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Effect of Applied Pressure During Sintering on the Densification and Mechanical Properties of SiC_f/SiC Composites Prepared by Electrophoretic Infiltration

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Abstract

SiC-fiber-reinforced SiC matrix ceramic (SiC_f/SiC) composites were successfully fabricated by electrophoretic infiltration and sintering at various applied pressures. The effect of applied pressure (i.e., 5, 10, and 20 MPa) was thoroughly examined, and applied pressure appears to influence the densification and mechanical properties of SiC_f/SiC composites. The densities of SiC_f/SiC composites prepared at applied pressure of 5, 10, and 20 MPa were 2.99, 3.10, and 3.16 g/cm^3, respectively. All samples showed dense microstructures in their matrix areas; however, many closed pores were found with increasing pressure. Pressure induced densification in the fiber area, and 20 MPa was determined to be the minimum pressure required for adequate densification. The liquid phase in the matrix area was effectively maintained at a high applied pressure, which resulted in densification. However, the liquid phase redistributed to the matrix or near the matrix area at applied pressures of <20 MPa, leading to pores in the fiber areas of these samples. The highest flexural strength of 262 ± 18 MPa for the SiC_f/SiC composite was achieved at an applied pressure of 20 MPa. Meanwhile, the flexural strengths of the composites prepared at 5 and 10 MPa were 198 ± 41 and 238 ± 32 MPa, respectively.

Keywords: applied pressure, electrophoretic infiltration, SiC_f/SiC composite, sintering

Introduction

High-temperature applications of SiC-fiber-reinforced SiC matrix ceramic (SiC_f/SiC) composites require these composites to be dense because the mechanical properties of these composites improve with density. Moreover, SiC fibers need to maintain their original shapes to prevent damage to the fibers.

Several well-known methods have been used to fabricate SiC_f/SiC composites. High-purity SiC_f/SiC composites are obtained by chemical vapor infiltration (CVI), which is an ideal method for the fabrication of these composites [1–3]. The precursors used in this method are gaseous; therefore, extended processing times are required to achieve porosities of approximately 10–20%. Polymer impregnation and pyrolysis (PIP) is another method for the fabrication of SiC_f/SiC composites [4–6]. In contrast to CVI, this method uses polymer precursors instead of gases; however, it requires multiple impregnations to obtain the desired density. To utilize the advantages of these two methods, Ortona *et al*. reported a combined CVI/PIP method [7]. Another approach for the fabrication of SiC_f/SiC composites involves slurry infiltration combined with sintering [8–9], and electrophoretic infiltration is one such method [10–16].

Electrophoretic infiltration was initially used to deposit a coating layer on a substrate [17–18]. Novak *et al*. investigated electrophoretic infiltration as a possible infiltration method for the production of SiC_f/SiC composites [10] and determined that submicron SiC powder sufficiently infiltrates SiC fibers compared with nanosized SiC powder. In another report, Novak *et al*. discovered that the zeta potential is the most important parameter for electrophoretic infiltration and dense deposits can be obtained from suspensions with high zeta potentials [11]. A method that combined electrophoretic infiltration and PIP was used to prepare high-density SiC_f/SiC composites [12], and a SiC_f/SiC composite with a relative density of 86.5% was obtained after six PIP cycles. Gil and Yoon reported that SiC_f/SiC composites prepared by electrophoretic infiltration exhibited the highest density [14], wherein a relative density of 99.5% was achieved by combined

electrophoretic infiltration and sintering. These authors used nanosized SiC powder to infiltrate SiC fibers and ultrasonication to minimize the sealing effect on the surfaces of SiC fabrics. The sintering conditions applied to electrophoretically infiltrated SiC fabrics need to be considered carefully because high temperatures can damage the fibers. Moreover, the pressure applied during sintering needs to be minimized to prevent fiber damage. To date, 20 MPa has been used as the applied pressure during hot pressing of SiC_f/SiC composites [14–16]; however, to the best of our knowledge, no studies that examine the effect of external pressure during sintering of SiC_f/SiC composites prepared by electrophoretic infiltration have been reported.

