

Effects of Pressure during Preform Densification on SiC/SiC Composites

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ABSTRACT

The nano-infiltration and transient eutectic-phase (NITE) method is one of the most attractive methods for fabrication of SiC and SiC/SiC composites. In the NITE method, preform densification is essential option for damage less near-net shaping technique. However, optimization of preform densification is insufficient yet. The objective of this study is to evaluate the effects of pressure during preform densification on SiC/SiC composites. The preform before preform densification has many pores in the inter-prepreg sheets. These pores were disappeared by preform densification. As the effects of pressure on preform, densification in the intra-fiber-bundle was improved due to increasing pressure. Flexural strength of the preforms with 1 MPa and 17 MPa indicated almost same value. The result suggested that increasing of pressure did not cause any change in fiber properties. In the effects of pressure on the composites, the composites with 17 MPa was exhibited improvement in bulk density and mechanical property, compared with that with 1 MPa.

Keywords: SiC/SiC Composites; NITE Method; Near-Net Shaping; Preform Densification

1. Introduction

SiC/SiC composites are promising high-temperature structural materials for advanced nuclear and aero-space applications. The advantage of SiC/SiC composites comes from their low specific mass, superior thermo-mechanical properties and low activation [1-3]. As fabrication processes of SiC/SiC composites, there are three common processes, such as chemical vapor infiltration (CVI) [4], polymer infiltration and pyrolysis (PIP) [5] and reaction sintering/melting infiltration (RS/MI) processes [6]. However, total performances of these composites are still not satisfied for going of industrial stage. Nano-infiltration and transient eutectic-phase (NITE) method is one of the most attractive processes for SiC/SiC composites fabrication to provide high performance on thermo-mechanical properties, size and shape flexibility and acceptable cost [7-9]. In order to produce the complex shape components of SiC/SiC composites by NITE method, the near-net shaping technique is necessary. In general, large volumetric shrinkage (~50 vol%) occurs during ceramic matrix composites fabrication by hot-pressing like NITE method. This volumetric shrinkage is caused due to infiltration and densification process of powder for matrix formation, resulting in unfortunately significant fiber-architecture and strength damage.

Therefore, the method development for suppression of large volumetric shrinkage during hot-pressing is essential to fabricate the production of complex shape by the damage-less near-net shaping, and one of method for that is preform densification before hot-pressing. In fact, the preform densification demonstrated the maintainability of fiber-architecture in composites due to suppression of large volumetric shrinkage and the improvement of composites' density and mechanical properties [10]. However, optimization of conditions (temperature, holding time and applied pressure) during preform densification is insufficient.

2. Objective

The objective of this study is to clarify the effects of conditions of preform densification on SiC/SiC composites. In particular, the effects of pressure during preform densification were investigated on microstructure and mechanical property of preforms and SiC/SiC composites.

3. Experimental Procedure

Pyrocarbon (PyC) coated-TyrannoTM SA fibers (Ube Industrials Ltd., Japan) were used as reinforcement for SiC/SiC composites fabrications. The PyC coating was

appropriately chosen at the thickness of 0.5 μm by chemical vapour deposition (CVD) process. β -SiC nanopowder (IEST, Japan, mean grain size of 32 nm) and sintering additives with Al_2O_3 (Kojundo Chemical Laboratory Co. Ltd., Japan, mean grain size of 0.3 μm , 99.99%) and Y_2O_3 (Kojundo Chemical Laboratory Co. Ltd., Japan, mean grain size of 0.4 μm , 99.99%) were used for matrix formation. For the fabrication of prepreg sheets, PyC-coated Tyranno-SA fibers were impregnated in “nano”-slurry, which consisted of the mixture of SiC nano-powders and sintering additives. Prepreg sheets were stacked for preparation of UD preforms, which is followed by preform densification. The preform densification is performed during heating under isostatic pressures of 1 - 17 MPa. The preforms prepared were hot-pressed at 1870°C for 1.5 h in Ar under a pressure of 20 MPa. The bulk density and open porosity of the preforms and the composites fabricated were measured by the Archimedes' principle. Mechanical property evaluation was performed by three point bending test with the crosshead speed of 0.5 mm/min and a support span of 16 mm at room temperature. The specimens were straight bar type, which measured $26^{\text{L}} \times 3^{\text{W}} \times 1.2^{\text{T}}$ mm³. Microstructural evaluation was inspected by a JEOL JSM-6700 F field emission scanning electron microscope (FE-SEM).

