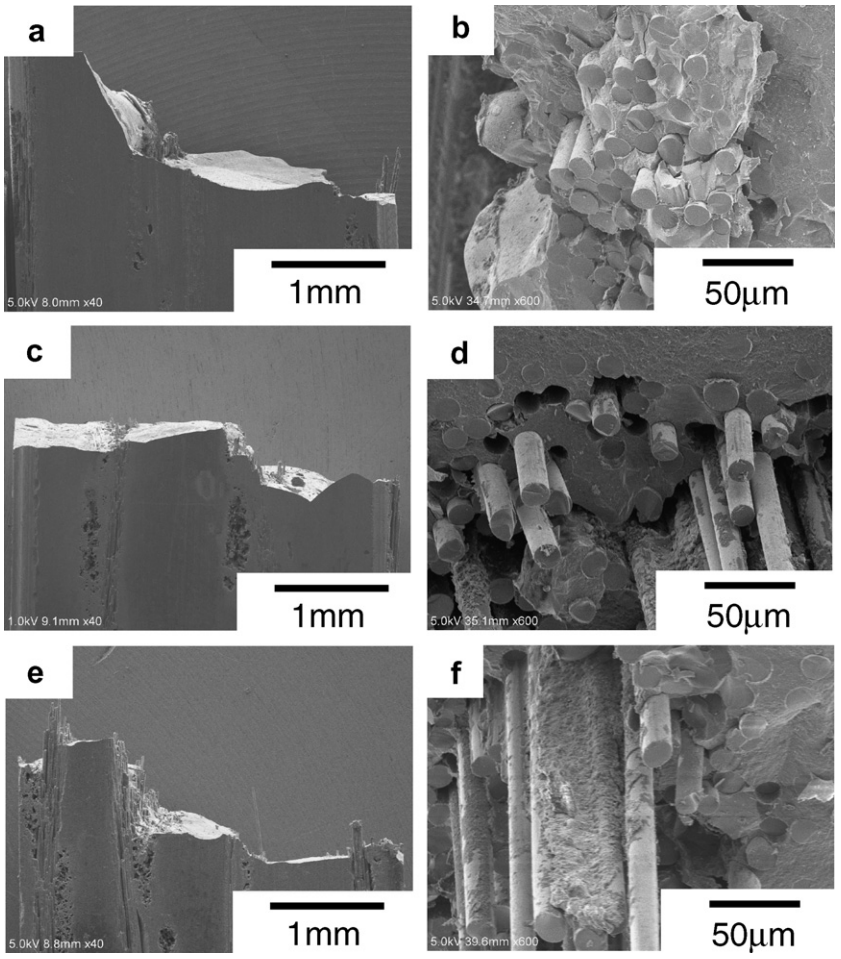

Fig. 5. SEM micrographs of fracture surface of SiC/SiC composite with the various weight ratio of C/SiC after three-point bending test at room temperature. The weight ratio of C/SiC is (a), (b) 0.5, (c), (d) 0.6 and (e), (f) 0.7.

are shown in Fig. 4. SEM micrographs of the corresponding fracture surfaces after the three-point bending test are shown in Fig. 5. Good SiC matrix deposition occurred between each fiber and a small amount of fiber pullout could be observed. However, the composite obtained in our study fractured in almost brittle manner. The low fiber volume fraction in the composite causes the brittle fracture behavior. When the load is applied to the thick matrix and the matrix elastically deforms and fractures during the bending test, the fiber cannot sustain the load when elastic energy is released and the fiber fractures with limited pull-out, resulting in brittle behavior.

4. Summary

Two-dimensionally plain woven SiC fiber-reinforced SiC composite was produced by reaction sintering using a sheet stacking method. The sinterability and mechanical properties of the composite were investigated. Dense composite, which is 90–93% of theoretical density, could be obtained by reaction sintering using a sheet stacking method. The bulk density and maximum bending strength of SiC/SiC composite with a C/SiC weight ratio of 0.6 was higher than that of the composite with C/SiC weight ratios of 0.5 or 0.7. The values were 2.9 g/cm^3 and 200 MPa, respectively. However, the composites obtained in this study fractured in almost brittle manner, attributed to the lower fiber volume fraction in the composite. It appears that the present fabrication process has the possibility for use in fabricating homogeneous and dense composites with good mechanical and thermal properties.

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