



Electrically conductive graphene-coated polyurethane foam and its epoxy composites

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ABSTRACT

A facile method is presented to manufacture electrically conductive graphene-coated polyurethane foam (GPF) by dip-coating polyurethane (PU) foams in a graphene (GN) aqueous suspension containing cellulose nanowhisker (CNW) as dispersion assistant. The electrical conductivity of the GPF/epoxy composites reaches 2.46 S/m at a relatively low GN content (0.9 wt%), and the electrical percolation threshold is around 0.3 wt%. The structure observations reveal that the high electrical conductivity results from the 3D networks formed by GN nanosheets coated on the inner surface of PU foams. Moreover, CNW is a key factor to facilitate the formation of GN networks on the PU skeleton. The highest electrical conductivity is obtained at the CNW / GN weight ratio of 1:20.

1. Introduction

Conductive polymer composites have attracted considerable attention and have wide applications in different industrial aspects due to their low density, corrosion resistance and easy processing [1]. One of the best fillers to fabricate conductive polymer composites is GN, owing to its unique and excellent properties, such as high electrical conductivity, high thermal conductivity, extraordinary elasticity and stiffness [2–4]. It has been reported that adding GN to polymers is able to boost the electrical property of the resulting nanocomposites [5]. However, because of the poor dispersion of GN in a polymeric matrix, the application of GN-based polymer composites is limited [6].

The introduction of three-dimensional (3D) GN networks has been considered as one of the most effective methods to apply GN practically, due to the high electrical and thermal conductivity of their composites [7,8]. 3D GN networks, including GN foams, GN sponges and GN aerogels, which are constructed with individual GN sheets by bonding them together, avoid re-stacking of individual GN sheets [9]. Jia et al. [10] achieved GN foam/epoxy composites with both high electrical conductivity and excellent mechanical properties via template-directed chemical vapor deposition. Zhang et al. [11] reported that 3D GN assemblies prepared from an in situ reduction-assembly method exhibited low density, highly porous structures, and electrically conducting properties. Although many efforts have been made to fabricate 3D GN networks via reducing graphene oxide (GO) or chemical vapor deposition methods, we notice that a general method using commercial

GN as raw materials to fabricate polymer composites with high electrical conductivity is still a challenge.

It has been reported that CNW is able to improve the dispersion of pristine GN in aqueous solution [12–14], which makes it possible to directly use commercial GN in conductive polymer composites. CNW can be derived from cellulose, which is biocompatible, biodegradable and renewable natural polymer [15], via various methods [12,13]. Additionally, CNW has high surface areas and high aspect ratios, and excellent mechanical properties.

In this paper, we present a facile, inexpensive and eco-friendly method to fabricate GPFs by using commercially available PU foams, GN, and CNW as the raw materials. CNW used in this study was prepared via the method reported in our previous work [13,14]. We started this work at the CNW/GN ratio of 1:20 as it has been demonstrated that the CNW/GN ratio has a notable influence on the electrical conductivity of the CNW/GN composites and that the conductivity of the composites at the CNW/GN weight ratio of 1:20 is higher than that of the composites at the ratio of 1:1 [12]. Highly conductive GPF/epoxy composites were fabricated by using GPFs as scaffold infiltrated with epoxy resin. Integral 3D GN networks can be observed in the composites. Besides, GPF/epoxy composites with different CNW/GN ratio were fabricated to investigate the effect of CNW. These highly conductive composites can be fabricated for practical applications, such as electronic devices, electromagnetic shielding and electronic packaging.

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2. Materials and methods

2.1. Materials

Commercial GN was purchased from Sichuan Jinlu Group (Deyang, Sichuan). Epoxy E-44, polyamide 650, 1,4-butanediol diglycidyl ether 691 are purchased from Shenyang Zhentraid Anti-corrosion Materials Co., Ltd. and commercial PU foams were from Qingdao Yuquan Haimian Co., Ltd.

2.2. Preparation of CNW and GPFs

CNW was obtained through the method reported previously [13,14]. The CNW suspension (2 mg/mL) was obtained by adding 0.1 g CNW to distilled water (50 mL) followed by ultrasonication (KH-1600TDE, Kunshan, China) in an ultrasonic bath for 2 h. Then, 2 g GN was added to the CNW suspension and uniform CNW/GN suspension was obtained after the mixture was ultrasonicated in an ultrasonic bath for 3 h. Extra water was added to control the GN concentration to be 2 mg/mL. PU foams were dip-coated into the CNW/GN suspension and then dried for 5 h at 70 °C to produce GPFs. The weight ratio of GN to PU foams was determined by weighing the initial and graphene-coated foams immediately after being taken out from the oven to avoid moisture absorption (all the initial weight of PU foams range from 0.184 g to 0.185 g). This dip-coating process was repeated to increase the amount of GN coated on the PU foams, thus controlling the weight ratio of GN to PU foams.

