EFFECT OF FIBER SURFACE TREATMENT ON THERMAL/MECHANICAL PROPERTIES OF LIQUID SILICON INFILTRATED CERAMIC MATRIX COMPOSITES

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Abstract

C/SiC composites are widely used in high temperature applications such as high performance brake system and aerospace component due to excellent heat resistant characteristic. However, due to the relatively low thermal conductivity, it is insufficient to be applied to the cooling system of aerospace component. For this reason, the material is required to improve the thermal properties for enhance cooling system efficiency. In this paper, C/SiC composites were fabricated with liquid silicon infiltration process that consisted with 3 steps as: polymer matrix composites fabrication, polymer carbonized C/C composites and liquid silicon infiltrated C/SiC composites. To improve vertical thermal conductivity, crack formation tendency was controlled by fiber surface treatment during carbonization step. Larger segment cracks area was induced that filled with molten silicon and it act

as a vertical path for thermal conduction in composites. As increasing interfacial bond strength between fiber and matrix by nitric acid treatment, the segment crack area was increased as shown in cross sectional SEM observation Figure 1 and 2. The thermal conductivity was measured by laser flash apparatus (LFA) method and increased with segment crack area increase but 3-point flexural strength was decreased as shown in Figure 3. Though the fiber surface treatment by nitric acid could enhance interfacial bonding strength so that induced larger segment crack area with decreasing fiber/matrix micro crack, damage was occurred on fiber surface that degrade mechanical properties. From the results, the optimum surface treatment time on carbon fiber was derived to increase thermal/mechanical properties of LSIed composites.

1. Introduction

Materials made of a single ceramic have poor thermal shock and toughness. Therefore, the disadvantages were reinforced by using ceramic fiber (carbon fiber and sic fiber). This reinforced product was called Ceramic matrix composites (CMC). C/C composites, C/SiC composites and SiC/SiC composites are typical materials of CMCs. C/SiC composites have been applied to aerospace parts or high-performance brake system because of the excellent heat resistance [1-5]. Currently, various methods such as polymer infiltration and pyrolysis (PIP), chemical vapor infiltration (CVI) and liquid silicon infiltration (LSI) are being developed and applied to manufacture the C/SiC composites.The main advantages of the CVI process are the relatively low process temperatures, uncritical for the carbon fibers and disadvantages are the limited fiber content and the remaining open porosity [6].The main advantage of PIP process is the controllable matrix buildup and disadvantagesare high costs for the preceramic precursors and the long manufacturing times [7]. In contrast to CVI and PIP, where the SiC matrix buildup does not influence or damage the carbon fibers, the molten silicon is highly reactive to the C matrix as well as to the C fibers. However, the LSI process is capable of not only flat plates but also large structures of complex shapes. Due to economical and some specific technical advantages, such as high thermal conductivity and wide variability in materials composition and properties, LSI process was selected [6, 8-9].

Currently, C/SiC composites are used as aerospace parts exposed to high temperature mechanical environment.These components are often used with a cooling system. However, since the thermal conductivity characteristics of C/SiC composites are relatively low compared to the matrix (SiC, Si), additional research and development is needed to improve the cooling efficiency.

In order to improve the thermal conductivity of composites, method of increasing the segment crack fraction inside the composite material was studied.Fiber-matrix bonding is an important factor affecting the thermal/mechanical properties of composites.Lots of people have studied the mechanical properties of C/C composites with or without fiber surface treatment [10-12].E. Fitzer et al. reported that when using surface-treated carbon fibers, a strong bond to the fiber-matrix was produced [12].L. M. MANOCHA et al. reported that when using surface-treated carbon fibers, mechanical properties were reduced due to the reduction of micro cracks between fiber-matrix after carbonization [13].And the thermal conductivity is related to the fiber-matrix fraction[14].In this paper, focus on the effect of the increase of the segment crack inside the composites and the

thermal conductivity of the C/SiC composites aftere the LSI process. The increase of the segment cracks became a Si infiltration path and affected the improvement of the thermal conductivity of C/SiC composites.