This study aimed to examine the effect of external pressure on the densification and mechanical properties of a SiC_f/SiC composite prepared by electrophoretic infiltration. The density, microstructural, and mechanical property data were analyzed to elucidate the effect of external pressure on the SiC_f/SiC composite.

Materials and Methods

Commercial β-SiC powder (>97.5% purity; 4620KE, NanoAmor Inc., USA) with average particle size and specific surface area of 52 nm and 80 m^2/g, respectively, was used as the matrix phase. The SiC powder particles contain thin SiO_2 layers because of the reaction between SiC and O_2 in the air [19]. To decrease the sintering temperature and increase the density of SiC, 12 wt% of sintering additives (i.e., Al_2O_3, D_m = 150 nm, >99.9% purity [Baikowski, Japan] and Y_2O_3, D_m = 200 nm, >99.9% purity [Acros Organics, USA] in a 60:40 mass ratio) was added to the commercial β-SiC powder. The mixed powder was dispersed in ethanol and subjected to ball milling for 24 h using 5-mm SiC balls to minimize contamination. Prior to ball milling, the mixed-powder dispersion was enhanced by the addition of 3 wt% of Hypermer KD1 as the dispersant. Moreover, the zeta potential of the mixed powder was controlled at >40 mV by setting the pH to 3. The β-SiC slurry containing the sintering additives was used to electrophoretically infiltrate a woven Tyranno SA3 fabric (Ube Industries Ltd., Japan) coated with 400-nm-thick pyrolytic carbon (PyC). The distance between fabric and electrode in the dual electrophoretic infiltration system was 20 mm. The applied voltage and electrophoretic infiltration time were 10 V and 30 min, respectively. Ultrasonication (HD 2070, Bandelin, Germany) was applied over the first 20 min to minimize the surface sealing effect. After the infiltrated fabric was dried, 15 layers of fabric were laminated under an applied pressure of 10 MPa at 80 °C. Binder burnout was conducted at 350 °C for 2 h in the air at a heating rate of 0.5 °C/min to remove organic matter from the infiltrated fabric. Sintering was performed in a hot-pressing furnace at 1,750 °C for 1 h under Ar. Three different applied pressures were used (i.e., 5, 10, and 20 MPa) during sintering.

The densities of the SiC_f/SiC composites were measured using the Archimedes principle, with distilled water as the medium. Scanning electron microscopy (SEM; S-4800, Hitachi, Japan) was used to analyze the microstructures of the sintered SiC_f/SiC composites. Thermal etching was performed at 1,500 °C for 30 min under vacuum after each sample had been polished. The average grain size of each sample (i.e., 100 grains) was estimated from the SEM image after thermal etching and analyzed statistically. Three-point bend testing was performed using an ultimate testing machine (UTM AG-50E, Shimadzu, Japan). Each composite was cut into a 4 mm × 2 mm × 20 mm piece and polished using a 1-μm diamond paste to remove scratches from the sample before bend testing.

Results and Discussion

Figure 1 shows the images of the SiC fabric after electrophoretic infiltration. Figure 1(a) shows that the SiC fabric is fully covered by SiC powder after electrophoretic infiltration for 30 min at an applied voltage of 10 V. SiC powder not only covers the surface of the SiC fabric but also deeply infiltrates the fiber area of the fabric to entirely cover its fibers (Figure 1(b)). These images indicate that electrophoretic infiltration is an effective method for the preparation of SiC_f/SiC composites.

Figure 2 shows the densities and relative densities of the SiC_f/SiC composites sintered at different applied pressures, which indicate that density increases with increasing applied pressure. The average densities of the SiC_f/SiC composites prepared at 5, 10, and 20 MPa were determined to be 2.99, 3.10, and 3.16 g/cm^3, respectively. The standard deviations of the average densities are large at 5 and 10 MPa. By contrast, the standard deviations of the average densities are small at 20 MPa. These results indicate that the SiC_f/SiC composites prepared at 5 and 10 MPa contain many pores and have not been completely densified. Given that the theoretical density of SiC is 3.21 g/cm^3, the relative densities of the SiC_f/SiC composites prepared at 5, 10, and 20 MPa are 93.1%, 96.5%, and 98.4%, respectively.

