Carbon fiber are widely used as reinforcements in composite materials because of their excellent properties, such as low coefficient of thermal expansion, high modulus and strength, low density and relative flexibility [1]. Nevertheless, their prolonged use in oxidising environments is limited to temperatures lower than 500°C because of gasification of carbon as carbon oxides [2]. And the researches for thermal conductive properties of the carbon fiber reinforced composites have been rarely reported. Numerous materials have been studied to solve this drawback based on metals (Al, Ti) or nonmetallic elements (Si, B). Ceramics have been widely used (carbides, nitrides) [3-5], but also oxides [1, 6].

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SiC has the ability to form a SiO2 layer in an oxidizing atmosphere which fills the cracks of the layer and increases the diffusion barrier efficiency [7]. Also, carbon fiber/silicon carbide composites (C/SiC) have been extensively studied due to their high thermal conductivity (TC), good thermal shock, low thermal expansion, good high-temperature strength, high hardness and ablation resistance [8-10]. Its sufficient thermal conductivity and effective oxidation protection system gives scope for a potential application in the various industries.

In this work, SiC coating on the carbon fiber (SiC/CF nanocomposites) was obtained, through the infiltration of a SiO2 sol onto carbon fiber by dip coating method and they were carried out in a furnace under an argon flow. The effect of the coating of silicon carbide on the microstructure and thermal properties of SiC/CF composites is investigated.

2 Experimental

2.1 Preparation of SiC/CF nanocomposites
Polyacrylonitrile-based carbon fibers with a diameter of about 6–7μm, purchased from Hyundai fiber (Korea), were used in this work. Before the coating reaction, the surfaces of the carbon fibers were sized by a thermal treatment at 950°C under argon during 15 h.

The SiO2 sol was synthesized with tetraethylorthosilicate (Si(OC2H5)4, TEOS) [99% Aldrich]. TEOS was mixed with isopropanol (C3H8O) [99+% Aldrich] as solvent, distilled water (H2O) for hydrolysis and hydrochloric acid (HCl) [37% Daejung] as catalyst at 70°C for 80 min.

Carbon fibers were impregnated with the SiO2 sol by dip coating method. A drying was then carried on in air at room temperature during 30 min. The oxide layer was densified by a first thermal treatment up to 1000°C with a heating rate of 5°C /min, followed by an isothermal step at 1000°C during 30 min. The final carbide layer was formed during a second thermal treatment at 1450°C with a heating rate of 5°C /min. The as-resulting solid was oxidized at 800°C in air to remove the unreacted carbon: a SiO2/SiC or SiC layer was observed.

2.2 Preparation of CF/epoxy composite for thermal conductivity measurement
The CF/epoxy composites were prepared by solution blending, casting, and pressing method, which involved as follow:
Uncoated carbon fibers and SiC coated carbon fibers were mixed in epoxy resin as the binder for measurement of thermal conductivity. The mixture was stirred for homogenization and sonicated. Subsequently, the mixture was kept in a vacuum oven to get rid of air bubbles and then the mixture was poured into a mold and the whole system was placed in an oven. The mixture was precured at 100°C. Finally, the CF/epoxy composites were pressed at 120°C for 1 h and cooling to room temperature, then demoulding.

2.3 Characterization of SiC/CF nanocomposites

The phases present in the SiC-coated carbon fibers were identified by an X-ray diffraction (XRD, X’Pert Philips instrument) using Cu-Kα radiation (λ=0.154nm). Scanning electron microscope (SEM, SM300, TOP CON) was used to examine surface morphology and to measure the coating thickness of the coated fibers. Thermal oxidation resistance of uncoated and coated fibers was investigated using a thermo-gravimetric analysis (TGA, TA Instrument Q20). The thermal diffusivity of composites was measured using a laser flash apparatus (LFA, LFA427, NETZSCH). The diffusivity data were then converted into thermal conductivity values by the product of the diffusivity, sample bulk density and specific heat.