2. Experiment

2.1 Preparation of C/SiC composites

The sizing of the PAN-based carbon fibers (Mitsubishi., Japan)with 2D plain weave structure was removed using nitric acid (60.0 %, OCI Company, Republic of Korea). The carbon fibers were immersed in nitric acid and placed in an oven at 110℃ for 1, 5, 15, 60 and 180 minutes, respectively. The treated fibers were laminated using a phenolic resin of resoltype(KRD-HM2, KOLON INDUSTRIES, Republic of Korea), and cured at 120℃ after vacuum bagging to produce FRP (fiber reinforced plastic). Primary carbonization (nitrogen atmosphere, 600℃) was followed by secondary carbonization (vacuum, 1000 ℃). Then, SiC matrix was formed by infiltrating silicon into C/C composites using the LSI process.

2.2. Evaluation

The density was measured by Archimedes' method, usingfivespecimens for each sample. After flexural tests, six specimens were observed by Scanning electron microscope (SEM; S-4800, HITACH Co., Japan). Then, Count-Program was used to analyzed and quantify the amount of segment cracks and micro cracks in SEM photographs. The 3 point flexural strength tests were carried out in accordance to ASTM C1341-13, using a material testing machine (Landmark, MTS, USA). The specimens were processed 1.2×4×45 mm, span length 40 mm, at room temperature, cross head speed set to 1 mm/min. Thermal conductivity of the specimens was measured by the laser flash method (LFA; 467, NETZSCH Co,. Germany). The specimens have a diameter of 12.7 mm and a thickness of 1.4 mm. The thermal conductivity measurements were carried out at 24.5, 500, 1000 and 1400℃, then 10 measurements, the average value was obtained.

3. Results and discussion

C/C composites were prepared by carbonizing FRP under nitrogen atmosphere of 600℃. Then, to analyze the area of crack in the matrix quantitatively, SEM images were evaluate using count-program. A change in the segment crack area due to the variation of the surface treatment time was shown in Figure 2. Segment crack area increased with surface treatment time. It was concluded that the amount of segment crack was increased due to the suppression and reduction of micro cracks between fiber-matrix by measuring bending strength between fiber-matrix. Thereafter, the amount of segment crack was increased. C/C composites were converted to C/SiC composites through LSI process, and density, porosity, flexural strength and thermal conductivity characteristics were evaluated. Table 1 shows the density and porosity results of C/SiC composites withvarious fiber surface treatment times. The density of the C/SiC composites was similar for all specimens except for 60 min treated specimen. The porosity of the specimens were 11~12% in all specimens and results were similar to the density. And the 180 min treated specimen has lowest value of 10.15%.

Figure 3 shows the flexural strength result of C/SiC composites and the mechanical properties were gradually reduced to 171.98MPa, 165.23MPa, 162.17MPa and 133.02MPa with fiber surface treatment time. The treated specimen for 180 minutes showed a drastic decrease of 62.83MPa. Decrease of fiber volume fractiondue to increase of segment crack fraction was also considered as a cause of decrease of physical properties.

