In-situ 중합법에 의한 Polyimide/Carbon Nanotube 복합재료의 제조 및 특성

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Preparation and Characterization of Polyimide/Carbon Nanotube Composites by in-situ Polymerization

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1. Introduction

Polyimides (PI) are widely used in applications ranging from microelectronics to aerospace. Due to their insulating nature, significant accumulation of electrostatic charge may result on their surface, causing local heating and premature degradation to electronic components or space structures.

Over the past decade, several publications have been made in fabrication and characterization of CNT nanocomposites [1,2]. Unfortunately, it is difficult to draw definite conclusions about electrical conductivity from these published studies because the reported levels of CNT loading required to achieve a percolation concentration vary widely, ranging from less than 1 to over 10%.

Difficulty in obtaining homogeneously dispersed CNT nanocomposites arises from the non-reactive nature of the CNT surface and the unavoidable bundle formation due to van der Waals attraction during synthesis. Most published work has focused on chemical modification of the CNT to improve dispersion through functionalizaton oxidization, melt mixing, and compounding. However, successful dispersion remains elusive.

Therefore, this work seeks to address these percolation issues through a combination of experimental methods and describes a multiwall carbon nanotubes (MWNTs)-reinforced polyimide composites with a level of electrical conductivity sufficient to permit electrostatic discharge, as well as mechanical and thermal properties superior to that of the base polymer.

2. Experimental

2.1. Materials and sample preparation

Nanotubes (MWNTs) used in this work were manufactured by CVD process (CVD MWNTs, supplied from iljin nanotech Co. of Korea, degree of purity is >90-94%, length: 10-50 ㎛, diameter: 10-20 nm). An aromatic colorless polyimide (PI) was selected as a polymer matrix. The MWNTs/PI composites were prepared by in situ polymerization under sonication. MWNTs dispersed in anhydrous dimethyl formamide (DMF) was used as a solvent for the poly(amic acid) synthesis. The MWNTs-PI solution was cast onto a glass plate and dried in a dry air-flowing chamber. Subsequently, the dried tack-free film was thermally cured in an air-circulating oven to obtain solvent-free freestanding MWNT-polyimide film. A series of MWNTs-PI nanocomposite films were prepared with MWNT concentrations ranging from 0.01 to 1.0 wt.%.

2.2. Measurements

DC volume and surface conductivities of the composites were evaluated according to ASTM D257 using a resistivity test fixture and an electrometer.

3. Results and Discussion

MWNTs-PI composites are prepared by in situ polymerization in the presence of sonication. No significant change in the glass transition temperature of the MWNTs-PI composites is observed, indicating that the polyimide condensation reaction is not adversely affected by the incorporation of MWNT and sonication. TEM picture reveals that thin MWNT bundles are dispersed uniformly throughout the whole polymer matrix, as shown in Fig. 1.

Fig. 2 shows the DC volume conductivity of MWNTs-PI composite film as a function of MWNT concentration. The conductivity of the pristine polyimide is about 6.3×10^{-18} S/cm. A sharp increase of the conductivity value is observed between 0.02 and 0.1 wt.%, where the conductivity changes from 3×10^{-17} to 1.6×10^{-8} S/cm. At loading levels in excess of 0.1 wt.%, the conductivity increases only moderately. At 0.5 wt.%, the conductivity is 3×10^{-7} S/cm, ten orders of magnitude higher than the value at 0.02 wt.%. This behavior is indicative of a percolation transition. Percolation theory predicts that there is a critical concentration or percolation threshold at which a conductive path is formed in the composites causing the materials to convert from a capacitor to a conductor. Fig. 2 indicates that the percolation threshold for this material resides between 0.02 and 0.1 wt.%.

4. References

1.Ray H. Baughman, Anvar A. Zakhidov, and Walt A. de Heer, Carbon Nanotubes-the Route Toward Applications, *Science*, **297**, pp.787-792(2002).

2.M. K. Seo and S. J. Park, Influence of Fluorination on Surface Characteristics of Carbon Nanotubes, *J. Phys. Chem.*, submitted.

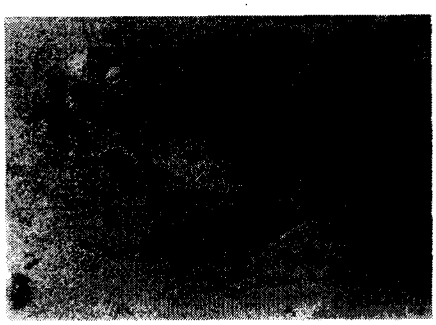

Fig. 1. TEM image of MWNTs embedded in PI resins.

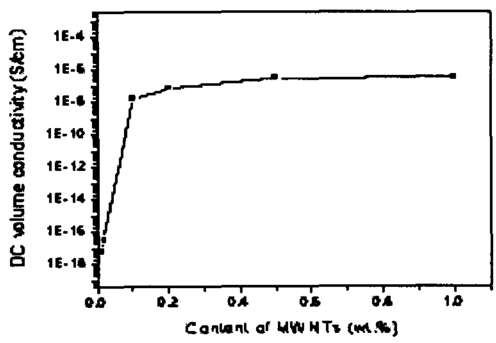

Fig. 2. DC conductivity of MWNTs-PI composites as a function of MWNT content.