Carbon Nanotube Modification Using Gum Arabic and Its Effect on the Dispersion and Tensile Properties of Carbon Nanotubes/Epoxy Nanocomposites

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In this study, the effects of a MWCNT treatment on the dispersion of MWCNTs in aqueous solution and the tensile properties of MWCNT/epoxy nanocomposites were investigated. MWCNTs were treated using acid and gum arabic, and MWCNT/epoxy nanocomposites were fabricated with 0.3 wt.% unmodified, oxidized and gum-treated MWCNTs. The dispersion states of the unmodified, oxidized, and Gum-treated MWCNTs were characterized in distilled water. The tensile strengths and elastic modulus of the three nanocomposites were determined and compared. The results indicated that the gum treatment produced better dispersion of the MWCNTs in distilled water and that gum-treated MWCNT/epoxy nanocomposites had a better tensile strength and elastic modulus than did the unmodified and acid-treated MWCNT/epoxy nanocomposites. Scanning electron microscope examination of the fracture surface showed that the improved tensile properties of the gum-treated MWCNT/epoxy nanocomposites were attributed to the improved dispersion of MWCNTs in the epoxy and to interfacial bonding between nanotubes and the epoxy matrix.

Keywords: MWCNTs, Gum Arabic, Epoxy, Tensile Strength, Fracture Surface.

1. INTRODUCTION

It is well-known that multiwall carbon nanotubes (MWCNTs) should be dispersed uniformly in the matrix to function properly as reinforcing nanomaterials. Accordingly, a number of studies have been performed to develop methods for homogeneous dispersion of MWCNTs into the polymer matrix. One method is an oxidative process utilizing strong acids in which hydroxyl and carboxylic acid moieties are created on MWCNTs. The oxidized MWCNTs show better solubility and can form electrostatically stabilized colloidal dispersions in water as well as in alcohols.1–3

The silanization of MWCNTs is another preferred method used to enhance the dispersion and interfacial adhesion between the MWCNTs and the epoxy.4,5 However, the acid-treatment and silane treatment of MWCNTs are chemical functionalizations. New attempts have been made to achieve environmentally-friendly modification of MWCNTs using natural materials.

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For instance, Bandyopadhyaya et al. reported the formation of homogeneous dispersions of individual MWCNTs in gum arabic (GA) solution and demonstrated that adsorption of GA led to disruption of the inter-tube interactions in the crystalline ropes.6 At present, very few studies have been performed on the effect of GA modification of MWCNTs on the tensile behavior of MWCNT/polymer nanocomposites.

For the present study, we investigated the effect of MWCNT treatment with GA on the tensile properties of MWCNT/epoxy nanocomposites. Tensile tests were performed on the untreated, acid-treated, and GA-treated MWCNT/epoxy nanocomposites. SEM examinations of the fracture surfaces were performed to investigate the effect of GA treatment on dispersion in the epoxy matrix.

2. EXPERIMENTAL DETAILS

The carbon nanotubes used in this study were MWCNTs synthesized via catalytic chemical vapor deposition (CM-95, Iijin Nanotech, Korea). The following reagents were used without further purification: nitric acid (60–62%,...
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Junsei, Japan), sulfuric acid (95%, Junsei, Japan), ethanol (99%, Aldrich, USA), and distilled water. Gum arabic (Junsei, Japan) was used as a surface active agent, the epoxy used was 118.2 g/eq of diglycidyl ether of bisphenol A (Kukdo Chemical, Korea), and the hardner was 60 g/eq of polyamidoamine (Kukdo Chemical, Korea). The oxidized treatment of MWCNTs (O-MWCNTs) was performed as follows. Three grams of untreated MWCNTs (U-MWCNTs) were dispersed in 300 ml of concentrated H₂SO₄/HNO₃ (3:2, v/v) solution at 50°C and stirred for 20 h. The solution was filtered with distilled water until a neutral pH was obtained to eliminate mixing an acid solution. The resulting oxidized MWCNTs were then dried under vacuum at 80°C for 12 h. The Gum Arabic modification of MWCNTs (G-MWCNTs) was performed as follows. Ten grams of Gum Arabic (GA) was dissolved in 200 ml of distilled water and stirred for 30 min. Then, two grams of untreated CNTs was dispersed in the GA solution and stirred for 1 h. The Gum treated MWCNTs (G-MWCNTs) were separated by filtration using ethanol and dried under vacuum at 80°C for 12 h.

MWCNT/epoxy nanocomposites were fabricated as follows: U-MWCNTs, O-MWCNTs, and G-MWCNTs were dispersed in the ethanol solution and ultrasonication was performed for 5 min. After sonication, 0.3 wt.% each of U-MWCNTs, O-MWCNTs, and G-MWCNTs were mixed with the epoxy resin and then stirred for approximately 2 h at 80°C to completely evaporate the ethanol. After removing the ethanol, a hardener was added (epoxy and hardener at a 2:1 ratio) and the mixture poured into a teflon mold. The mixture was de-gassed in a vacuum oven at 760 mm Hg for 30 min and hardened in an oven at 120°C for 4 h. The fabricated nanocomposite plates were machined as tensile specimens and the dimensions of the gauge section were 5 mm thick × 80 mm.

