Mechanical behavior of SiC_f reinforced SiC composites with fiber coating — Stress alleviation in SiC fiber by soft coating layer

Kyung Soon JANG, Eugene LEE, Tae Woo KIM, In Sub HAN,* Sang Kuk WOO* and Kee Sung LEE†

School of Mechanical and Automotive Engineering, Kookmin University, Seoul 136-702, Korea

*Convergence Materials Research Center, Korea Institute of Energy Research, Daejeon 305-343, Korea

The present research attempts to analyze and evaluate the mechanical behavior of SiC_f in SiC_f/SiC composites with and without soft coatings. The effects of soft fiber coating in the alleviation of indentation or flexural stress in the layered structure are investigated. Boron nitride and/or pyrocarbon as soft coating layer on SiC fiber are selected for the structure. Modeling is performed by changing the type of fiber, the type of soft layer, and layered stacking sequence of the soft layer. Finite element method (FEM) analyses are conducted to evaluate stress distributions under indentation or flexure loadings by displacement loading. Experimentally, the fiber coatings are conducted in the SiC fiber reinforced SiC composites. It was found that BN and/or pyrocarbon coating on the fiber alleviate the stress under the contact or flexure loading, which indicates that the appropriate design by soft layer on the fiber can reduce the maximum stress.

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1. Introduction

Silicon carbide (SiC) exhibits high thermal conductivity, hardness, chemical stability at high temperature as well as good resistance to corrosion, thermal shock.^{1)–5)} The use of SiC, when either carbon fibers or silicon carbide fibers were incorporated to form ceramic/ceramic composites, made wide industrial applications of SiC possible. The thermo-mechanical applications of SiC include radiation heaters, heat exchangers, gas turbines and structural parts in nuclear fusion reactors.^{6,7)}

Among ceramic composites reinforced with whiskers, particles, and fibers, composites with continuous fibers have shown effective strengthening and toughening.⁶⁾ Most of SiC fibers have been produced in Japan, and they have good purity and mechanical properties.⁸⁾ SiC fibers have also been woven into a woven mat and the fiber preforms have been infiltrated with the matrix materials to be densified. The fiber/matrix interface properties, which are determined by judicious selections of coating material, the level of interface bonding, may change the mechanical behavior of the composite. The potential coatings include pyrocarbon (PyC) and boron nitride (BN).^{9)–12)}

The present study is to intend to determine the role of coating in the mechanical behavior of SiC fiber/SiC composites with only considering one or two soft coating layers on the fiber. Since the coating is the key material in determining the mechanical behavior, the present model was focused on determining the direct effect of coating on the stress distribution and deformation analytically and experimentally.

2. Materials and procedures

2.1 Materials

The properties of fiber were used for SiC from UBE Ind., Ltd. (Japan). The grades are Tyranno grade S (type: TY-S1H16Px, Bobbin No. 08407K0724) and Tyranno grade LoxM (type: Tn-

S1E08px, Bobbin NO. 07785K0703). The Young's modulus for Tyranno S and Tyranno LoxM is 170, and 187 GPa, respectively. The Poisson's ratio is assumed to be 0.2 for both types of fibers.

Coatings selected for the present study were BN and PyC. BN coating was selected because it has low Young's modulus and good interface properties. PyC coating was also chosen with the same reasons. The Young's modulus for BN coating and PyC coating is 10.3, and 29.4 GPa, respectively. The BN coating that has a lower Young's modulus than PyC is considered to be relatively softer material as compared to PyC coating. The same value of Poisson's ratio that was used for the fiber was used for the coating. Assuming the same Poisson's ratio for both fiber and coatings eliminated any deformation difference at the interface due to Poisson's ratio mismatch.

2.2 Modeling and analysis

The present study is concerned with the mechanical behavior for the SiC/SiC composite system, and particularly for the coating and fiber. The matrix was not included intentionally in the analysis. The analysis was done by changing parameters of number and sequence of the coating. The mechanical behavior was determined with indentation and flexural loadings as shown in **Figs. 1(a)** and **(b)**, respectively.

