TENSILE PROPERTIES AND FRACTURE MECHANISMS OF UNIDIRECTIONAL SIC/SIC COMPOSITES

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1 Introduction

Silicon carbide fiber reinforced silicon carbide matrix composite (SiC/SiC composite) is expected to be used as the structural material of airframe and engine of future aerospace transportation system, because of its superior mechanical performances at high temperature [1-3]. In ceramics matrix composites (CMCs) entirely consisted of essentially brittle materials such as SiC/SiC composite, a fiber/matrix interface bears an important role in various mechanical properties4. According to W. A. Curtin's theory [5], a matrix also has an important role on tensile properties in load transferring mechanics. In a fracturing process, it receives a distributed load from the broken fibers due to the frictional resistance of an interface. This provides the material with pseudo-ductility and consequently enhance the mechanical performance of CMC. A microstructure to yield many long pull-out fibers on fracture surface is not sufficient, but the microstructure in which the load carrying-capability of fiber and the probability of fiber breakage are well balanced and those efficiently contribute to bear a load is preferable. In the aspect of manufacturing technique, the matrix of CMC is generally difficult to densify sufficientlly and its microstructure much varies depending on process conditions [6-9]. So that, CMCs need to be considered as "process defect tolerant material". The important matter is to make the best condition that enables fibers to maximize their potential performances in limited fabrication techniques. For this purpose, the effects of interface layer and matrix on tensile properties need to be sytematically understood.

In this work, the effects of the fiber/matrix interface and matrix of the SiC/SiC composites on the tensile properties were experimentally investigated. Tests were conducted for the bundle composites, that were composed of a fiber bundle densified with a matrix, because of its structural simplicity and the applicability of its data to higher order reinforcement structures. The samples were prepared by polymer impregnation and pyrolysis (PIP) method, focusing on its wide applicability in size and shape, and controllablility in microstructure. The microstructure of the samples was systematically controlled by the deposition time for carbon interface layer, the precursor polymer, the filler blending rate and the number of densification processing.

2 Experimentals

Bundle composites that are fiber bundle with interface layer and matrix in various conditions were prepared for the evaluation.

Tyranno-ZMI SiC fiber (Ube Industries LTD., Japan) was used for the reinforcement. As the precursor for the matrix, allylhydridepolycarbosilane (AHPCS, Starfire Systems Inc., USA) and polycarbosilane (PCS, Nippon Carbon Co., Ltd., Japan) were adopted. For the filler material of the matrix, SiC fine powder, ultra-fine grade of β-randomTM, (Ibiden Co., Ltd., Japan) was used. Its average particle size was 270 nm.

Fabrication procedures for the composite samples were as follows; 1) fiber bundle was fixed to the carbon fixture, 2) C layer was formed by chemical vapor infiltration (CVI) method on the fibers, SiC layer of about 100 nm thickness was subsequently formed on the layer as its protective coating, 3) the precursor polymer or its powdery slurry was impregnated into the bundle in vacuum, 4) the prepreg bundle was heated in argon atmosphere, 5) the sample was subjected to multiple polymer-impregnation and heating. The procedure until 4) is defined as "first densification processing" (N=1) in this article. The source gas of C layer and SiC layer were methane (CH_4) and methyl trichlorosilane (CH_3SiCl_3) respectively. PCS was used as a hexane solution. Through these processes, linear composite samples of more than 150 mm length with a diameter of around 1mm were obtained.

The samples were evaluated by tensile test in the fiber direction, monofilament push-out test and microscopy for the polished section and the fractured section by optical microscope (OM) and scanning electron microscope (SEM). The tensile test was conducted under 0.5 mm/min of displacement rate and 90 mm of gage length. At least 5 samples were tested for each fabrication condition. The samples fractured at more than 5 mm distance from the grip part were defined as "5 mm condition clearing sample". The push-out test was conducted using a Berkovich-shape indenter with flat bottom at the loading rate of 20 mN/s. The disc specimens of around 200 μm thickness with both sides polished were prepared for the test. At least 20 effective data points were obtained for each sample. The interfacial shear strength was calculated from a load at the flat area of a load-displacement curve being divided by the area of an interface area that contributed to the slide. The fiber breakage locations were manually scanned using OM. The pull-out fiber length of the fractured surface was manually measured using SEM micrograph. All fiber that could be observed for a bundle were counted.

