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Tensile behavior of polyethylene fiber composites with polyethylene fiber surface-modified using ion irradiation

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It is well-known that for fiber-reinforced composites, the condition of the interface between fiber and matrix plays an important role because the interface is responsible for the load transfer between fiber and matrix. Accordingly, various methods have been developed for the surface modification of polymer fibers to improve the interfacial adhesion between fibers and matrix [1–5]. At present, ion-assisted reaction is prominent because the chemical and physical properties of polymer fibers can be easily tailored by the method. For instance, Koh *et al.* [6] have modified the surface of polytetrafluoroethylene (PTFE) using the ion-assisted reaction method to improve adhesion strength between the PTFE and aluminum. Their result showed that the adhesion strength of aluminum/PTFE was improved 375% when the PTFE was surface-treated at the Ar^+ ion dose of 1×10^{16} ions/cm^2 in an oxygen environment.

In recent years, we applied the ion-assisted reaction method to improve fracture toughness of graphite/epoxy composites [7]. That is, we made surface-treatment of graphite/epoxy prepregs using Ar^+ ion irradiation in an oxygen environment and determined the treatment effect on the mode I fracture toughness of laminated graphite/epoxy composites. The mode I fracture toughness of the surface-treated case was found to be 24% higher than that of untreated (as-received) case. The improvement was attributed to increased bonding strength between plies and possibly also to increased fiber/epoxy adhesion in the vicinity of the ply-to-ply interface.

The aim of this study, therefore, is to investigate the surface modification of polyethylene fibers using Ar^+ ion-assisted reaction method and its effect on the tensile behavior of polyethylene fiber composites.

Woven type UHMWPE (ultra high molecular weight polyethylene) fibers (Honeywell Inc, Spectra 900) were prepared and cut into 200 × 200 mm pieces for Ar^+ ion irradiation treatment. The Ar^+ ion irradiation treatment of the fibers was carried out in an oxygen environment. The treatment was made with a cold hollow cathode ion gun (50 mm diameter) in a vacuum chamber that was maintained below 10^{-2} Pa. Argon gas was injected into the ion gun at a constant flow rate, 8×10^{-6} m^3/min, to produce the Ar^+ ion beam. Oxygen gas as an environmental gas was blown onto the surface of the Spectra fibers at a flow rate of 4×10^{-6} m^3/min during the irradiation. The Ar^+ ion dose and the ion beam energy applied were 1×10^{16} ions/cm^2 and 1 keV, respectively. The reason we chose this ion energy is that it was shown in the previous study that the surface modification of polycarbonate with 1 keV energy Ar^+ ion beam irradiation in an oxygen environment produced much better wettability than the conventional surface ion beam modification methods using a few MeV energy [8]. Also, the optimal ion dose in the treatment of polymer material is known to be in a range of 1×10^{15} ions/cm^2 to 1×10^{17} ions/cm^2 [6, 9]. The ion dose was controlled by discharge current and measured by a Faraday cup.

Three-plied polyethylene-fiber/vinylester composites were used for tensile tests. In order to investigate the effect of Ar^+ ion irradiation on the tensile behavior of polyethylene-fiber/vinylester composites, the untreated (as-received) polyethylene-fiber/vinylester composite panels and the surface-modified polyethylene-fiber/vinylester composite panels by ion irradiation were fabricated at a same time in an autoclave under 0.5 MPa of pressure at 130 °C for two hours. The cured thickness of surface-modified panel was equal to that of untreated panel and was about 1 mm. The panels were machined as dogbone tensile specimens, and Fig. 1 shows a schematic diagram of tensile test specimen. Displacement-controlled (2 mm/min) tensile tests were performed using a universal test machine, and the elongation was measured using an extensometer. Six tensile tests were carried out each for two cases of tensile specimens to determine the reproducibility of the results.

The load-displacement curve of surface-modified polyethylene-fiber/vinylester composite specimen was compared with that of untreated polyethylene-fiber/vinylester composite specimen. Fig. 2 shows the typical load-displacement curves obtained in tensile tests for both specimens. It can be seen in the figure that for both cases, the load increased nonlinearly with displacement at the early stage, and the load increased almost linearly with displacement up to the maximum load. Then, the load varied in a serrated fashion with further displacement up to fracture. The serrated

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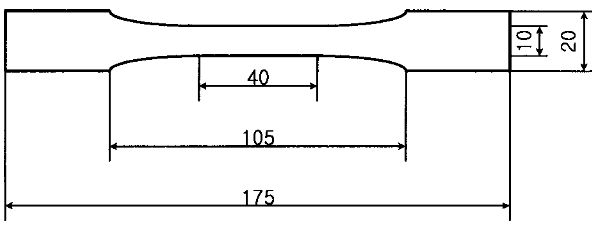

Figure 1 Schematic diagram of dogbone tensile polyethylene-fiber/vinylester composite specimen (unit: mm).