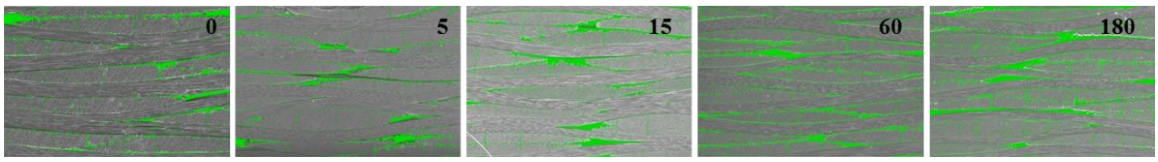

Fig 1.Image analysis result for segment crack area

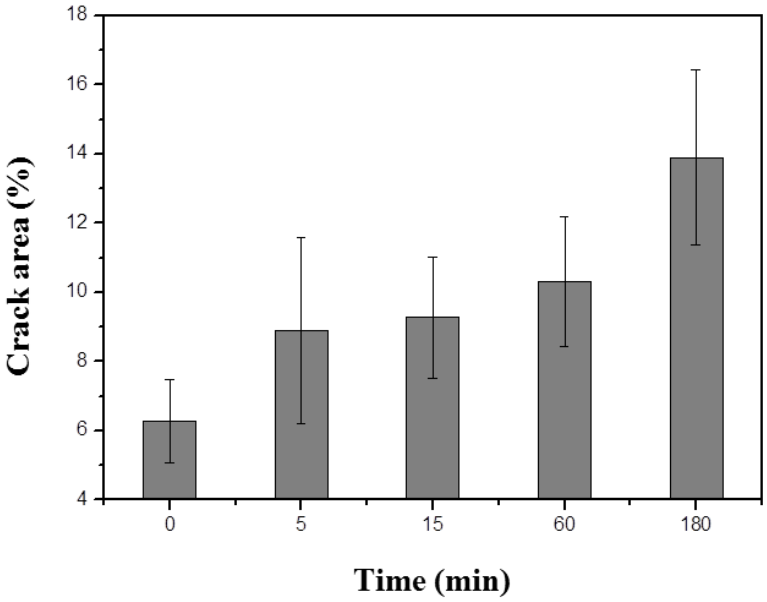

Fig 2.Change of fraction of segment cracks by surface treatment time

Time (min)	Density (g/cm^3)	Porosity (%)
0	1.82 ($\pm$0.04)	11.64 ($\pm$0.86)
5	1.81 ($\pm$0.02)	12.23 ($\pm$0.75)
15	1.80 ($\pm$0.02)	12.71 ($\pm$0.45)
60	1.73 ($\pm$0.03)	12.32 ($\pm$1.29)
180	1.82 ($\pm$0.04)	10.15 ($\pm$1.75)

Table 1.Density and porosity of C/SiC composites according to fiber treatment time

To verify this the cross section was analyzed using specimens after flexural strength test. Figure 4 shows the result of observing the cross section of the specimen. As the fiber surface treatment time increased, it was observed that the C/C region in the segment crack and this area was very clear. The specimen with short surface treatment was reacted with the infiltrated Si into the C/C region in the segment crack and confirmed that the SiC phase was formed also. During the carbonization process, the specimens with excellent surface bonding

were prevented from micro-cracking between the fiber and matrix, and the infiltration of Si into the micro cracks in segment did not occur. When the surface treatment was not performed or treated time was short, a lot of micro cracks in segment were generated during the carbonization process. Micro cracks became a channel for Si to melt infiltration.

Figure 5 shows the thermal conductivity results of the specimens prepared using differently surface treated carbonfibers. The thermal conductivity of the other specimens, except for specimens made of untreated fiber, showed similar result after 500℃. The highest thermal conductivity result was obtained with 15 min fiber surface treated specimen in each temperature range.

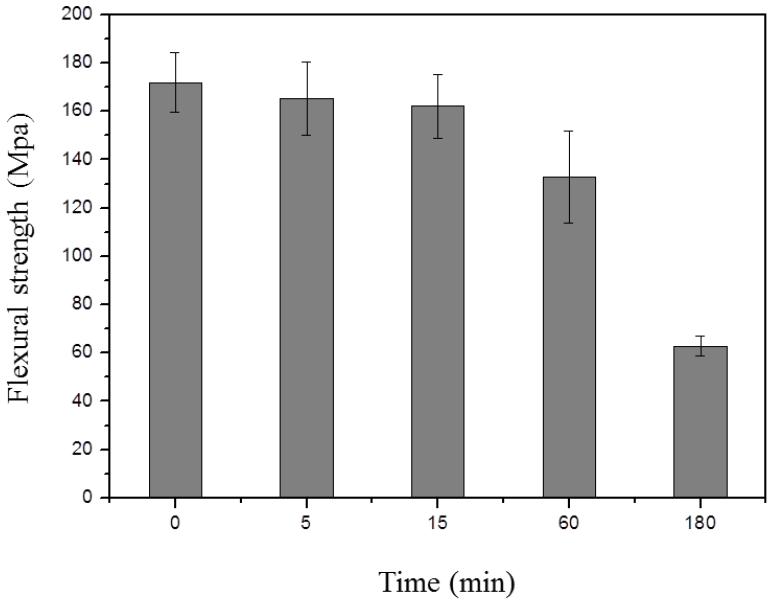

Fig 3.Flexural strength result of C/SiC composites according to fiber treatment time