3 Results and Discussions

Because the bundle were normally consisted of almost same number of filament, the comparison in the maximum tensile loads could be directly connected with that in those strengths. No significant difference was recognized for either sample in the results of comparison between the average values of the maximum load for the "5 mm condition clearing samples" and "all samples". Since there was no significant effect to the tensile characteristics due to the breaking position within the same sample, it can be concluded that the tests were properly conducted. The average values of the "5 mm condition clearing samples" are shown as follows.

3.1 Number of Densification Processing

The maximum tensile loads of the samples after respective times of densification processing are shown in Fig. 1. The maximum tensile load improved by applying PIP processing 1 through 8 times, in comparison with the fiber bundle only with interface treatment (n=0). It was confirmed that the densification degree of a sample after 8 times PIP processing almost reached saturation in the previous work [10]. Accordingly, the tensile strength of a bundle with coating layer was around half of that of a highly matrix-densified composite.

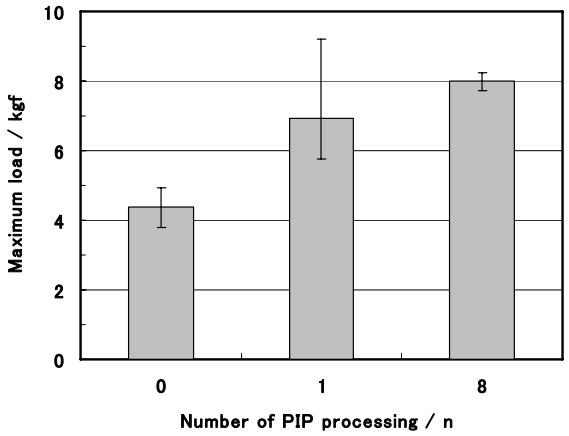

Fig. 1. Maximum loads of the composites of various number of densification processing.

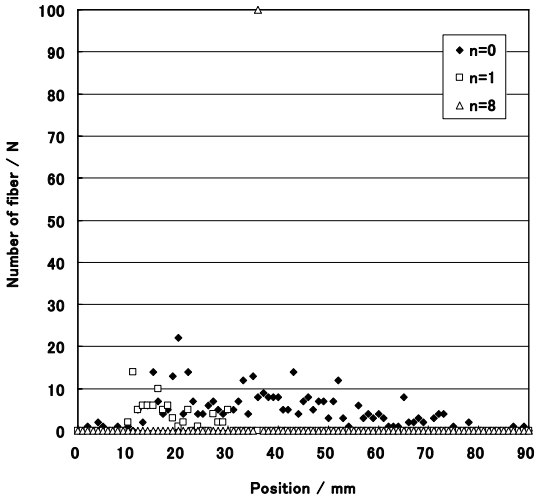

Fig. 2. Distributions of fiber breakage location in the entire gauge length for the bundles after various number of densification processing.

Fig. 2 exhibits the distribution of fiber breakage location in the gage area. For the sample of n=0, each fiber was mechanically almost independent, so that the fiber was broken randomly at its weakest points through the entire gauge length. On the other hand, fiber breakage area was limited in some ten milimeter

and further a milimeter for the sample of n=1 and n=8, respectively. It is suggested that as the matrix surrounding the fiber was densified, the matrix break in fiber direction decreased and the breaking at a weakest point of each fiber was suppressed so that the fracture was limited around the main crack point, resulting in the increase of the maximum load. Consequently, the importance of matrix densification was confirmed.

3.2 Thickness of Interface Layer

The maximum tensile loads of various carbon layer thickness sample are shown in Fig. 3. Testing for the samples without carbon layer was not possible because they were so weak that they fractured during preparation. Mechanical properties were greatly improved by adding a carbon layer of only 50 nm. Then the maximum load improved as the thickness was increased. In consideration of the interfacial properties, these behaviors are quite reasonable. Although there have been done many studies for the range of some 100 nm to 1000 nm in the past [11], no clear difference was recognized here. Rather, mechanical properties have even improved by increasing the carbon layer thickness up to the range of micron order.