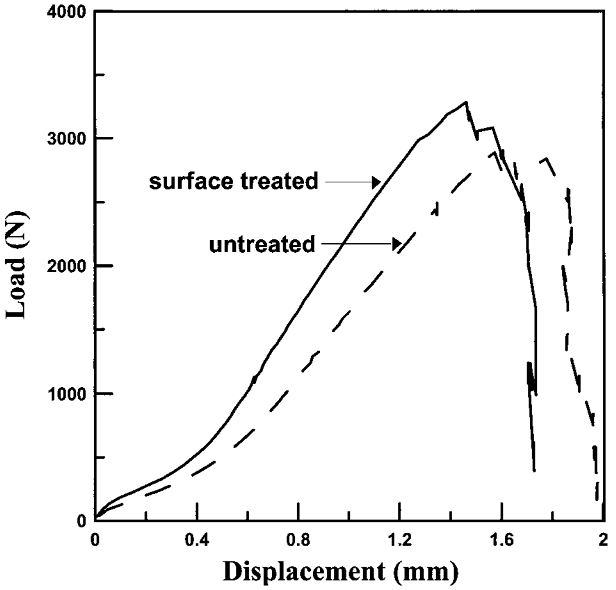

Figure 2 Typical tensile load-displacement curves of untreated and surface-modified of polyethylene-fiber/vinylester composites.

TABLE I Comparison of test results between surface-modified polyethylene-fiber/vinylester composite specimen and untreated polyethylene-fiber/vinylester composite specimen

	Maximum load (N)	Stiffness (N/mm)	% increase of maximum load	% increase of stiffness
Untreated specimen	2950	2390	–	–
Surface-modified specimen	3451	2916	17.0	22.0

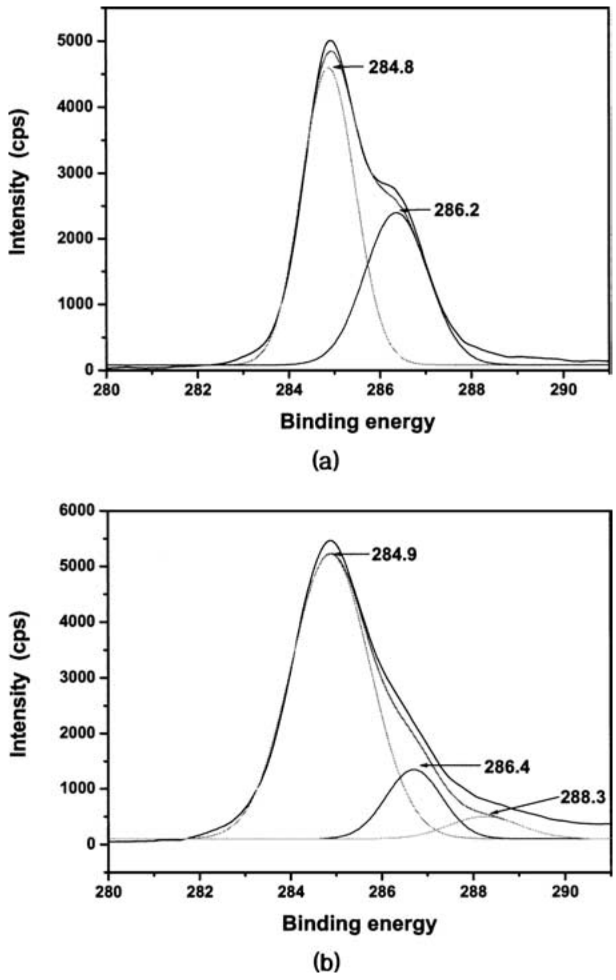

Figure 3 Evolution of Cl s spectra of polyethylene fibers: (a) untreated and (b) surface-modified by Ar$^+$ ion irradiation in an oxygen environment.

behavior of load with displacement indicates that the maximum load corresponds to the load for the onset of delamination.

The tensile strength of the surface-modified polyethylene-fiber/vinylester composite specimen was compared with that of the untreated polyethylene-fiber/vinylester composite specimen to determine the effect of surface modification on the tensile strength. The tensile strength in each specimen was determined as the maximum load in the corresponding load-displacement curve. The tensile strength is significantly improved by the surface modification of the polyethylene fibers using an ion-assisted reaction method. The average tensile strength of the surface-modified composite case was about 17% higher than that of the untreated composite case. The tensile stiffness of the surface-modified specimen was also compared with that of the untreated specimen. Similar to the result of tensile strength, the tensile stiffness is significantly improved by the surface modification of polyethylene fibers using an ion-assisted reaction method. The average tensile stiffness of surface-modified specimen was about 22% higher than that of untreated specimen. Table I shows a comparison of test results between surface-modified polyethylene-fiber/vinylester composite specimen and untreated polyethylene-fiber/vinylester composite specimen.

