Electromechanical strain sensing using polycarbonate-impregnated carbon nanotube–graphene nanoplatelet hybrid composite sheets

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ABSTRACT

We report an experimental study on the electromechanical strain sensing ability of polycarbonate-impregnated hybrid sheets consisting of exfoliated graphite nanoplatelets, nanographene platelets, and multi-walled carbon nanotubes. The hybrid sheets were fabricated through surfactant-aided carbon nanomaterial dispersion followed by vacuum-induced filtration. The inherently porous sheets were impregnated with polycarbonate by infiltrating a polycarbonate–chloroform solution through the sheets. SEM analyses revealed that combining nanomaterials of various sizes and dimensions can serve as a means to control the porous network structure, which allows controlled polymer impregnation and tailored strain sensitivity. The wide-area strain sensing ability of the polymer-impregnated composite sheets was demonstrated by subjecting the composites with multiple electrodes to a flexural load and measuring the piezoresistivity *in situ*. The study demonstrated successful hybridization of 1D fiber-like and 2D platelet-like carbon nanomaterials into freestanding sheets with controlled nanostructure and properties, which can be used as preforms for easy-to-handle, high-carbon-content, multi-functional composite sheets.

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1. Introduction

Both carbon nanotubes (CNTs) and graphene consist of sp^2-bonded carbon atoms, which make them remarkably stable materials with great mechanical strength and fascinating electronic properties [1]. Hence, a wide range of smart materials have been developed for practical applications, addressing challenges in advanced aerospace, mechanical parts, bionics and medical technologies with these carbon allotropes [2,3]. Recently, in the area of structural health monitoring, strain sensing using polymer nanocomposites have received much attention due to the distinct advantages of polymers and nanofillers. In particular, a number of theoretical and experimental research has been carried out on strain sensing behavior of CNT-polymer [4–7] and graphene–polymer [8,9] composites. Many of the early studies on carbon nanocomposite based strain sensing involved Raman spectroscopy. For instance, Frogley et al. [10], reported the relationship between Raman spectrum shift and mechanical strain in CNT-epoxy composites. A similar Raman spectroscopic study was conducted on graphene monolayer composites by Huang et al. [11], who observed the same Raman spectrum shift under uniaxial tension. The 2D and G bands exhibit a redshift and the G band splits into 2 distinct (G^+,G^-) features because of the strain-induced breaking of the crystal symmetry. Although this method provides sufficient accuracy for monitoring molecular-level stretching and load transfer between polymer and CNTs (or graphene sheets) in nanocomposites, the area that can be monitored at a time is limited by the spot size of the irradiated laser, typically a few hundred square microns at most, which is not suitable for a field-deployable structural health monitoring system. Hence, monitoring the strain-induced resistivity change, known as piezoresistivity, is more appropriate for wide-area strain sensing.

To date, many studies had been reported on the reversible electromechanical characteristics of CNT buckypapers [12–14], CNT-polymer composites [15–18] and graphene–polymer composites [19,20] for strain sensing. Recently, Li and Chou [21] reported that CNT films can be used as strain sensors at the macroscale due to the dependence of the electrical properties of the SWCNT films on mechanical deformation at the nanoscale. Dharap et al. [13] reported a linear change in voltage when a CNT film was subjected to tensile strain, and they proposed that strains at multiple locations can be measured in the isotropic CNT film. Hu et al. [6] exclusively

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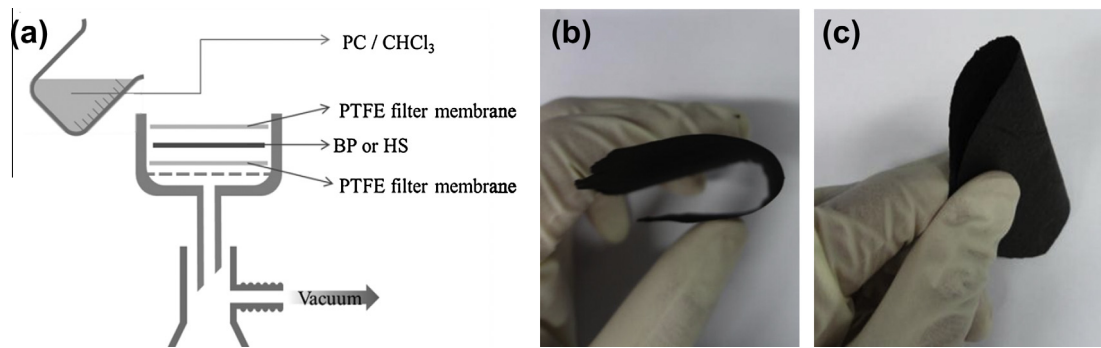
Fig. 1. (a) Schematic diagram of PC impregnation process and digital images of HS (CM-250/M-5) (b) before and (c) after polymer impregnation.

studied the strain sensitivity of the CNT-polymer composites based on the statistical resistive network using theoretical and experimental results. Eswaraiah et al. [19] demonstrated the real-time strain response of functionalized graphene-PVDF nanocomposites at the macroscale under tensile loads and the use of these nanocomposites as strain sensors.