Figure 3 shows the SEM images of sintered SiC_f/SiC composites prepared at different applied pressures, which indicate that all samples have dense structures in their matrix areas (Figures 3(a), 3(c), and 3(e)); however, pores are visible in the fiber areas, particularly for the composites prepared at 5 MPa (Figure 3(b)) and10 MPa (Figure 3(d)). These results are consistent with the data presented in Figure 2, that is, the SiC_f/SiC composites prepared at 5 and 10 MPa are less dense than that

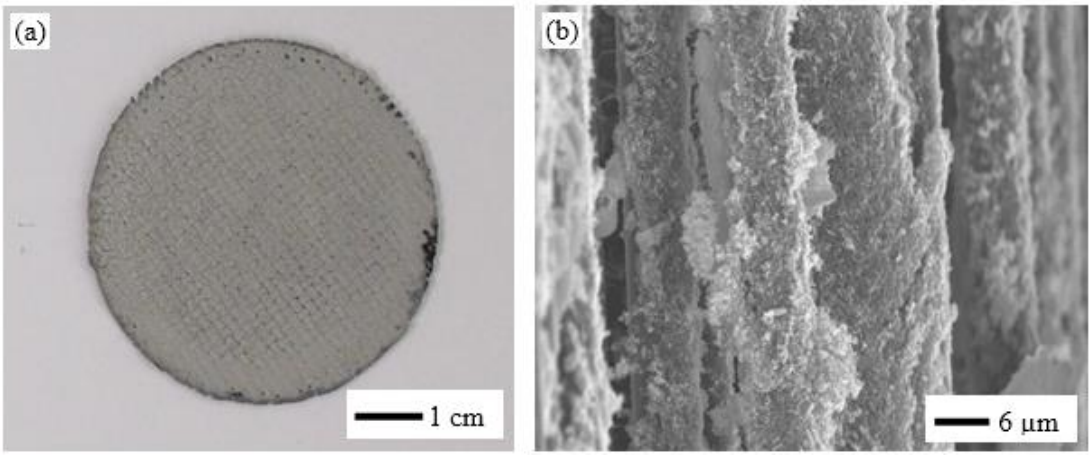


Figure 1. (a) Photographic Image of the SiC Fabric After Electrophoretic Infiltration, (b) SEM Image of The Infiltrated SiC Fabric, Which Shows That the Fiber is Covered by SiC Powder

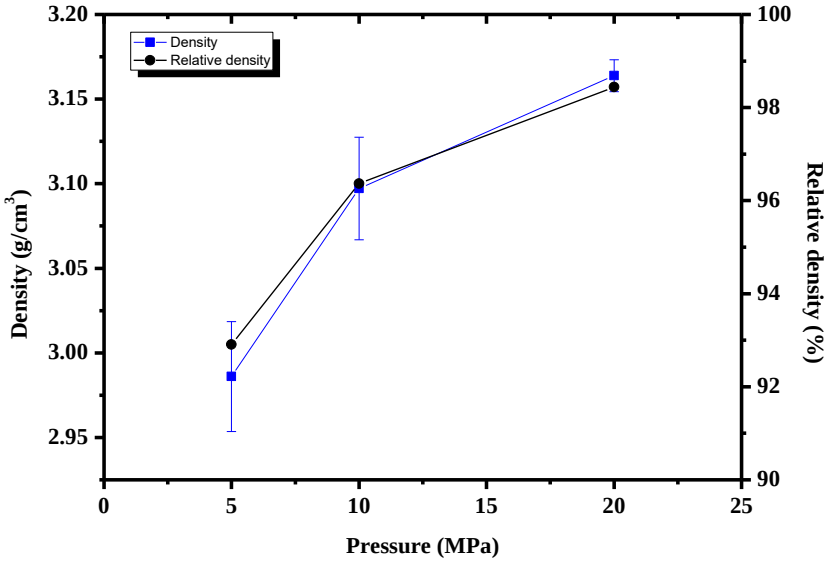


Figure 2. Density and Relative Density of the SiC_f/SiC Composite as Functions of Applied Pressure