The maximum stress developing on the system with the change of the fiber was compared in order to choose the preferred fiber/coating combination to reduce the maximum stress. The elastic modulus mismatch is the parameter in changing the stress distribution within coating/fiber structure. The sequences of two different coatings (PyC and BN) were checked for double-layered structure. The effect of soft coating onto the fiber on the alleviation of indentation or flexural stress in the layered structure was investigated by using FEM (finite element methods).

It is difficult to model the individual fiber in the woven mat. Instead the bundles of the fibers were modeled as a layer of fibers having the same properties of individual fiber. The thickness of the woven fiber is commonly measured in the order of about 1

† Corresponding author: K. S. Lee; E-mail: keeslee@kookmin.ac.kr

mm, thus the thickness of the fiber was modeled to be 1 mm in the present study.

Each thickness of the coating was modeled to be 0.2 mm, which is about 20% of the fiber thickness. The actual thickness of the coating may be thinner than the modeled thickness of 0.2 mm. Because this study is concerned with the comparison of the mechanical behavior, the thickness of the coating was intentionally enlarged for easier comparison.

For indentation modeling, the rigid sphere of diameter 3.18 mm was used and the displacement of the rigid sphere was fixed to be 0.01 mm. Instead of applying the same indenting force, the same indenting displacement was applied in the present study. It was because the ceramic can be displacement limited, and the displacement control provides better numerical convergence in computations. Axisymmetric plane was modeled, but the results were shown for the half-plane for better presentation of the results.

Single coating represent either BN or PyC coating and the double coating implies either BN/PyC or PyC/BN coating on the top of fibers. The stress distribution and the maximum stress were compared with the change of material system type. Besides, the largest displacement of the fiber was compared to determine the effect of fiber type and coating sequence.

For flexural loading, the matrix was again excluded to find the direct effect of coating type and coating sequence on the fiber on the stress distribution. The thickness of coating was 0.2 mm which is the same thickness for modeling indentation. The plane stress condition was selected, and the maximum in-plane stresses were compared.

2.3. Experiment

SiC_f/SiC composite including soft coating layers onto the fiber interface were prepared experimentally. The composites were produced by RMI (Reactive Melt Infiltration) process in the laboratory after preparing 2D-fiber architecture. The matrix is formed by chemical reaction between a Si liquid precursor and a carbon material in the fiber preform.

The commercially available 2-D fiber mats (Tyranno S grade, Tyranno, Japan) were prepared. PyC coating layers onto the fiber mats were deposited in a CVD (chemical vapor deposition) reactor. The constant furnace temperature, 800–1100°C was uniformly maintained across all zone in nitrogen atmosphere. Propane (C₃H₈) gas was flowed with nitrogen gas into the chamber

Fig. 1. Schematic diagrams of (a) indentation and (b) 3-point flexural loading.

and the reaction was performed for 60 min. The flow rates of propane and nitrogen gas were adjusted 14 cc/min and 140 cc/min, respectively.

Several fiber mats with a dimension of 5 mm × 5 mm × 4 mm were stacked. The phenol resins were impregnated into the fiber mat under vacuum condition. BN coating layers were deposited onto some fiber mats using spray coating machine. The BN slurry was prepared from BN powder, PVB, fish oil, DBP in the mixed solution of toluene and 2-PrOH. Constant velocity of sprayer was maintained during coating.

The coated and impregnated fiber mats were in contact with Si powder (Boram-chemical, 1mm, Korea) in a graphite crucible. The crucible was placed in a graphite resistance furnace, slowly heated up to 600°C to remove the organic additive, and heated up to 1600–1700°C in a vacuum at 13.3 Pa for 30 min. After the heat treatment, the excess free Si which existed on the surface of fully infiltrated bodies were ground off by grit blast.

The surface of the samples were cut into pieces, 3 mm × 4 mm × 25 mm, ground and polished to 1 μm of diamond suspension finish. For obtaining load-deflection curves of the composites during fracture, the tests were conducted on bar specimens in a flexure. The specimens were fractured at 0.02 mm/min in the flexure.