Fig. 1. Sedimentation of dispersed MWCNTs (A: unmodified MWCNTs, B: oxidized MWCNTs, C: Gum treated MWCNTs) in distilled water (a) 10 min after ultrasonication, (b) 240 h after ultrasonication.

Fig. 2. TEM images of dispersion states: (a) unmodified MWCNTs, (b) oxidized MWCNTs, (c) Gum treated MWCNTs.
long × 15 mm wide. Tensile tests were performed in a universal test machine according to the ASTM D 638. At least three tensile tests were performed for the three types of nanocomposites.

3. RESULTS AND DISCUSSION

Unmodified, Acid-treated, and Gum-treated MWCNTs (U-MWCNTs, O-MWCNTs, and G-MWCNTs) were dispersed in distilled water via ultrasonication to determine the effect of Gum treatment on dispersion in the aqueous solution. Figure 1 shows the dispersion states of the U-MWCNTs, O-MWCNTs, and G-MWCNTs observed at times of 10 min and 240 h. As shown in Figure 1(a), U-MWCNTs, O-MWCNTs, and G-MWCNTs showed good dispersion in distilled water after 10 min of ultrasonication. However, the U-MWCNTs gradually settled due to their agglomeration and hydrophobic nature, whereas the O-MWCNTs and G-MWCNTs exhibited good suspension stability even after 240 h of sonication.

This agrees well with the results in the Ref. [8]. According to Bagheri et al., Gum Arabic adsorbs to the agglomerated MWCNTs and works as a repulsive force. Thus, the overall potential of the intertubes becomes repulsive and the dispersion of the MWCNTs becomes thermodynamically stable.

The microscopic structures of MWCNTs with different surface modifications were investigated using TEM. Figure 2 presents TEM images of U-MWCNTs, O-MWCNTs, and G-MWCNTs. As shown in Figure 2(a), the U-MWCNTs were severely agglomerated and their end tips were closed, which are the features of unmodified MWCNTs.

For O-MWCNTs, as shown in Figure 2(b), the agglomeration of the carbon nanotubes was reduced and the end tips of many carbon nanotubes were open, which enables the generation of functional groups at the open ends. Similar dispersion results were obtained for Gum-treated MWCNTs. As shown in Figure 2(c), the dispersion of the nanotubes was significantly improved compared to that of the U-MWCNTs. Figure 2 also shows that the MWCNTs retained their external average diameter of 10–15 nm after oxidation and Gum treatment, which is in good agreement with the suspension stabilities described in Figure 1. Figure 3 shows the comparison of the tensile strengths of unmodified, acid-treated, and Gum-treated MWCNT/epoxy nanocomposites. The tensile strength of the G-MWCNT/epoxy nanocomposites was 22% higher, and that of the O-MWCNT/epoxy nanocomposites was 15% higher than that of the U-MWCNT/epoxy nanocomposites. The fracture surfaces of the U-MWCNT, O-MWCNT, and G-MWCNT/epoxy nanocomposites were examined by SEM. Figure 5 shows the comparison of the fracture surfaces of the MWCNT/epoxy nanocomposites. In the U-MWCNT/epoxy nanocomposites (Fig. 5(a)), the carbon nanotubes curled, entangled, and pulled out from the epoxy matrix, which indicates poor dispersibility and weak interfacial bonding between the U-MWCNTs and the epoxy matrix. However, as can be observed from Figures 5(b) and (c), the oxidation and Gum treatment of the MWCNTs increased the dispersibility. In both cases, the MWCNTs were tightly held to the epoxy matrix and

![Fig. 3.](image1.png) **Fig. 3.** Comparison of tensile strength of unmodified, oxidized, and Gum treated MWCNT/epoxy nanocomposites.

![Fig. 4.](image2.png) **Fig. 4.** Comparison of elastic modulus of unmodified, oxidized, and Gum treated MWCNT/epoxy nanocomposites.
many surface cleavages were observed, which indicates an improvement in the interfacial bonding between functionalized MWCNTs and the epoxy matrix. However, it can be observed in the O-MWCNT/epoxy nanocomposites that some nanotubes were pulled out from the matrix, which shows that the stress transfer from the matrix to the O-MWCNTs was lower than to the G-MWCNTs. The uniform dispersion of nanotubes restricted the mobility of polymer chains under loading, and improved the modulus and strength of the MWCNT/epoxy nanocomposites.

4. CONCLUSION

It was found that Gum-treated MWCNTs showed good dispersion stability even after 240 h of ultrasonication, but exhibited little difference in dispersion from the acid-treated MWCNTs. The improved dispersion stability of Gum-treated MWCNTs was due to their enhanced negative ion abilities due to the functionalization effect on their surfaces. TEM analysis showed that little damage occurred on Gum-treated MWCNTs compared to acid-treated MWCNTs. The tensile strength...
and modulus of G-MWCNT/epoxy nanocomposites were improved by 22% and 29%, respectively, compared to the U-MWCNT/epoxy nanocomposites, whereas the improvements were 15% and 13% respectively, over O-MWCNT/epoxy nanocomposites. This enhancement is attributed to the good dispersibility and strong interfacial bonding energy between the functionalized MWCNTs and the epoxy matrix.

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References and Notes

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