prepared at 20 MPa. The SiC_f/SiC composite prepared at an applied pressure of 20 MPa exhibits dense microstructures in its matrix area (Figure 3(e)), as well as in its fiber area (Figure 3(f)). Notably, white phases are present in the matrix and fiber areas, particularly for the SiC_f/SiC composite prepared at 20 MPa. Energy-dispersive X-ray spectroscopy (EDS) analysis (Figure 4) showed that the white phase is composed of Al, Y, Si, O, and C, which correspond to the elements present in the sintering additives (i.e., Al_2O_3 and Y_2O_3). The source of Si may be SiO_2 on the surface of SiC, which reacts with Al_2O_3 and Y_2O_3 and reduces the temperature to induce the formation of the liquid phase. The eutectic temperatures of Al_2O_3–Y_2O_3 and Al_2O_3–Y_2O_3–SiO_2 are 1,760 °C and 1,379 °C [20], respectively; hence, the white phases are deduced to be the liquid phases that form because of the reactions between Al_2O_3, Y_2O_3, and SiO_2. This observation is consistent with that reported in other studies [16,21,22] in which a liquid phase was detected in the fiber area at an applied pressure of 20 MPa. This liquid phase helps densify SiC through a liquid-phase sintering mechanism. The liquid phase is homogeneously dispersed in the matrix area of the SiC_f/SiC composite at an applied pressure of 5 MPa. The liquid phase segregates near the fiber area (Figure 3(c)) or exists in the fiber area (Figure 3(f)) at high pressures. The absence of a liquid phase in the fiber area of the SiC_f/SiC composite prepared at 5 or 10 MPa results in the formation of pores and is the reason why densification is ineffective at these pressures. Moreover, poor densification in the fiber area results in damage to the PyC coating, as shown in Figures 3(b) and 3(d).