3. Results and discussion

Figure 2 shows the comparison of maximum stress distributions for two different fibers receiving indentation loading. Indentation loadings are good mechanical tool to induce damage in the layered specimens. When a hard spherical indenter is pressed on a flat surface of material, the contact force produces a crack with cone shape trajectory from maximum stress region outside the contact zone.¹³⁾ The maximum stress under the same indenting displacement for Tyranno S grade was found lower than that for Tyranno LoxM grade by about 7 percent. As the strain is constant in the fixed constrain condition during indentation, the stress is proportional to elastic modulus if the material

Fig. 2. Comparison of maximum stress contours at two different fibers under indentation (a) Tyranno S fiber, (b) Tyranno LoxM fiber.

is not deformed. The lower stress is caused by the lower elastic modulus of Tyranno S grade.^{14),15)} Note the maximum stresses under indentation are 289 MPa for Tyranno S fiber and 318 MPa for Tyranno Lox M fiber.

The maximum stress and distribution with the change of single coating onto Tyranno S fiber were compared in Fig. 3. The maximum stress with BN coating onto fiber was lower than that with PyC coating. It is considered that the higher modulus of PyC was resisting the indenting and the stress was developed higher as shown in Fig. 3(b). These results were consistent with the results found for coating/substrate system in the reference.^{16),17)} Because similar stress contours were found for the Tyranno LoxM fiber with a coating, the contours were not shown in the paper. It is notable the maximum stresses under indentation go down to 26–67.5 MPa relative to high stresses of fiber without coating as shown in Fig. 2.

In order to determine the effect of coating material on the mechanical behavior, the largest stress and displacement within the fiber were compared and shown in Fig. 4. As shown in Fig. 4(a), the stress with PyC coating was found to be higher than the system with BN coating. The maximum displacement was also larger for fiber with PyC coating. The present study used the same indenting displacement for both coatings. In order to induce the same indenting displacement, the PyC with higher stiffness has pushed down the underlying fiber more which has resulted in larger displacement.

When both PyC and BN coatings were stacked to form double coatings, the mechanical behavior can depend on the coating stacking sequence. Figure 5 shows the comparison of maximum stress for two different sequences. Because of the BN coating with relatively lower elastic modulus as soft interlayer between the upper PyC coating and the fiber, the PyC tended to be bent downwards and thus the high tensile stress was thought to develop within the PyC coating. It is flexural stress to dominate the damage patterns of PyC on BN coating during indentation

Fig. 4. Comparison of maximum (a) stress, (b) displacement developing within the fiber depending on the type of fiber and coating.

(a) BN-Tyranno S

(b) PyC-Tyranno S

Fig. 3. Contours showing the location for maximum stress with single coating of (a) BN-fiber and (b) PyC-fiber.

(a) PyC-BN-Tyranno S

(b) BN-PyC-Tyranno S

Fig. 5. Contours showing the location for maximum stress with double-coating of (a) PyC-BN and (b) BN-PyC.

because the under layer is relatively softer.^{18)–20)} On the other hand, the presence of relatively soft BN coating when located directly under the rigid indenter produced lower maximum stress at the contact surface as shown in Fig. 5(b). The result of Fig. 5(b) was consistent with the results of Fig. 3(a), where the soft BN coating was directly under the rigid indenter.

Maximum stress and displacement developing within the fiber were compared depending on the coating sequence and shown in Fig. 6. As shown in Fig. 6(a), maximum stress with BN–PyC coating sequence produced slightly higher stress. The coating sequence of PyC–BN resulted in high stress within PyC due to bending of PyC onto soft BN. However, the presence of soft BN top layer alleviated stress developed within fiber. It is notable that the maximum displacement with BN–PyC coating was only slightly lower as shown in Fig. 6(b).

Summarizing results in Fig. 7 shows that the presence of soft coating can alleviate the large stress as found in the fibers alone. All types of soft coating in this study alleviated the stress. The PyC with larger elastic modulus when located directly under the indenter yielded relative large stress. Besides, the reason for high stress for PyC–BN sequence is because the maximum stress was developed within PyC coating lying onto the soft BN coating. The flexural deformation of PyC yielded high tensile stress. However, the maximum principal stress of PyC–BN coated fiber system is still lower than that of the system without coating. Coatings are applied to SiC/SiC composite to improve its durability with oxidation and failure resistance. Therefore damage-insensitivity is good strategy of development in high-reliable ceramic composites for high temperature applications. As a damage accumulation from mechanical fatigue at high temperature can cause the failure of the composites, damage tolerance by stress alleviation is desirable.²¹⁾

As shown in Fig. 7, the stacking sequence of double layer of BN–PyC produced lower stress than that of PyC–BN. This sequence indicates that the coating with the lowest modulus of BN at top and PyC of medium modulus, and the fiber with the largest modulus at bottom was the stacking sequence yielding the lowest stress among all types under study.