2.3. Preparation of GPF/epoxy composites

Epoxy E-44, polyamide 650 and 1, 4-butanediol diglycidyl ether 691, were mixed uniformly via mechanical stirring, where polyamide 650 was used as a curing agent and 1, 4-butanediol diglycidyl ether 691 was used to reduce the viscosity of the epoxy. The weight ratio of these three materials was kept at 10:5:8, as the instruction suggested. The GPFs with different weight ratio of GN to PU foams were completely immersed into the epoxy mixture and then placed in a vacuum chamber for approximately 15 min to remove air bubbles and cured at 70 °C for 18 h. Excessive epoxy adhered on the foam surface was removed after

curing. The process of preparing GPF/epoxy composites is illustrated in Fig. 1a. The GN content, defined as weight ratio $W_{GN}/W_{composite} \times 100\%$, was determined by the weight of GN and that of GPF/epoxy composites.

2.4. Preparation of reference composites

PU foams were dipped directly into GN aqueous suspension to fabricate graphene-coated foams without CNW (GF-WC). Then, reference epoxy composites without CNW were prepared by using GF-WC as the scaffold.

2.5. Characterization

The specimens of GPFs and GPF/epoxy composites were fractured after being dipped into liquid nitrogen. The fracture surface of these specimens were coated with a thin layer of gold and investigated by using a JSM-6301F Scanning Electron Microscope (SEM) with an acceleration voltage of 20 kV to examine the morphology. Electrical conductivity of the GPFs and GPF/epoxy composites was measured at room temperature using a two-probe measurement with two copper electrodes (Keithley-2000 digital multimeter (100 $\mu\Omega$, 100 M Ω)). The specimens used for the electrical conductivity measurement (GPFs: 30 \times 30 \times 4.5 mm. GPF/Epoxy composites: 46 mm in diameter and 4.0 mm in length) were silver-pasted to minimize the contact resistance between the composites and the electrodes. The electrical conductivity σ can be calculated from the Eq. (1):

$$\sigma = l/R \times S \quad (1)$$

where l , R , and S are thickness, resistance and area, respectively.

3. Results and discussion

Fig. 1b presents the typical morphology of CNW whose length is several hundred nanometers and the aspect ratio is about 10. SEM image of GN is shown in Fig. 1c. GN has a flaky sheet structure in shape with lateral size of around 10 μm . Macroscopic GPFs were prepared from CNW/GN suspension, as shown schematically in Fig. 1a. It has been demonstrated that CNW is an efficient GN stabilizer that can be

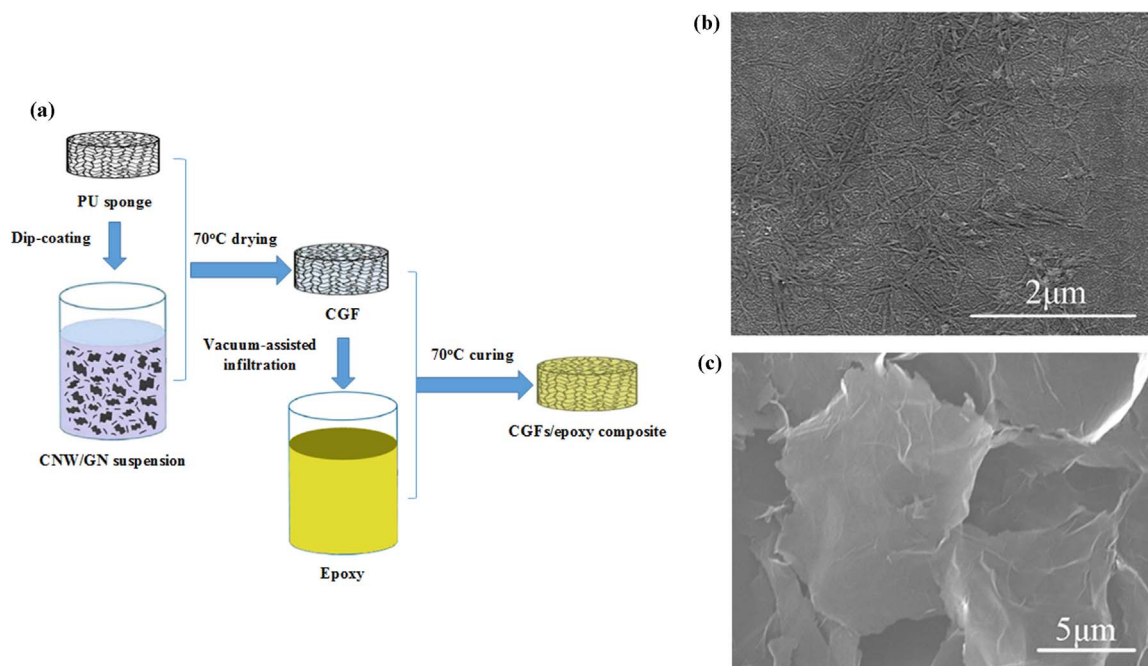


Fig. 1. (a) Schematics of the fabrication processes of GPF/epoxy composites and SEM images of (b) CNW and (c) GN.

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