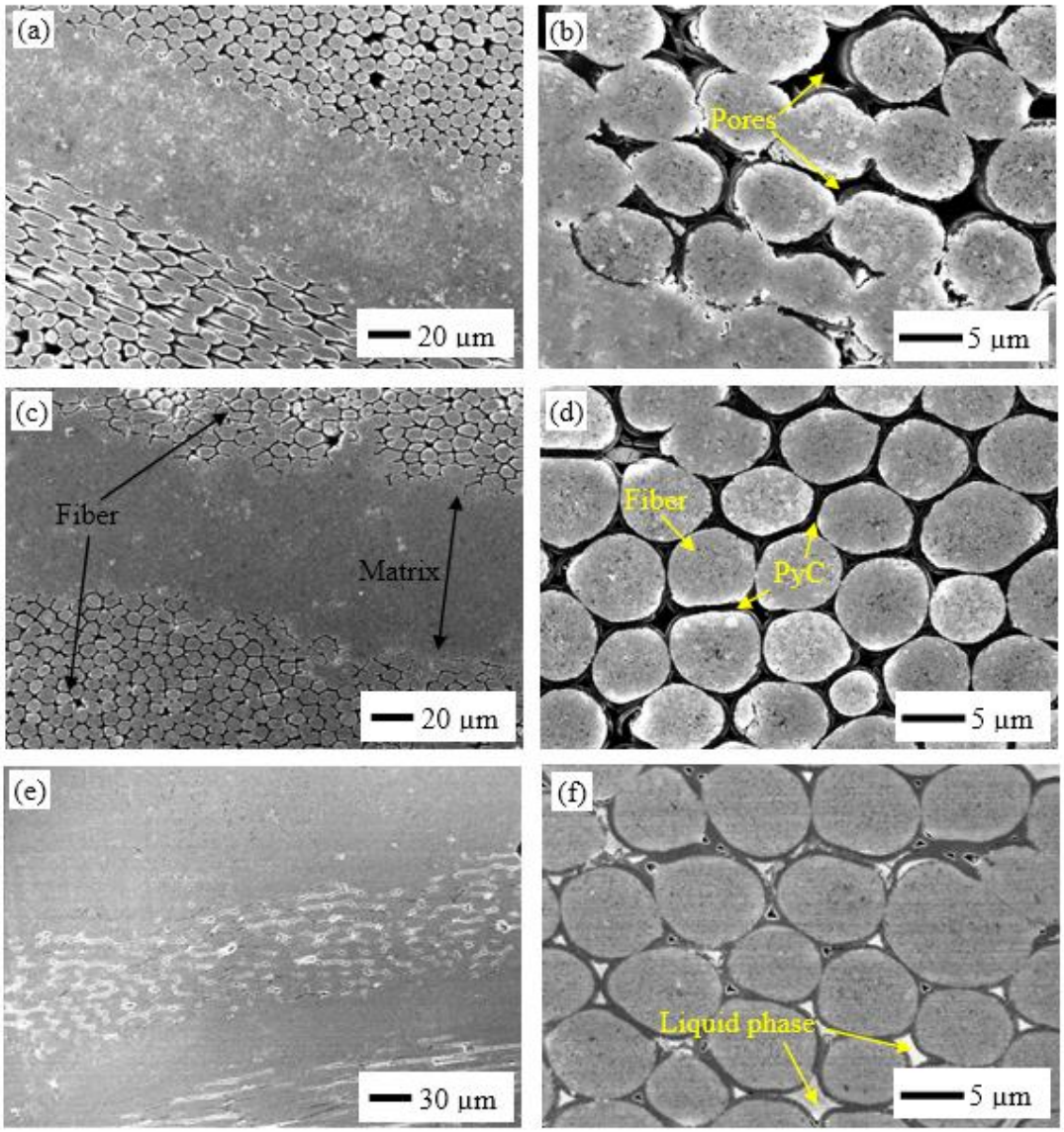
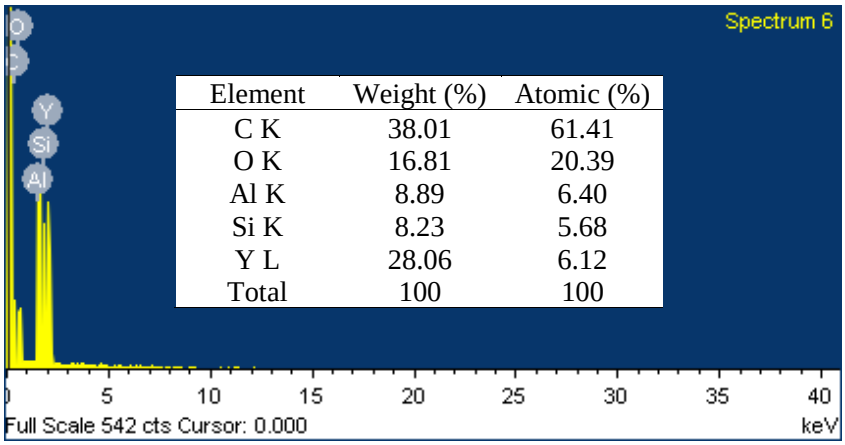


Figure 3. SEM Images of Sintered SiC_f/SiC Composites at Low Magnification ((a) 5 MPa, (c) 10 MPa, and (e) 20 MPa) and at High Magnification ((b) 5 MPa, (d) 10 MPa, and (f) 20 MPa) in the Fiber Area



Element	Weight (%)	Atomic (%)
C K	38.01	61.41
O K	16.81	20.39
Al K	8.89	6.40
Si K	8.23	5.68
Y L	28.06	6.12
Total	100	100

Figure 4. EDS Analysis Results of the White Phases Observed in Figure 3(f)