Similar to CNT and graphene nanocomposites, exfoliated graphite nanoplatelet (xGnP) and graphene oxide (GO) can be dispersed in selected polymer matrices to yield composite materials with preferred properties [22,23]. xGnP- or GO-based polymer composites exhibit outstanding thermal, electrical and mechanical characteristics. The graphitic nanostructured particles were "hybridized" with CNTs and were mainly studied as functional materials for particular applications, such as supercapacitors [24,25], transparent electrodes [26,27], catalyst supports [28,29], field emission devices [30,31], chemical sensors [32] and photonic applications [33]. The knowledge of synergistic effect was obtained from the recent electrochemical study of "hybrid films," while their effects on electromechanical properties, more specifically piezoresistive behavior, are still illusive.

Here, we present a novel hybrid composite for efficient wide-area strain sensing, utilizing various combinations of size and length of xGnPs, nanographene platelets (NGPs) and CNTs that form freestanding sheets, which are subsequently impregnated with polycarbonate by vacuum filtration. The aim of the present work is to study the interactions between 2D platelet-like xGnPs and NGPs and 1D fiber-like CNT, and their effects on the electromechanical properties of the composite sheets. The feasibility of wide-area strain sensing was studied employing multi-probe resistivity monitoring under flexural loading.

2. Experimental

2.1. Materials

Polycarbonate (PC) with average molecular weight of 22,000 g/mol was obtained from Samsung Cheil Industries Inc. CVD-grown multi-walled carbon nanotubes (MWCNTs) with a purity rating of >95%, 5–10 nm inner diameter, 60–100 nm outer diameter, and two different lengths – 100 μm (CM-100) and 250 μm (CM-250) – were purchased from Hanwha Nanotech. Two types of xGnP with average lateral dimensions of 5 μm (M-5) and 15 μm (M-15) were purchased from XG Sciences. Nanographene platelets (NGPs) with a documented average lateral dimension of <10 μm, average thickness of <1 nm, oxygen content of <2.1%, and surface area of 400–800 m^2/g were purchased from Angstron Materials. The carbon nanomaterials were used without any chemical modification. Surfactants, sodium lauryl sulfate

Table 1
Filler compositions in PC-BP and PC-HS composite sheets.

Material type	Sample name	xGnP or NGP content in HS (wt.%)	Filler content in PC-BP or PC-HS (wt.%)
CM-250	PC-CM-250	0	59.6
CM-250 & M-5[a]	PC-CM-250/M-5 (8:2)	20	62.4
	PC-CM-250/M-5 (5:5)	50	64.9
	PC-CM-250/M-5 (3:7)	70	71.4
CM-250 & M-15[a]	PC-CM-250/M-15 (8:2)	20	65.2
	PC-CM-250/M-15 (5:5)	50	71.3
	PC-CM-250/M-15 (3:7)	70	75.3
CM-250 & NGP[b]	PC-CM-250/NGP (8:2)	20	69.3
	PC-CM-250/NGP (7:3)	30	76.28
	PC-CM-250/NGP (6:4)	40	81.3
	PC-CM-250/NGP (5:5)	50	81.3
CM-100	PC-CM-100	0	61.3
CM-100 & M-5[a]	PC-CM-100/M-5 (8:2)	20	63.4
	PC-CM-100/M-5 (5:5)	50	69.3
	PC-CM-100/M-5 (3:7)	70	75.3
CM-100 & M-15[a]	PC-CM-100/M-15 (8:2)	20	65.2
	PC-CM-100/M-15 (5:5)	50	71.3
	PC-CM-100/M-15 (3:7)	70	72.2
CM-100 & NGP[b]	PC-CM-100/NGP (8:2)	20	70.5
	PC-CM-100/NGP (7:3)	30	70.2
	PC-CM-100/NGP (6:4)	40	74.8
	PC-CM-100/NGP (5:5)	50	79.7

[a] 70 wt.% was the highest xGnP content achievable.
[b] 50 wt.% was the highest NGP content achievable.