1 Introduction

Carbon nanotube(CNT)-based polymer composites have shown to exhibit piezoresistive behavior, i.e., their resistivity changes when subjected to mechanical strains [1,2]. Nanocomposites are sensitive to small strain and the piezoresistive behavior is fairly reversible within certain strain ranges [1].

This study investigates multi-walled carbon nanotube(MWCNT)- and exfoliated graphite nanoplatelet(xGnP)-polymer composites as sensing materials by monitoring the electrical resistance of the sensors under flexural loading. The use of electrical resistance has been explored for damage detection in CNT composite parts [3,4] with promising results. This work focuses on MWCNT- and xGnP-polymer composite sensors as stand-alone devices that can be affixed, embedded or otherwise integrated into existing structures. The sensing performance was studied by bonding the sheet-type nanocomposite sensor to a substrate and subjecting it to three-point bending. In addition, nanocomposite sheets with mechanically aligned MWCNTs via extrusion were used to as anisotropic strain sensors with pre-defined orientations that allow tailored sensitivity in different directions.

2 Experimental

2.1 Materials and Nanocomposite Fabrication

The thermal CVD-grown MWCNTs (CM-100) used in this study were provided by Hanwha Nanotech (Incheon, Korea). Two types of xGnP(xGnP-M-5 and xGnP-M-15) with sizes of approximately 5 and 15 microns, respectively, and individual nanoplatelet thicknesses of 6-8 nm were purchased from XG Sciences (East Lansing, MI, U.S.A.). The polymer matrix used was poly(methyl methacrylate) (PMMA) produced by Evonik Industries. PMMA pellets were dissolved in chloroform at a concentration of 1 g/mL using a mechanical stirrer. A weighted amount of carbon nanomaterial (CNM) was added to the solution and sonicated for 70 min (10 min at high power followed by 60 min at low power) using a S-4000 horn sonicator (manufactured by Qsonica). Solvent was subsequently removed by air-dry, then by vacuum oven at 60°C for 3 hours to produce PMMA-CNMs composites. A nanocomposite film was fabricated by hot pressing the solution-cast nanocomposite mixture in a 10-ton hydraulic Carver press (Wabash, IN). Stainless steel shim stock was used to produce 100 mm by 100 mm films with a constant thickness of approximately 100 µm. The nanocomposite films were prepared at 0.5, 0.8, 1, 2, 4, 6, and 10 wt%. A neat PMMA film was made as a control sample.

The coagulation method was also employed to fabricate nanocomposite films. In the coagulation method, the procedure described by Du et al. [5] was used, in which the suspension of PMMA-CNMs mixture dissolved in dimethylformamide (DMF) was dripped into a large amount of distilled water. The process resulted in precipitation of PMMA molecular chains, which, in effect, entrapped the CNMs, thus locking their positions and preventing reaggregation. The resulting solid nanocomposite was dried in a vacuum oven, and was subsequently hot pressed into 100 mm by 100 mm sheets as described above.

In order to investigate the effect of CNT alignment on the sensing behavior of the nanocomposites, an
laboratory-scale extruder (RCP-0625 manufactured by Randcastle, Cedar Grove, NJ; single-screw; vertical-type; screw diameter = 16 mm; L:D = 36:1) was used to produce continuous 76.2-mm-wide nanocomposite films.

2.3 Piezoresistivity under Flexural Loading

A 5-mm-thick PMMA plate (approximately 150 mm by 150 mm) was used as the substrate onto which a nanocomposite sheet was bonded using an epoxy adhesive. The PMMA plate unit was installed onto a three-point bending fixture in an Instron 5982 universal materials testing system, as shown in Fig. 1(a). Four electrodes were attached on the surface of the sheet, as shown in Fig. 1(b), such that the resistances in longitudinal (R_L between L_1 and L_2) and transverse (R_T between T_1 and T_2) directions were measured in situ as the flexural load was applied.