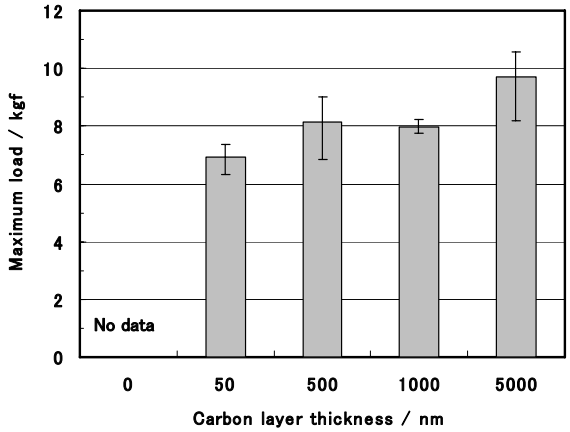

Fig. 3. Maximum loads of the composites of various carbon layer thicknesses.

Fig. 4 exhibits the distribution of the pull-out fiber length obtained for the samples of the carbon layer thickness of 50 nm, 1000 nm and 5000 nm. The average lengths of each sample are also noted near their lines. It was clearly found that the distribution of the pull-out fiber length was shifted to higher value as the thickness increased. In case of the sample of 50 nm, only a few fibers were beyond 100 μm. Consequently the average length became higher from 12 μm at 50 nm to 116 μm at 5000 nm.

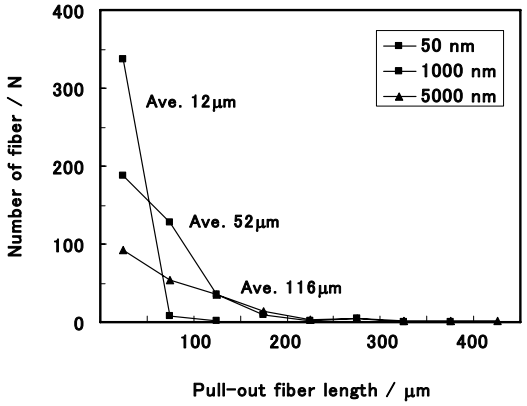

Fig. 4. Distributions of pull-out fiber length for the composites of the carbon layer thickness of 50 nm, 1000 nm and 5000 nm.

Fig. 5 shows the interfacial shear strength of the composites of various carbon layer thickness. As with previous reports [11,12], the shear strength continuously decreased as the thickness increased. It was found that the effect of a carbon layer thickness on the shear strength was not saturated below 1000 nm but extensively continues up to several micron order in this work.

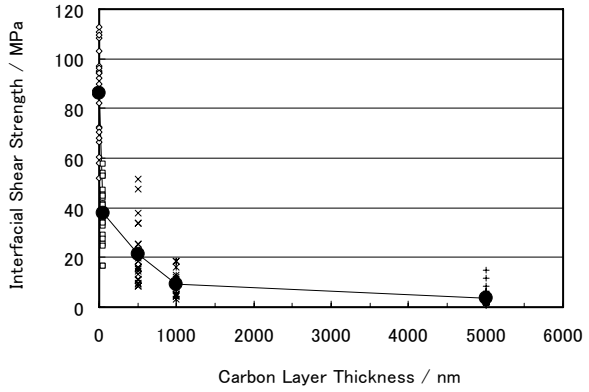

Fig. 5. Interfacial shear strengths of the composites of various carbon layer thicknesses.

For the results of the pull-out fiber length and the interfacial shear strength, it was simply shown that the length increasingly became longer as the shear strength decreased in such wide range as the several nm-to-several μm. It is easily conceivable that this behavior is closely related with the efficiency of the deflection and propagation of a crack to interface direction. On the other hand, the behavior of the tensile strength was not simple. It can not be explained only with the amount of fiber pull-out. The increase shown from 50 nm to 500 nm should be certainly due to crack deflection efficiency. The tensile strength became almost same between 500 nm and 1000 nm, and again increased between 1000 nm and 5000 nm. This implys the existence of another factors that enhance and decline the tensile strength. One is the load transfer capacity that declines the strength as the interfacial shear strength decreases and the other is the structural compensation and/or stabilization for a fiber by an interface layer that enhance the strength as the interface layer thickness increases. Further study for this issue is ongoing.