4. Results and Discussions

4.1. Effects of Pressure on Preform

Figure 1 shows optical microscopic image taken on the cross-sectional samples of the preforms before and after preform densification. **Table 1** shows density of preforms after preform densification with different pressure. In the preform before preform densification, pores in the inter-prepreg sheets were observed at many parts. These many pores are possible to affect formation of defects in products. By preform densification, there were disappeared and preform' density also was improved. In the previous study, deformation of fiber and damage of PyC interphase due to preform densification were not seen [11]. The change in microstructure in optical microscope

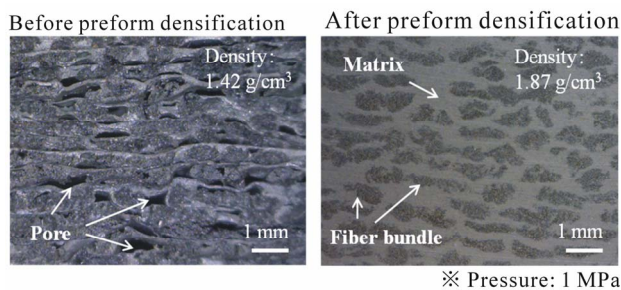


Figure 1. Microstructure of preforms before and after preform densification with different pressure.

observation and improvement in density are not clearly depended on pressure. **Figure 2** shows scanning electron images of the cross-sectional samples of preforms in the intra-fiber-bundle with different pressure. Noticeable pores were not conformed in the all preforms. Matrix slurry seems to be effectively infiltrated by the preform densification. Fiber deformation and damage of PyC interphase were not conformed due to increasing pressure. Contacts between fibers were partially slightly observed. As SEM image analysis, the number of fibers in 50 μm diameter circumference was measured and densification in the intra-fiber-bundle was evaluated. **Figure 3** shows change in number of fibers on the circumference due to pressure. With increasing pressure, the number of fibers on the circumference was increased and tends to saturate with more than 5 MPa. The increasing of number of fibers means disappearance of pores and decreasing of distance between fibers. These results suggest that increasing of pressure might be effective to densification in the intra-fiber-bundles. **Figure 4** shows stress-strain curves of the preforms during the three-point bending test. Both preforms are indicated a pseudo-ductility fracture behavior. The flexural strength of the preform with 1 MPa and 17 MPa are 172 MPa and 173 MPa, which are almost same value. The preform can be defined as fiber-reinforced plastic (FRP), which is consisted of polymer resin matrix. So, the flexural strength of the preform is considered strength of reinforced fibers. Thus, since both of preforms have almost same strength, no influence on fiber properties is suggested due to increasing of pressure. However, these trends due to increasing of pressure are presumed to change by difference in stacking direction of sheets, content and type of binder.

4.2. Effects of Pressure on SiC/SiC Composites

Table 2 lists fiber volume fraction, density and mechanical properties of the composites fabricated in this study. The bulk density is almost same value regardless of the increasing of pressure during preform densification.

Table 1. Density of preform after preform densification with different pressure.

Pressure	1 MPa	5 MPa	10 MPa	17 MPa
Density (g/cm ³)	1.87	1.90	1.89	1.94

Table 2. Characterizations of the SiC/SiC composites fabricated in this study.

Pressure during preform densification	Fiber volume (%)	Density (g/cm ³)	ED/TD ^a (%)	Open porosity (%)
1 MPa	43	2.64	85	10.9
17 MPa	40	2.74	88	7.9

^aED: experimental density, TD: theoretical density.

