Effect of the Processing Parameters on the Densification and Strength of 2D SiC Fiber-SiC Matrix Composites Fabricated by Slurry Infiltration and Stacking Process

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ABSTRACT

2D SiC fiber-SiC (SiC/SiC) composites were fabricated via slurry infiltration and a stacking process. The effects of the additive composition and content in SiC slurries and the effect of the sintering time on the sintered density and strength of SiC/SiC composites were investigated. A slurry containing Al_2O_3-Y_2O_3-MgO (AYM) additives led to a higher strength compared to a slurry containing Al_2O_3-Y_2O_3-CaO (AYC) additives. The sintered density increased as the sintering time increased and showed a maximum (>98%) at 4 h. In contrast, the flexural strength increased as the sintering time increased and showed a maximum (615 MPa) at 6 h. The relative density and flexural strength increased as the additive content increased.

Key words : Fiber composites, Silicon carbide, Densification, Strength, Processing parameter

1. Introduction

S iC fiber-reinforced SiC composites (SiC/SiC composites) are potential candidates in various application fields where the service conditions are extremely severe including gas turbines, space shuttles, and nuclear fusion reactors.[1-4] The latter application deals with the fabrication of the inner walls of the toroidal chamber facing the plasma, extracting the heat generated by fusion reactions and transferring that heat to the coolant. The main advantages of SiC in this area are its high melting point, and excellent mechanical and thermal properties at high temperatures. The best SiC/SiC composites in this field are those that are as dense as possible and as pure as possible in compositions with an interface stable under radiation.[4,5]

Several processing routes for SiC/SiC composites have been suggested including chemical vapor infiltration (CVI),[6] polymer impregnation and pyrolysis (PIP),[7] reaction sintering (RS),[8] slurry impregnation and hot pressing (nano-infiltrated transient eutectoid process, NITE),[9] and hybrid (or combined) processes.[10,11] In terms of densification, the CVI and PIP processes yield composites with significant residual porosity levels. The RS process retains free Si in the matrix, and the Si limits its refractoriness and creep resistance. The NITE process produces a relatively dense matrix, but extremely expensive nano-sized SiC powders must be used for the impregnation and enhanced densification.[9] Recently, a new process based on stacking SiC fiber fabric and SiC tapes alternately for fabricating SiC/SiC composites was developed.[12] The process involves (i) stacking SiC fiber fabric and SiC tapes alternately at room temperature, (ii) pyrolysis of the stacked composites, and (iii) hot-pressing the pyrolyzed composites. By controlling the hot-pressing temperature, it was possible to obtain fully dense 2D SiC/SiC composites with relative densities of >98%.

In the present study, the effect of the processing parameters on the densification and strength of 2D SiC fiber-SiC matrix composites fabricated by slurry infiltration and a stacking process was investigated.

2. Experimental Procedure

2D woven Tyranno SA fiber fabrics (SiC fiber fabrics, Ube Industries Ltd., Japan) were used as the reinforcement in the fabrication of SiC/SiC composites. A SiC tape with a thickness of 180 μm and a width of 150 mm was prepared via tape casting. The details of the tape casting were described in a previous paper.[12]

Powders of β-SiC (~0.3 μm, Betarundum, Ibiden Co. Ltd., Ogaki, Japan) with various compositions of additives (see Table 1) were mixed for 24 h by ball milling. A binder of 3 wt% polyvinyl alcohol was added into the slurry. The fiber fabric was infiltrated in the slurries and dried at RT and at 100°C in a dry oven. The fiber fabrics and the SiC tapes

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Table 1. Batch Composition of Slurries for Fabricating SiC Fiber-Reinforced SiC Composites

Sample designation	Slurry composition (wt%)				
	SiC[†]	Al₂O₃[‡]	Y₂O₃[i]	MgO[§]	CaO[*]
10AYC-1	90	7	2		1
5AYM-1	95	3.2	1.3	0.5	
10AYM-1	90	6.4	2.6	1	
20AYM-1	80	12.8	5.2	2	
10AYM-2	90	6.4	2.6	1	
10AYM-4	90	6.4	2.6	1	
10AYM-6	90	6.4	2.6	1	

[†] ~0.34 μm Ultrafine, Lbiden Co., Ltd, Nagoya, Japan.
[‡] AKP30, Sumitomo Chemical Co., Tokyo, Japan.
[i] Grade fine, H.C. Starck, Germany.
[§] 99.9%, High Purity Chemicals, Japan.
[*] 99% up, High Purity Chemicals, Japan.

were alternately stacked at room temperature under a pressure of 10 MPa. The stacked sheets were placed in a BN-coated graphite mold and then calcined at 800°C for 1 h under a flow of argon to remove the organic components. The calcined compact was hot-pressed at 1750°C for 1-6 h under a pressure of 15 MPa in an argon atmosphere.