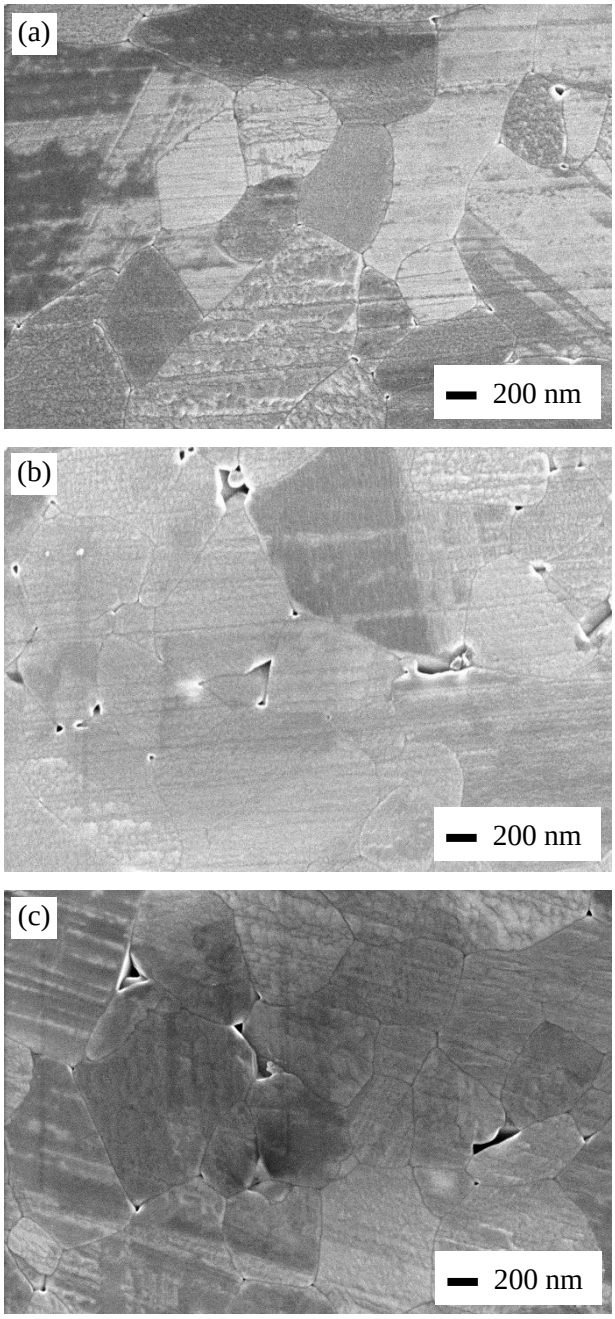


Figure 5. SEM Images Showing the Sizes of the Grains in the Matrices of the SiC_f/SiC Composites Prepared at (a) 5 MPa, (b) 10 MPa, and (c) 20 MPa

Shimoda *et al*. reported that densification in the fiber area is essential to protect the PyC coating and the fiber from damage [21], which leads to minimal degradation of the mechanical properties of the SiC_f/SiC composite. One notable finding in this study is the lack of infiltrated particles in the fiber areas of the SiC_f/SiC composites prepared at applied pressures of 5 and 10 MPa, even though electrophoretic infiltration covered the fabric, as well as the fibers, as shown in Figure 1. This observation is possibly ascribable to particle redistribution resulting from the formation of liquid phases in the first stage of sintering, as well as capillary forces, Ostwald ripening, and attraction of particles in the fiber area of the matrix or the vicinity of the matrix. Consequently, the fiber area was poorly densified because of the absence of infiltrated particles composed of SiC and the sintering additives. In contrast, an applied pressure of 20 MPa hinders the redistribution of infiltrated particles to the matrix area, resulting in the densification of the fiber area. Therefore, 20 MPa is the minimum applied pressure required for the fabrication of SiC_f/SiC composites prepared by electrophoretic infiltration to ensure the densification of the matrix and fiber areas.

The sizes of the grains in the matrices of the SiC_f/SiC composites prepared at various applied pressures are shown in Figure 5. Notably, pressure influences grain size, even though the differences are insignificant. The average grain sizes in the matrix areas of the composites prepared at 5, 10, and 20 MPa are 1,464, 1,354, and 1,282 nm, respectively. Moreover, the matrix of the SiC_f/SiC composite prepared at 5 MPa has a dense microstructure with a few pores, as shown in Figure 5(a). The number of pores in the matrix area increases with increasing applied pressure, as shown in Figures 5(b) and 5(c), which is due to the homogeneous distribution of the liquid phase in the matrix area of the SiC_f/SiC composite prepared at 5 MPa (as discussed in the previous paragraph). Meanwhile, the other samples exhibited segregated liquid phases.