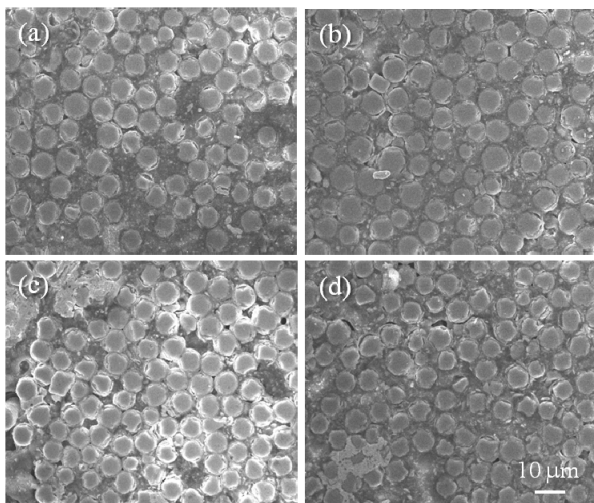


Figure 2. FE-SEM images of preform in the intra-fiber-bundle with different pressure.

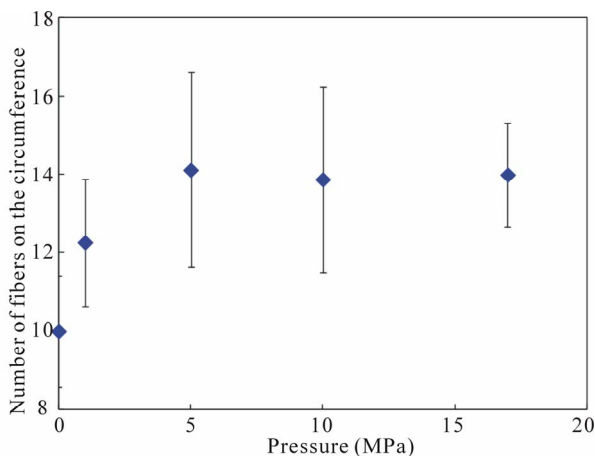


Figure 3. Change in number of fibers due to pressure.

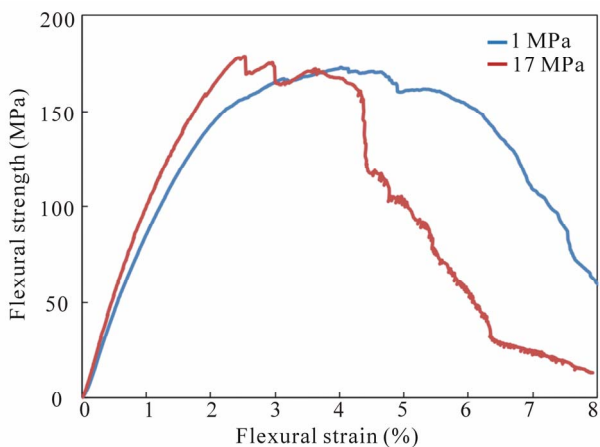


Figure 4. Flexural stress-flexural strain curves of preform with different pressure.

When the pressure is increased during preform densification, the bulk density is slightly increased from 2.64

g/cm³ to 2.74 g/cm³ and the open porosity simultaneously decreased from 10.9 % to 7.9 %. FE-SEM images of polished cross-sectional samples of the composites with different pressure during preform densification are shown **Figure 5**. The cross-sectional areas can be classified into intra-fiber-bundle regions and inter-fiber-bundle regions. Large pores could not be identified at inter-fiber-bundle regions. Pores are mainly distributed in the intra-fiber-bundle regions. In the intra-fiber-bundles, pores of the composites with 17 MPa are slightly decreased than that of the composites with 1 MPa. This result is considered contribution of densification in the intra-fiber-bundles due to increasing of pressure during preform densification. In the backscattered electron images, the bright contrast phase indicates remnants of the oxide additives with high Z elemental compositions (**Figures 5(c)** and **(d)**). Shimoda *et al.* have reported that this phase is mainly consisted of Al, Y and O elemental compositions by the energy-dispersive X-ray spectroscopy (EDS) analysis [12]. This phase was agglomerated in the inter-fiber-bundle regions and around intra-fiber-bundle regions. The difference in scatteration of remnants of the oxide additives due to pressure during preform densification was not clear. The fiber deformation is slightly observed in both the composites. Fiber deformation ratio of composites with 1 MPa and 17 MPa, defined as a fiber aspect ratio of longer dimension to shorter dimension, is 1.2 and 1.2, which is consistent. The PyC interphase of both composites is maintained regardless of pressure during preform densification. **Figure 6** shows stress-strain curves of the composites during the three-point bending test. The sample number for each composite was five. Both of the composites displayed a pseudo-ductility fracture behavior. The average flexural strength of the composites with 17 MPa is 471 MPa, which is slightly higher than that of the composites with 1 MPa. The difference of flexural strength might be due to