![Fig. 1. Schematic of piezoresistivity measurement test under bending](image)

3 Results and Discussions

Figure 2 compares the longitudinal and transverse resistance changes of 1 wt% MWCNT-PMMA composite sheets (bonded on PMMA plate surfaces) upon three-point bending. In three-point bending, as the plate undergoes flexural deformation, the top and bottom layers become subjected to compression and tension, respectively. In this study, as the nanocomposite sheet is bonded on the “bottom” surface of the PMMA plate (Fig. 1(a)), it is subjected to tension in the longitudinal direction. As the flexural load increases, the resistances increase in both longitudinal and transverse directions, indicating piezoresistive behavior. As can be observed in Fig. 2, the resistance increases in the longitudinal direction is greater than those in the transverse direction.

![Fig. 2. Longitudinal and transverse resistance changes of 1 wt% MWCNT-PMMA composites upon flexural loading](image)

Similar trends were observed for 1 wt% xGnP-PMMA composite sheets bonded on the bottom surfaces of the PMMA plates subjected to three-point bending (Fig. 3). As can be seen from Fig. 3, the longitudinal resistance curve for the xGnP-M-15 nanocomposite is steeper than that for the xGnP-M-5 nanocomposite, indicating xGnP-M-15, which is the larger of the two, provides higher sensitivity. This is attributed to the fact that at the same xGnP content, the conductive network formed by larger conductive fillers (in this case, xGnP-M-15) is more susceptible to, that is, it is more prone to be disrupted when subjected to, external loading.

![Fig. 3. Longitudinal resistance changes of 1 wt% xGnP-PMMA composites upon flexural loading](image)
The resistance ratio $R_\parallel/R_\perp$ was used to assess the degree of CNT alignment in extruded MWCNT-PMMA films, where $R_\parallel$ and $R_\perp$ are the resistances measured along the longitudinal and transverse directions, respectively, with respect to the CNT-aligned direction. A lower take-up speed resulted in a lower degree of alignment, which indicated that the alignment of the MWCNT can be varied by controlling the take-up speed (Fig. 4).

In both the directions parallel and perpendicular to the fiber axis of nanocomposite sheets, the resistance increase in the longitudinal direction was greater than those in the transverse direction, which was more pronounced in the perpendicularly oriented CNT composite (Fig. 5). Also, the change in resistance was about twice as high in the perpendicularly oriented CNT composite as compared to the parallel oriented CNT composite. This is attributed to the fact that in the perpendicular orientation, there is higher susceptibility of the CNT network to disruption, leading to substantially large increase in resistivity.

**Fig. 4.** Dependence of CNT alignment on take-up speed during film extrusion

(a) Longitudinal and transverse resistance changes in the sample in which CNTs are aligned in the transverse direction

(b) Longitudinal and transverse resistance changes in the sample in which CNTs are aligned in the longitudinal direction

**Fig. 5.** Longitudinal and transverse resistance changes of 1 wt% MWCNT-PMMA composites upon flexural loading

**4 Conclusions**

This paper presents an experimental study on the electrical and piezoresistive behavior of MWCNT- and xGnP-filled PMMA composite sheets and their application in large-area strain sensing of structures subjected to flexural loading. The effect of not only the type of carbon nanomaterial but also its orientation (for MWCNTs) was investigated. The study paved the way for large-area structural health
monitoring using smart carbon nanomaterial-filled polymer composite sheets, which can be bonded to or embedded into structures to be monitored. The SHM technologies presented have the following advantages:

1. The sheet-form nanocomposite sensors can cover large areas on the surfaces of the structures and allow macroscale strain measurement. In addition, based on the arrangement probe arrays, strains can be measured in any desired directions using a novel neural network algorithm, and three-dimensional strain mapping is possible.

2. The sensing performance can be tailored by controlling various material and process parameters, including carbon nanomaterial type, concentration and orientation, polymer matrix type, and degree of nanomaterial dispersion.

3. The nanocomposite sensors can be integrated in the composite structure without adversely affecting or altering the manufacturing processes.

4. The onboard signal acquisition and data analysis software implementing cumulative stress tracking, flaw detection, and fatigue life prediction algorithm will enable accurate and reliable structural health monitoring.

5. The inclusion of carbon nanomaterials will enhance the mechanical properties and durability of the composite structures while not increasing the overall weight significantly.

References