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Comparative study on monitoring structural damage in fiber-reinforced polymers using glass fibers with carbon nanotubes and graphene coating

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ABSTRACT

This paper reported the preparation of glass fibers (GFs) with carbon nanotubes (CNTs) and graphene coating by using an electrophoretic deposition (EPD) process. The developed GFs were embedded into epoxy to illustrate the role of different coatings on monitoring the damage in fiber reinforced polymers (FRPs). Piezoresistive effects of GFs with different coatings and corresponding composites were studied and compared. The results showed that GFs with both coatings exhibited a negative temperature coefficient behavior, and graphene coated sample was more sensitive to temperature and strain than that of CNTs. Huge dissimilarities on piezoresistivity of FRPs were also observed when the samples were subjected to the mechanical loading, which originated from the different interphases generated between the fibers and polymer matrix. GFs with graphene coating provided more details on the damage accumulation in FRPs and could be more practical for the early warning of structural damage in FRPs.

1. Introduction

Fiber reinforced polymers (FRPs) have become one of the most important materials for engineering applications due to their high specific stiffness and strength, outstanding fatigue performance, good chemical and thermal resistance as well as low cost [1–3]. The concept of stress-transfer between the fiber and polymer matrix in FRPs is central to predict the mechanical behavior of composites [4,5]. Principally, the stress-transfer depends on the structure and properties of interphase formed between the fiber and matrix. A high interfacial shear stress will dominate the effective transfer of the applied load to the fiber, resulting in improved mechanical properties of composites. Therefore, the understanding on the interfacial failure in a FRP structure is important not only for the design of materials, but also for the life assessment of composites in service conditions. Various materials, such as optical fibers [6], shape memory alloys [7], carbon fibers [8], have been employed to study the interfacial failure in FRPs. However, these conventional materials cause large stress perturbations in FRP structure due to their large diameters and weak interfacial adhesion between the fibers and polymer matrix.

The emergence of nanotechnology as a major field of research, especially based on carbon-based nanoparticles (NPs), such as carbon nanotubes (CNTs) and graphene, has impacted almost every scientific discipline, and their assemblies and composites were designed towards various applications [9–11]. The use of CNT or graphene as a sensory material for damage monitoring in FRPs was reported as well [12–16]. The bulk of previous and ongoing studies is mainly based on polymer nanocomposites reinforced by CNTs or graphene, where the structural deformation is measured by monitoring the deformation of the matrix. Coupling the electrical properties and mechanical deformation in NPs within a matrix material makes the composites with sensory capability. A major drawback based on this approach is that one cannot predict the damage in the reinforcements, which actually sustain the majority of the load applied to the structure.

As an alternative, CNT or graphene as fiber coating/sizing was developed in recent years, and the NP-coated fibers exhibited comparable mechanical properties to the structural reinforcements (fibers without coating/sizing), making them an integral part of composite structures without any negative effects on their performance. Such implementation offers a new route for the advanced

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warning of damage in composites by integrating the interphase modification concept into the FRPs. For example, Gao and co-workers [17,18] used a single CNT-glass fiber to monitor the initiation and growth of micro-cracks in composites, providing an early warning system to detect fracture in materials. Fu at al [19] used graphene as a modifier on glass fibers (GFs) to control the interfacial crystallization of polymer, thus improving the interfacial adhesion between the fiber and matrix. When polymer-based nanocomposite containing CNTs or graphene were applied as coatings on GFs, the flaws on fiber surface produced during the fiber production were repaired, and the fibers showed much enhanced barrier performance to the environmental attacks [20]. The coating of carbon-based NPs brings additional benefit in making glass fibers Raman-active, thus provides a way to measure the local strains in glass fiber reinforced polymers (GFRPs) by using optical method. Young et al. [21] introduced strain-sensitive coatings made of CNTs and epoxy to measure stress transfer from matrix to the fiber by studying the Raman shift of CNT under mechanical strain.

While CNT or graphene has been utilized as a functional coating for GFs, few reports were found in the literature in controlling and optimizing the structure and morphology of coating layer, less effort was put to compare the role of coating on the properties of fibers and structural damage process of corresponding FRPs under the mechanical loading. It is known that CNTs have tube-like morphology, whereas graphene exhibits a plate-like structure, such significant difference on dimension may result in the different function of coating in governing the behavior of load-transfer from polymer matrix to reinforcement via an interphase. Therefore, comparative studies to address these issues are highly desirable.

Motivated by the above efforts, this paper reported the preparation of functional GFs with CNTs and graphene coating. The prepared GFs were embedded into epoxy to fabricate model GFRPs. The piezoresistivity of single GFs with different coatings and corresponding GFRPs were studied and compared, and the feasibility of employing GFs to detect structural damage in composites was demonstrated.