3.3 Precursor polymer

The maximum tensile loads of the composites fabricated using AHPCS and PCS are exhibited in Fig. 6. It has been known that AHPCS has better weight yield, which well contributes to the higher densification for the same number of PIP processing as compared with PCS10. Contrary to the degree of densification, higher maximum load was obtained for PCS-derived composite rather than AHPCS-derived one. Although an effectiveness of matrix densification on tensile strength was confirmed above, it was not directly shown in this case. There should be another critical factor in tensile property.

Fig. 7 shows the length distribution of pull-out fiber for the samples fabricated using AHPCS and PCS. Compared with the AHPCS-derived sample, much longer pull-out fibers was seen for the PCS-derived sample for the same thickness of interface layer. The average length of the PCS sample was 116 μm which was almost same value as that observed in the AHPCS sample with the carbon layer thickness of 5000 nm.

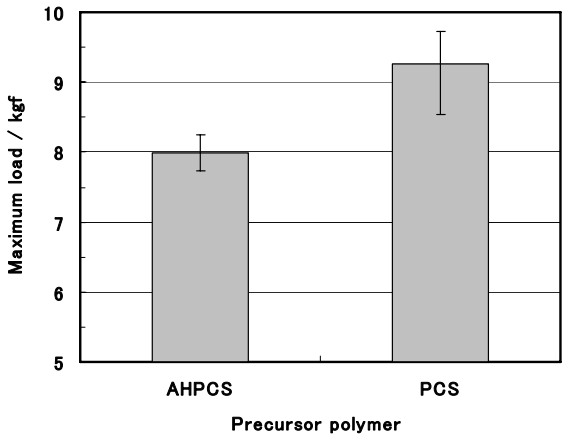

Fig. 6. Maximum loads of the composites obtained using AHPCS and PCS.

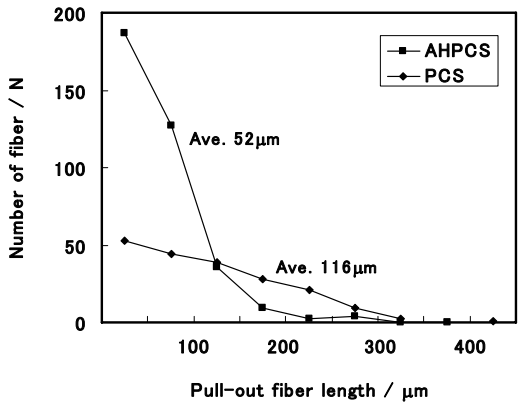

Fig. 7. Distributions of pull-out fiber length for the composites fabricated using AHPCS and PCS.

The interfacial shear strength of both composites were exhibited in Fig. 8. The values obtained for the PCS-derived composite clearly higher than that obtained for the AHPCS-.

Although the PCS one has higher interfacial shear strength, longer pull-out fibers were made. The relationship of these results is presently supposed to be related with the mechamical characteristics of the matrices. There would be some morphological and/or compositional advantage in improving structural reliability and/or uniformity for PCS-derived matrix rather than AHPCS-derived one. Further investigation is necessary for this matter.

Anyway, longer fiber pull-out directry contributed for enhancing a composite strength.

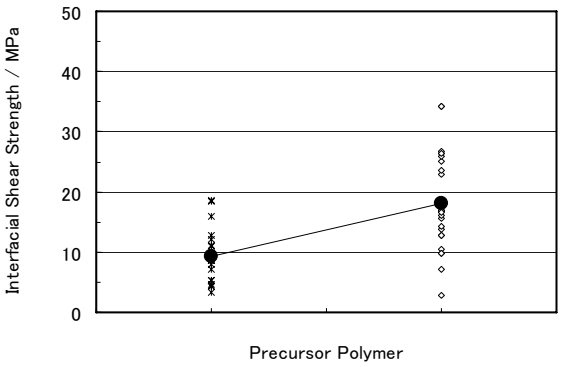

Fig. 8. Interfacial shear strength of the composites fabricated using AHPCS and PCS.

3.4 Filler Blending Rate

The maximum tensile loads of the composites to which SiC powder was added at each admixture rate in the first PIP processing are shown in Fig. 9. In comparison with the samples without filler (0 wt.%), maximum load was improved by mixing up to 30 wt.%. However, there approved to be no significant effect on tensile strength by increasing the mixture rate from 30 wt.% to 60 wt.%.