The density of each sample was measured using the Archimedes method. Bar-shaped specimens were cut to a size of 3 mm × 2.5 mm × 25 mm for strength measurements. Bend tests were performed at room temperature on four to six specimens for each condition using a four-point method with inner and outer spans of 10 and 20 mm, respectively. Fracture surfaces were observed by scanning electron microscopy (SEM).

3. Results and Discussion

The processing strategy for the fabrication of the 2D SiC/SiC composites involves: (i) infiltration of the SiC fiber fabric using a SiC slurry, (ii) stacking the slurry-infiltrated SiC fiber fabric and SiC tapes alternately at room temperature, (iii) pyrolysis of the stacked composites, and (iv) hot-pressing the pyrolyzed composites.

Fig. 1 shows the microstructure of fracture surfaces for composites infiltrated with SiC slurries containing different additive compositions. The sintered densities of 10AYC-1 and 10AYM-1 (refer to Table 1) were 3.07 g/cm³ and 3.03 g/cm³, respectively. The first and the last numbers in the sample designation denote additive contents in the SiC slurry and the number of hours at the hot-pressing temperature, respectively. Under the present fabrication conditions, the distinction between SiC fiber bundle and SiC matrix was very clear in the composites. Typically, no large pores or defects were observed at the interfaces between the SiC fiber fabric and the SiC tape in both specimens. As in the case for the densification of monolithic SiC,[13] the matrix layers were well sintered. However, micropores existed in
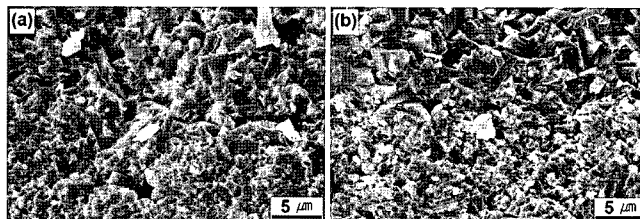

Fig. 1. Typical fracture surfaces of 2D SiC fiber-SiC composites sintered with different slurry compositions: (a) 10AYC-1 and (b) 10AYM-1 (refer to Table 1).

the inter-fiber areas (see Fig. 3(a)), as the spherical morphology of the fibers was maintained at the sintering temperature. It appears that the infiltration of SiC slurries into the inter-fiber region was not sufficient to fill the inter-fiber space. Fig. 1 also shows that the grain size of SiC grains (1~5 μm) in the matrix was larger than that (0.2~1 μm) in the fibers. The slower grain growth rate of SiC grains in the fibers might have resulted from the lack of the liquid-phase in the fibers at the hot-pressing temperature. There were no remarkable differences between the two specimens in terms of the microstructure and sintered density. However, the strength (490±32 MPa) of 10AYM-1 was higher than that (368±22 MPa) of 10AYC-1. This difference is attributed to the chemistry effect of the SiC matrix. This type of chemistry effect in liquid-phase sintered SiC ceramics has been reported by a number of researchers.[14,15] As the AYM additive system resulted in higher strength compared to the AYC additive system, further experiments were carried out using the AYM additive system.

Fig. 2 shows typical fracture surfaces of 2D SiC fiber-SiC composites sintered at 1750°C as a function of the sintering time. The fiber morphology changed from spherical (Fig. 2(a)) in the cross-section to a matrix-like morphology (Fig. 2(d)) as the sintering time increased due to the densification of the inter-fiber spaces at 1750°C under an applied pressure of 15 MPa and the grain growth of the SiC grains in
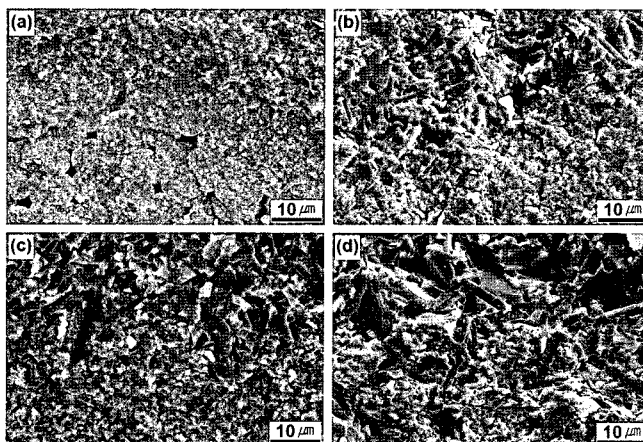