Figure 6 shows the flexural strength of the SiC_f/SiC composite as a function of applied pressure. Indeed, the SiC_f/SiC composite prepared at an applied pressure of 20 MPa has higher flexural strength than those prepared at 5 and 10 MPa. The flexural strengths of the 5, 10, and 20 MPa samples are 198 ± 41, 238 ± 32, and 262 ± 18 MPa, respectively. The flexural strength of the composite is observed to be influenced by its density. For instance, the dense structure of the SiC_f/SiC composite prepared at an applied pressure of 20 MPa reached its maximum flexural strength earlier than those prepared at applied pressures of 5 and 10 MPa. Interestingly, although the SiC_f/SiC composite prepared at 20 MPa shows an abrupt decrease in load after fracturing, which indicates the brittle nature of the composite, it still exhibits a short tail extension, which is ascribable to single-fiber pullout, as shown in Figure 7(c). Meanwhile, the SiC_f/SiC composites prepared at applied pressures of 5 and 10 MPa show relatively long tail extensions, as shown in Figure 6. The tail-extension mechanism of the composites prepared at 5 and 10 MPa is different from that of the composite prepared at 20 MPa. It seems that composite delamination is the main factor responsible for tail extension, as shown in Figures 7(a) and 7(b). The SiC_f/SiC composites prepared at 5 and 10 MPa delaminate because of the presence of pores, which results in low-density fiber areas.

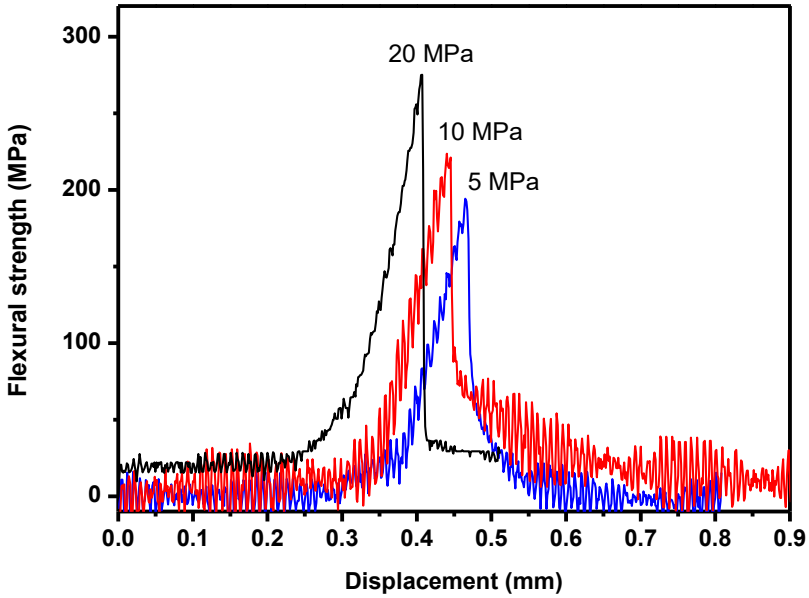


Figure 6. Flexural Strengths of SiC_f/SiC Composites Prepared at Various Applied Pressures