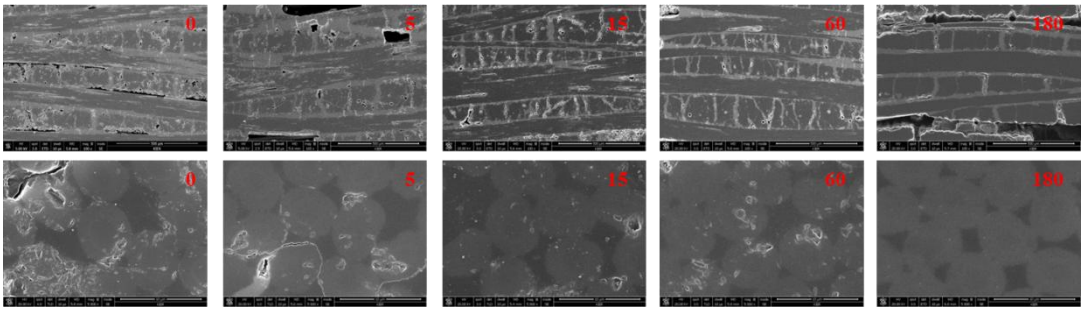

Fig 4.SEM result of C/SiC composites cross section area (top)×100 magnification, (bottom)×5000 magnification

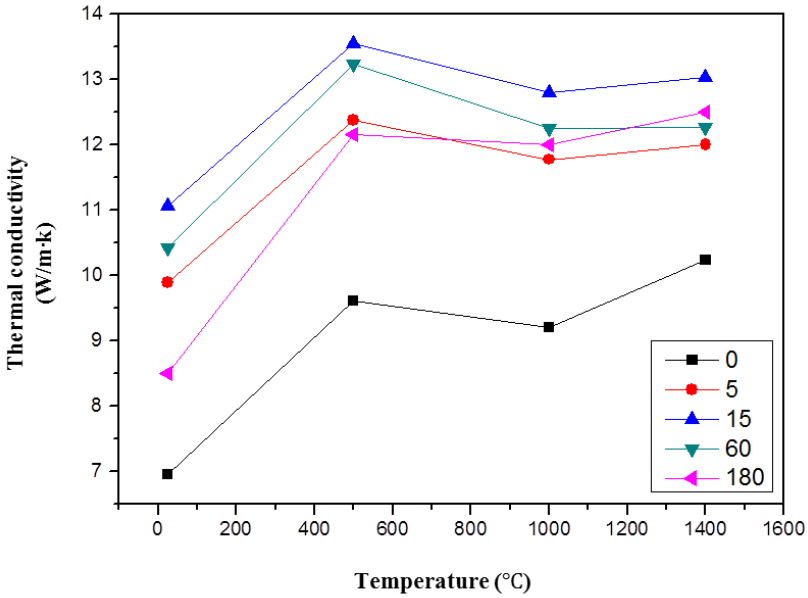

Fig 5.Thermal conductivity result of C/SiC composites according to fiber treatment time

4. Conclusion

In order to improve the thermal conductivity of the LSI processed CMC, study was conducted to control the optimum matrix fraction.

The optimum matrix fraction was obtained by controlling the amount of segment cracks within the composites. Thesegment crack area control was achieved by treating the surface of the fiber with nitric acid. The segment cracks were increased while micro cracks in each segment decreased due to enhanced bonding strength between the fiber and matrix.

The inside of the C/C area in the segment crack of CMC made with LSI is formed very clearly without infiltration of Si when the surface treatment is sufficiently performed. In the case of no surface treatment or insufficient processing time, Si infiltrates through the micro crack and is present in the segment crack.

The thermal conductivity of composite with surface treated fiber showed all higher than the untreated fiber used composite.

5. Acknowledgments

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