X-ray photoelectron spectrometry (XPS) analysis was performed to examine the chemical bond environment for the modified surface of polyethylene fibers by Ar$^+$ ion irradiation in an oxygen environment. Fig. 3 shows Cls core level spectra of the untreated polyethylene fibers (Fig. 3a) and the surface-modified polyethylene fibers (Fig. 3b). As shown in Fig. 3a, the C—C peak was located at 284.8 eV and C—O at 286.2 eV for untreated polyethylene fibers. The signal corresponding to the C=O bond was so small that it was neglected. In the case of surface-modified polyethylene fibers (Fig. 3b). binding energies of carbon peaks are shifted to higher energy level. The ratio of each peak area is also changed compared with that of the untreated polyethylene fibers. Particularly, a hydrophilic bond of C=O is newly formed. This can be explained that unstable chains generated by Ar$^+$ ion irradiation react with oxygen and create a hydrophilic bond of C=O (carbonyl group).

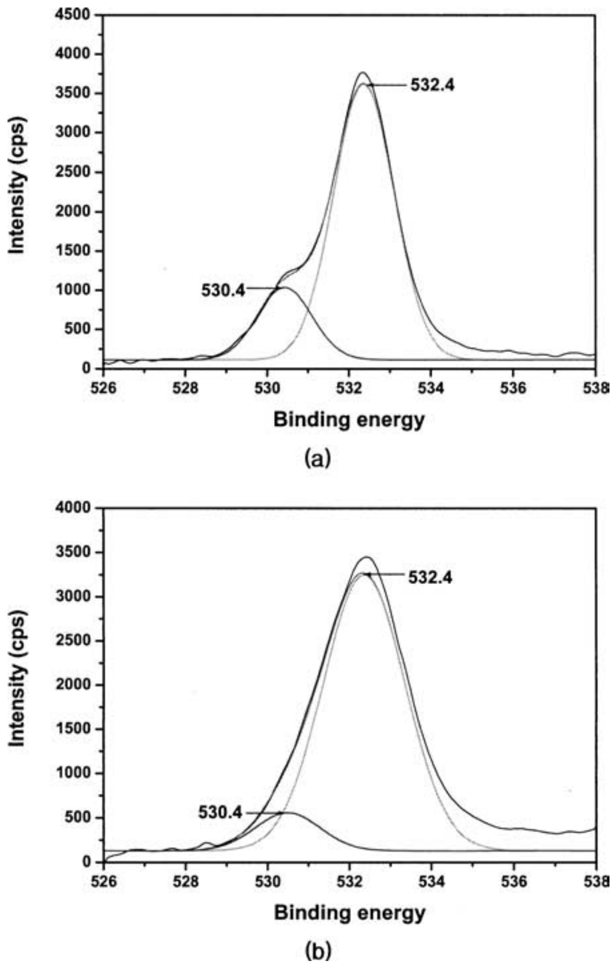

Figure 4 Evolution of Ol s spectra of polyethylene fibers: (a) untreated and (b) surface-modified by Ar$^+$ ion irradiation in an oxygen environment.

Similar results were obtained in the analysis of Ols spectra. Fig. 4 shows Ols core level spectra of the untreated polyethylene fibers (Fig. 4a) and the surface-modified polyethylene fibers (Fig. 4b). For the untreated polyethylene fibers, the peak positions of C—O and C═O were located at 530.4 and 532.4 eV, respectively. For the surface-modified case, the peak positions of C—O and C═O are the same as those of the untreated case. However, the area ratio and intensity of C═O peak significantly increased compared with those of the untreated case. The area ratio of C—O and C═O was 20 and 80% for untreated polyethylene fibers while the ratio was 8 and 92% for surface-modified polyethylene fibers. This indicates that more C═O group are formed by the surface modification using the ion-assisted reaction method.

In conclusion, it has been shown that the tensile behavior (tensile stiffness, tensile strength) of polyethylene-fiber/vinylester composites can be improved if polyethylene fibers are surface-modified by Ar$^+$ ion irradiation in an oxygen environment. The improvement of tensile behavior is attributed to the formation of hydrophilic functional groups on the surfaces of polyethylene fibers. In particular, the C═O functional group plays an important role in enhancing the tensile properties of polyethylene-fiber/vinylester compósities.

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