Fig. 2. Effect of the sintering time on the fracture surfaces of 2D SiC fiber-SiC composites infiltrated with 10AYM slurry and sintered at 1750°C: (a) 1 h, (b) 2 h, (c) 4 h, and (d) 6 h (refer to Table 1).
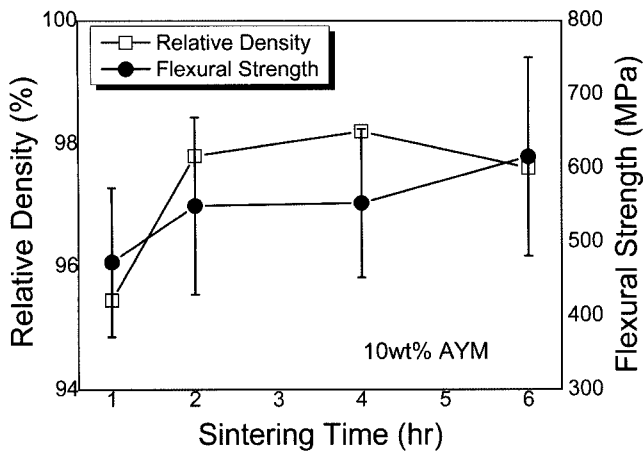
Fig. 3. Effect of the sintering temperature on the sintered density and strength of 2D SiC fiber-SiC composites.

Fig. 4. Effect of the additive content in the slurry on the fracture surfaces of 2D SiC fiber-SiC composites sintered at 1750°C for 1 h: (a) 5AYM-1, (b) 10AYM-1, and (c) 20AYM-1 (refer to Table 1).

the fibers. Elongated grains were observed in the matrix after sintering for two hours and in the fiber after sintering for six hours. The occurrence of the $\beta \rightarrow \alpha$ phase transformation of SiC was responsible for the growth of the elongated grains.[16,17] These results suggest that the occurrence of the phase transformation was activated in the matrix due to the presence of a liquid phase. The grain size of the SiC grains in the matrix increased from $1 \sim 5$ µm to $8 \sim 18$ µm as the sintering time increased from 1 h to 6 h. The grain size of the SiC grains in the fibers increased from $0.2 \sim 1$ µm to $2 \sim 10$ µm as the sintering time increased from 1 h to 6 h. The effect of the sintering time on the density and flexural strength of the 2D SiC/SiC composites is shown in Fig. 3. The sintered density increased as the sintering time increased and showed a maximum ($\sim 98.2\%$) at 4 h. A slight decrease in the density after sintering for six hours (97.6%) can be attributed to the weight loss that is frequently observed in oxide additive systems after prolonged sintering.[18-20] The flexural strength increased as the sintering time increased and showed a maximum (615 ± 135 MPa) at 6 h. The increase in strength after prolonging the sintering time was due to the increased densification in the 2-h-sintered specimen and the growth of the elongated grains to moderate sizes ($5 - 20$ µm) in both the matrix and fibers (see Fig. 2(d)) in the 6-h-sintered specimen. The growth of the elongated grains with a moderate size in both the matrix and fibers generally led to increased strength and toughness in the SiC matrix composites.[17,21]