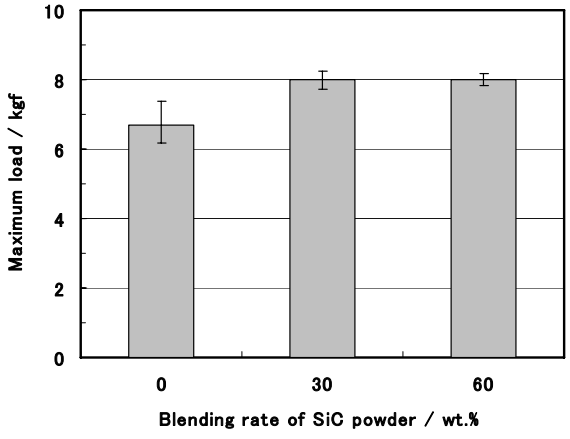

Fig. 9. Maximum loads of the composites fabricated at various filler blending rates.

The distribution of pull-out fiber length of the composites fabricated at various filler blending rate was exhibited in Fig. 10. The length of pull-out fiber increased along with the blending rate. The behavior between 30 wt.% and 60 wt.% was inconsistent with the result of tensile strength.

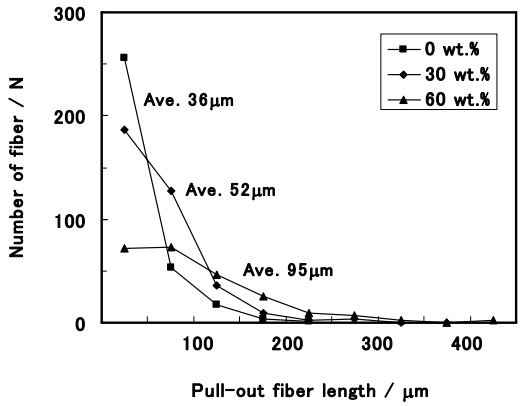

Fig. 10. Distributions of pull-out fiber length of the composites fabricated at various filler blending rates.

Fig. 11 shows the interfacial shear strengths of the composites fabricated at various filler blending rates. Compared with the interfacial shear strength obtained for 0 wt.%, that obtained for 30 wt.% decreased. Then it increased from 30 wt.% to 60 wt.%. It was found that fine filler particle distribution in a matrix much affected an interfacial property of a composite as well as density. These beharios shold be predominantly affected by the behaviors of shrinkage and crack initiation around a fiber in processing.

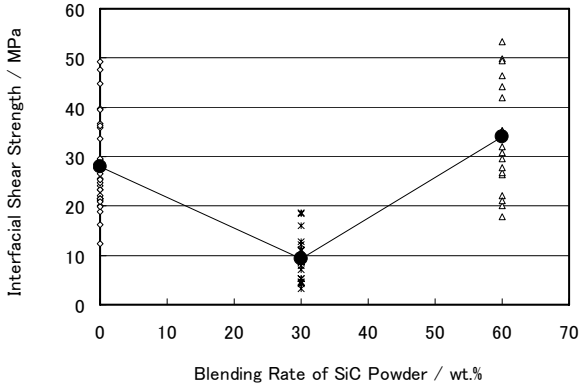

Fig. 11. Interfacial shear strengths of the composites of various filler blending rates.

For the range between 0 wt.% and 30 wt.%, crack deflection and propagation efficiency

should be predominant factor. The behabior between 30 wt.% and 60 wt.% is much complicated. Further investigation will be done.

4 Summary

The tensile property of the polymer-derived unidirectional SiC/SiC composites was experimentally evaluated. The following findings are obtained.

1) For a matrix, it was confirmed that sufficient densification by repeating the PIP processing and adding filler materials was effective for improving the tensile strength.
2) As the effect of an interface, crack deflection and propagation efficiency, load transfer capacity, and structural compensation and/or stabilization for a fiber were implied. Also it was suggested that some hundred nanometer thickness of carbon layer is sufficient in the aspect of crack deflection and propagation function.
3) Preferable average pull-out fiber length for tensile strength was found to be around 100 μm.

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