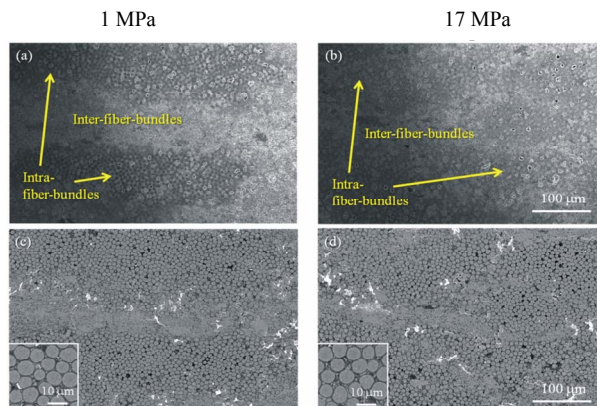


Figure 5. FE-SEM images taken on the polished cross-sectional samples of the composites: (a), (b) Scanning electron image; (b), (c) Backscattered electron image.

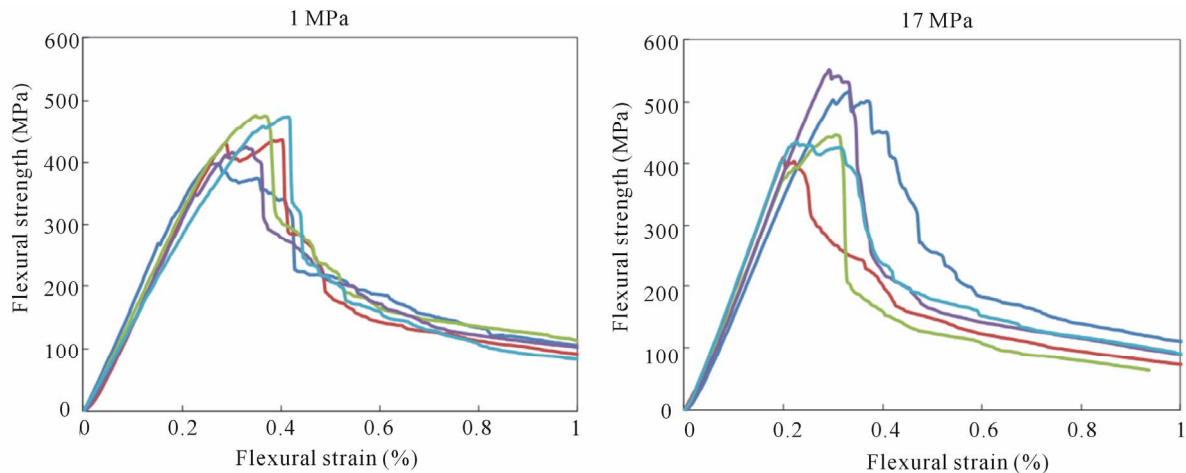


Figure 6. Flexural stress-flexural strain curves of the composites with different pressure during preform densification.

enhanced densification in the intra-fiber-bundles.

5. Conclusion

The effects of pressure during preform densification on microstructure and mechanical properties of preforms and SiC/SiC composites were evaluated. The preform before preform densification had many pores in the inter-prepreg sheets. These pores were disappeared by preform densification. As the effects of pressure on preform, densification in the intra-fiber-bundle is improved due to increasing of pressure. Flexural strength of the preforms with 1 MPa and 17 MPa is indicated almost same value. The result suggested that increasing of pressure did not cause any change in fiber properties. In the effects of pressure on the composites, the composites with 17 MPa was exhibited improvement in bulk density and mechanical property, compared with that with 1 MPa.

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