Fig. 4 shows the effect of the additive content in the slurry on the fracture surface of 2D SiC fiber-SiC composites sintered at 1750°C for 1 h. The fiber morphology changed from spherical (Fig. 4(a)) in the cross-section to a matrix-like morphology (Fig. 4(c)) as the additive content increased due to the enhanced densification of the inter-fiber spaces at 1750°C under an applied pressure of 15 MPa. Although the additive content in the SiC slurries changed, the grain size and morphology did not change significantly. The densification process for the composites can be divided into two main
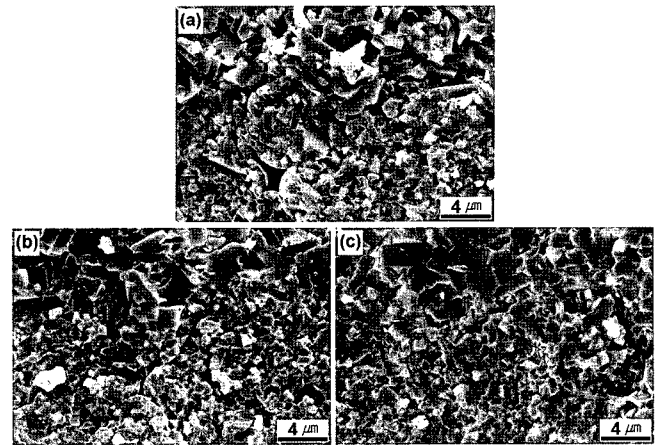
parts: matrix densification and inter-fiber densification. Densification of the matrix is similar to that for monolithic SiC and is a temperature-dependent process, according to a previous paper.[12] The inter-fiber densification is controlled by the deformation of fibers during the hot-pressing process when the amount of liquid is insufficient to fill the inter-fiber spaces. The present result suggests that a larger addition of additive into SiC slurries enhances the densification of inter-fiber spaces without causing the deformation of fibers, resulting in a higher sintered density (Fig. 5). Thus, increasing the additive content in SiC slurries is an efficient technique to enhance the densification of 2D SiC/SiC composites without changing both the morphology of fibers and the microstructure in matrix. Generally, a larger amount of liquid-forming additive leads to changes in the microstructure,[22] but 1-h annealing at 1750°C was insufficient to trigger this type of microstructure change in the specimens.

As shown in Fig. 5, both the sintered density and flexural strength increased as the additive content increased in the
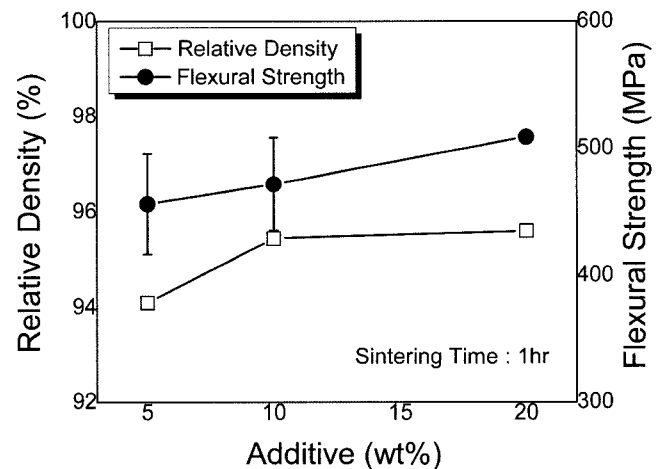

Fig. 5. Effect of the additive content on the sintered density and strength of 2D SiC fiber-SiC composites.

SiC slurries. The strength increased from 456 MPa to 509 MPa as the additive content increased from 5% to 20%. The increase in strength is attributed to the increased density of the composites. A flexural strength of ~280 MPa was reported in a 2D SiC fiber-SiC composite fabricated by a CVI process when no pyrolytic carbon coating was applied.[6] The sintered density of the composite was 2.78 g/cm^3. Thus, the superior strength of the 1750°C-sintered composite fabricated by the present process is attributed to the higher sintered density (3.03-3.18 g/cm^3) and the absence of the degradation of the fibers. A modification of the fiber-matrix interface, such as an application of pyrolytic carbon coating on the fibers, would improve the mechanical properties further, as was found in a study involving composites fabricated by the CVI process.[6]

4. Summary

Dense 2D SiC/SiC composites with sintered densities greater than 3.03 g/cm^3 were fabricated by slurry infiltration and a stacking process. The AYM additive system was more effective than an AYC additive system in terms of increasing the strength of 2D SiC/SiC composites. The sintered density of 2D SiC/SiC composites increased as the sintering time increased at 1750°C and showed a maximum (3.18 g/cm^3) at 4 h. The flexural strength increased as the sintering time increased and showed a maximum (615±135 MPa) at 6 h. The increase in strength after prolonging the sintering time was due to the increased densification and the development of a toughened microstructure in both the matrix and fibers. A larger addition of additive into SiC slurries enhanced the densification of the inter-fiber spaces and did not deform the fibers, resulting in a higher sintered density.

Acknowledgement

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