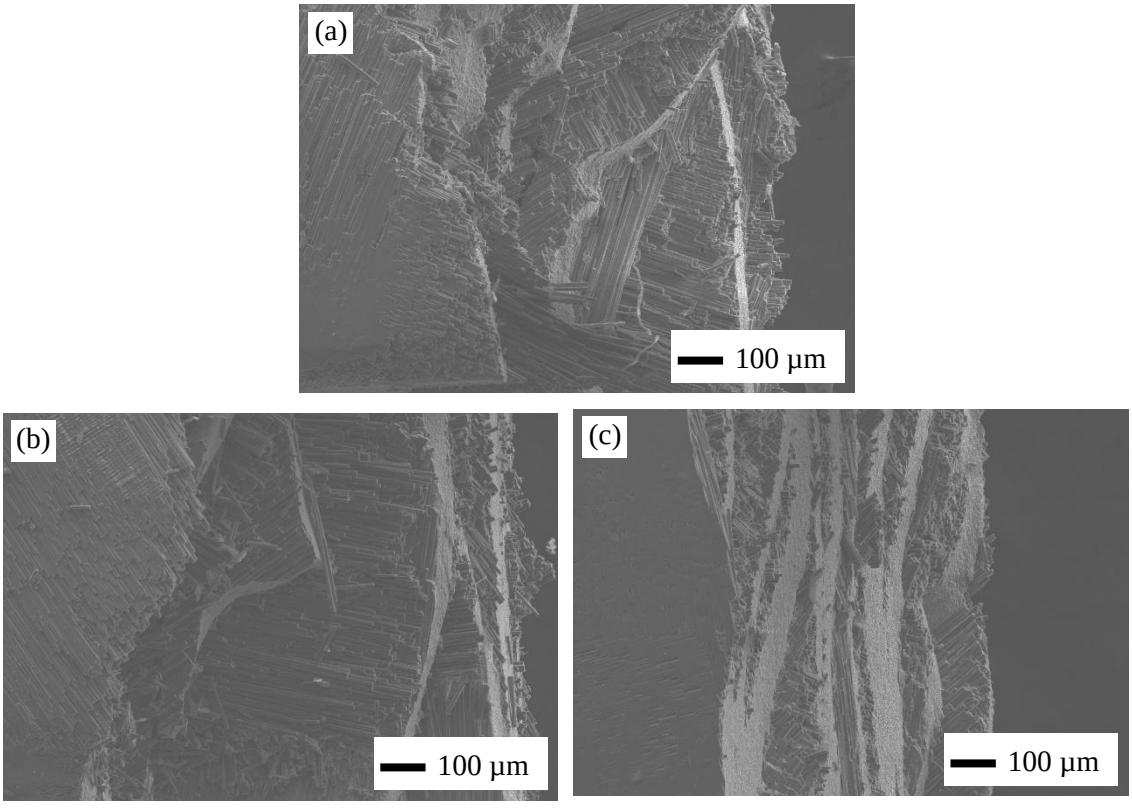


Figure 7. SEM Images of Fractured SiC_f/SiC Composite Surfaces: (a) 5 MPa, (b) 10 MPa, and (c) 20 MPa

Conclusion

In this study, the effect of pressure on the densification and mechanical properties of SiC_f/SiC composites prepared by electrophoretic infiltration was examined. Applied pressure was found to play an important role in the densification of the SiC_f/SiC composite, particularly for the fiber area. The minimum applied pressure that ensures adequate densification was determined to be 20 MPa. Meanwhile, pores remained in the fiber area at

applied pressures of <20 MPa. Poor densification in the fiber areas of the SiC_f/SiC composites prepared at applied pressures of 5 and 10 MPa is due to liquid phase redistribution to the matrix or near the matrix area. Contrary, the SiC_f/SiC composite prepared at 20 MPa successfully maintained infiltrated particles in its fiber area, which resulted in the formation of dense fibers. This result is supported by the homogenous distribution of the liquid phase in the matrix area of the SiC_f/SiC composite prepared at an applied pressure of 5 MPa, with the liquid phase more likely from the fiber area. Hence, a dense microstructure with few pores was detected in the matrix area of the SiC_f/SiC composite prepared at an applied pressure of 5 MPa compared to those prepared at applied pressures of 10 and 20 MPa. Therefore, the density of the SiC_f/SiC composite increases with increasing pressure. For instance, the densities of the SiC_f/SiC composites prepared at applied pressures of 5, 10, and 20 MPa were 2.99, 3.10, and 3.16 g/cm^3, respectively, and the highest flexural strength of 262 ± 18 MPa was obtained at an applied pressure of 20 MPa. This high flexural strength is certainly due to the excellent density of this composite. In contrast, the flexural strengths of the composites prepared at applied pressures of 5 and 10 MPa were 198 ± 41 and 238 ± 32 MPa, respectively.

Acknowledgements

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