3 Results and discussion

3.1 Characterization of SiC/CF nanocomposites prepared by carbothermal reaction

In most cases, the overall reaction between the carbon and the oxide (carbo-reduction) reaction can be written as follows:

$$3C + MeO_2 \rightarrow MeC + 2CO$$  \hspace{1cm} (1)

Nevertheless, the mechanism of the reaction depends on the final carbide formed. In the case of SiC, the reaction proceeds via two stages. A reaction between silica and carbon or between silica and silicon carbide leads to the formation of SiO through reaction (2) and (3):

$$C + SiO_2 \rightarrow SiO + CO$$  \hspace{1cm} (2)

$$SiC + SiO_2 \rightarrow 2SiO + CO$$  \hspace{1cm} (3)

Reaction (2) will occur at the beginning of the reaction. Then, when the conversion yield increases, the interface between SiO2 and carbon disappear and reaction (3) becomes the main SiO formation path. SiO cannot undergo further reduction at the SiO2/SiC interface, so the only possibility is the diffusion of SiO in the SiC layer and the formation of SiC at the C/SiC interface [11]:

$$SiO + 2C \rightarrow SiC + CO$$  \hspace{1cm} (4)

Fig. 2 shows the XRD spectrum of materials prepared by carbothermal reduction. Two different phases, namely, SiC and C, are identified. The XRD spectra of the SiC coated carbon fiber obtained at
1450 °C. The bands of diffraction at 2θ = 35.59 and 2θ = 59.9 correspond to the (111) and (220) plans of the β type SiC according to the Ramsdell notation, respectively. This suggests that a SiC coating may have been formed on the carbon fibers. SEM images of the coatings obtained with one layer of SiO2/SiC or SiC are presented in Fig. 3a, respectively. After removal of the unreacted carbon, the tubular morphology of the initial carbon fibre is kept, indicating that the carbon surface was uniformly covered with the silicon carbide. However, the SiC surface exhibits a strong rugosity. The diameter of the tubules is equal to ca.6 um; the thickness of the wall is less than 1 um. The C, Si and O elements peaks, as Fig. 3(b) shown, prove that the SiO2/SiC or SiC coatings exist in the surface of carbon fibers.

3.2 Thermal properties of SiC/CF nanocomposites

In order to further reveal the influence of the SiC coating on the oxidation resistance, TGA was carried out both for uncoated and SiC-coated carbon fibers. Fig. 4 shows the TGA curves of uncoated carbon fibers and SiC-coated carbon fibres prepared at 1450 °C for 1 h. As can be seen from curve, the uncoated fibers begin to oxidise at a temperature of about 500 °C and are burnt away completely at about 700 °C. The weight loss of SiC-coated carbon fibers begins at around 700 °C, increasing the onset temperature by about 200 °C in comparison to uncoated carbon fibres, and there is a 10% (wt.) residue. CF/epoxy composites were prepared by uncoated and SiC coated carbon fiber using a mixing-hot pressing method to improve thermal conductivity. CF and SiC coated CF with the different contents can be used as a filler in the polymer composites system. The specimens of the epoxy resin, the composites with uncoated CF 6% (wt.), 50% (wt.) and the composites with SiC coated CF 6% (wt.), 50% (wt.) were prepared. The thermal conductivity of SiC coated CF are larger than that of uncoated CF single (shown in Fig 5.). Disadvantage of low thermal conductivity of epoxy composites could to be improve using SiC coated CF with high thermal conductivity.
4 Conclusion

The SiC coatings on carbon fiber surfaces were successfully fabricated by sol–gel method. The results indicate that the carbon surface can be uniformly covered with the silicon carbide. After removal of the unreacted carbon, the diameter of the tubules is equal to ca. 6 um; the thickness of the wall is less than 1 um. In comparison to uncoated carbon fibers, SiC-coated carbon fibers show a good oxidation resistance due to the ability to form a SiO2 layer in an oxidizing atmosphere of the SiC layer. And the study on the CF/epoxy composites prepared by uncoated and SiC coated carbon fiber using a mixing-hot pressing method was performed to improve thermal conductivity. The thermal conductivity of the composites with SiC coated CF 50% (wt.) are larger than that of uncoated CF due to the synergy effect of CF and SiC with high thermal conductivity.

References