2. Experimental

2.1. Materials

E-glass fibers with an average diameter of 12 μm were used in this study. The nanotubes were multi-walled CNTs (Nanotech Port, China), and the diameter and length ranged between 20 and 40 nm and 5–15 μm, respectively, according to the supplier's specification. Graphene was obtained by reducing graphene oxide (GO) under the thermal treatment [22]. For the preparation of GO, briefly, 5 g of graphite flakes (Asbury) were mixed with H_2SO_4 (30 mL, 98%) and fuming nitric acid (50 mL, 90%). The mixture was stirred for 24 h and separated from the solution to obtain graphite intercalation compound (GIC). The GIC was treated at 1050 °C for 15 s to obtain expanded graphite (EG). Then 1 g EG and 200 mL H_2SO_4 were combined and stirred in a three-neck flask, followed by addition of 10 g $KMnO_4$. The mixture was kept at 60 °C for 24 h, then deionized (DI) water (200 mL) and H_2O_2 (50 mL, 30%) were slowly poured into the system. The mixture was washed by water and following gentle ultrasonication to obtain GO suspension (2 mg/mL).

2.2. Preparation of functional GFs and GFRPs

The coating of CNT and GO on GFs was accomplished by an electrophoretic deposition (EPD) method (See Supporting Information Fig. S1). CNT dispersion (0.05 wt%) was prepared by sonicating CNTs into 100 mL of sodium dodecyl sulfate solution (1 mg/mL) using a probe sonicator (Digital Sonifier, Branson) for 30 min. GO dispersion (0.05 wt%) was obtained by diluting the prepared GO suspension. Two copper plates (length 12 cm × width 6 cm × thickness 0.3 cm) were employed as electrodes in EPD. Single GFs were fixed on a plastic framework mounted on the surface of copper electrode. EPD experiment was performed under a voltage of 20 V with deposition time of 3.5 min, and the distance between two electrodes was kept at 1 cm. The above process was repeated on the reverse side of GF to get a homogenous coating on the fiber surface. This was followed by processing the fibers at 400 °C in N_2 for 2 h to eliminate the impurities and reduce GO to graphene in the coating layer [22]. The obtained samples were labeled as CNT-GF, GO-GF and graphene-GF.

For the preparation of model GFRPs, a 60 mm coated fiber was placed in the center of a dog-bone shaped Teflon mold and connected with copper wires on both fiber ends by conductive carbon paste. Epoxy monomer (LR135) and curing agent (LH136, both from Momentive) were mixed with weight ratio of 100:35 and degassed at 50 °C for 10 min prior to pouring into the mold. The samples were cured at room temperature for 24 h, followed by a post curing at 65 °C for 16 h. Composites with single fiber were labeled as CNT-GFRPs or graphene-GFRPs.

2.3. Characterization

A scanning electron microscopy (SEM, Zeiss Supra55VP) was employed to characterize the morphology of GFs with different coatings. Energy-dispersive X-ray spectroscopy (EDX, Bruker XFlash-SDD-5010) in mapping mode was used to evaluate the distribution of element in the composites. The electrical properties of single fiber and model GFRPs were determined using a two-probe method on a digital multi-meter (34410A, Agilent). For single fiber test, at least three specimens were measured and the average value of resistance was reported with standard deviations. The tensile test for the samples was performed on a universal testing machine (UTM, C43-104, MTS). The piezoresistance of fibers was evaluated by combining the electrical resistance measurement with tensile test. For single fiber test, the specimen was placed straightly on a perforated cardboard by gluing fiber ends on conductive carbon paste, which acted as an adhesive to connect the fiber with copper wires linked with the multi-meter. The specimens had a gauge length of 4 cm and a crosshead velocity of 0.05 cm/min was chosen for loading on the UTM equipped with a load cell of 10 N. At least 20 samples were tested and the results on tensile test were analyzed using a two-parameter Weibull model [20]. For model GFRP, the in-situ resistance of sample under the load was obtained by connecting copper wires at the end of composites to the multi-meter along with a tensile rate of 0.5 cm/min on the UTM with a load cell of 1000 N.

3. Results and discussions

3.1. Morphology and electrical properties of GF with coatings

Fig. 1 shows the morphology of GF with different coatings. The EPD process produced a fairly uniform coating layer consisting of either graphene or CNTs on fiber surface. Graphene exhibits a full coverage to the GF (Fig. 1A), and CNTs form networks along the fiber direction (Fig. 1C). These morphological differences originated from the structures of coatings: graphene has a plate-like structure and can be easily assembled layer by layer on GF surface, whereas CNTs are one dimension materials with high aspect ratio, the connectivity of individual CNTs leads to the continuous networks on fiber surface. A close examination on the cross-section of coated fibers shows that the thicknesses of graphene and CNT coating are 250