Figure 8 compares locations for the maximum tensile flexural stress for three cases; without coatings, with a single coating and double coatings. The tensile stresses were applied onto the coating layer for the coated systems. Even though the strain varied linearly with the depth of the beam, the higher elastic modulus of fiber as compared to that of coating produces higher tensile stress. However, it is noteworthy that the maximum stress regions are moved from the outermost surface for the fiber without coating to the inner regions for the fiber with single or double coating.

Figure 9 shows comparison of maximum stress from contours in Fig. 8. The presence of soft coating was found to alleviate the tensile stress. For a single coating, the stress was lower with PyC coating. For double coatings stresses were similar regardless of coating sequence of BN–PyC or PyC–BN. For all material cases, the material with Tyranno fiber S yielded lower stress than that with Tyranno fiber LoxM grade.

The failure behaviors of SiC without coating, with BN coating and with BN/PyC coated samples are compared in Fig. 10. The load-deflection curves are obtained experimentally for samples fabricated by reactive melt infiltration process. In flexural loading mode, different behaviors of load-deflection curves are observed, depending on the coating type and sequence. While the SiC composites without coating shows brittle behavior, the fiber composites with soft coatings show a strong nonlinear behavior,

Fig. 6. Comparison of maximum (a) stress and (b) displacement developing within the fiber depending on the sequence of double coatings.

Fig. 7. Comparison of maximum stress depending on the fiber type, the presence of coating, and the sequence of two coatings.

especially after failure occurred. Saw-like fracture behavior was found for coated composites because high strength fibers do not deform prior to fracture of coatings. Even though the maximum load is lower due to lower strength of BN itself, the experiment evidence on the composites suggests damage-tolerant fracture behavior of SiC_f/SiC composite with soft coating layers.

Figure 11 shows SEM micrographs of a section from a SiC_f/SiC composite. This image shows the pull-out is apparent along the interface surrounding the fibers during 4-point bending test. Such pull-out phenomena surrounding the fibers tend to increase the saw-like fracture behavior as the fiber with a volume fraction

Fig. 8. Location change for the maximum stress under flexural loading (a) without coating, (b) with a single coating and (c) double coatings layer.

Fig. 9. Comparison of maximum flexural stresses depending on the fiber type, the presence of coating, and the sequence of two coatings.

of 40% can support the load without failure.

4. Conclusions

In order to determine the mechanical behavior of composite, important design information with coating were studied. The present study was focused on direct effect of coating on the stress

Fig. 10. Load-deflection curves of SiC/SiC composites without coating, with a single coating and double coatings layer.

Fig. 11. SEM micrographs of a section from a SiC_f/SiC composite, showing pull-out of fiber at (a) higher magnification, $\times 5000$ and (b) lower magnification, $\times 1000$.

and deformation on the fiber, and matrix was not included. The information can be used as guidelines for selecting coating structure in composite including matrix. Several conclusions were drawn based on the present study.

In general, the lower modulus of Tyranno S fiber as compared to Tyranno LoxM fiber induced lower stress.

The presence of soft coating was found to alleviate the tensile stress under indentation and flexural loading. For a single coating

under flexural loading, the stress was found lower with PyC coating. For double coatings stresses within the fiber were similar regardless of coating sequence of BN-PyC or PyC-BN.

The stacking sequence of double layer of BN-PyC produced lower maximum stress than that of PyC-BN. This sequence indicates that the coating with the lowest modulus of BN at top and PyC of medium modulus, and the fiber with the largest modulus at bottom was the optimum stacking sequence yielding the lowest stress among all types under study.

Experimentally measured load-deflection behaviors of SiC_f/SiC composites with soft coating layers indicate damage-tolerant fracture behavior.

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