



Research paper

Study on interfacial and mechanical improvement of carbon fiber/epoxy composites by depositing multi-walled carbon nanotubes on fibers

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ARTICLE INFO

Article history:

Received 27 February 2018

In final form 8 May 2018

Available online 8 May 2018

Keywords:

Epoxy resin

Carbon fiber

Carbon nanotubes

Composite

ABSTRACT

To improve the interfacial properties between carbon fiber (CF) and epoxy resin (EP), T300 carbon fibers were coated with multi-walled carbon nanotubes (MWCNTs) using aqueous suspension deposition method. The carbon fiber/epoxy laminated composites were prepared by molding process. The wettability and interfacial properties between MWCNTs deposited carbon fibers (MWCNTs-T300) and EP were studied. The mechanical properties of carbon fiber/epoxy laminated composites were tested, and the mechanism of the interface strengthening was discussed. The results show that the surface energy of T300 carbon fiber is obviously increased after MWCNT deposition. The contact angle between MWCNTs-T300 and EP is reduced, and the interfacial energy and adhesion work are greatly improved. The MWCNTs-T300/EP laminated composites have excellent mechanical properties, the flexural strength is 822 MPa, the tensile strength is 841 MPa, and the interlaminar shear strength (ILSS) is 25.68 MPa, which are increased by 15.1%, 17.6% and 12.6% compared with those of the original carbon fiber/EP laminated composites (original T300/EP) respectively. The MWCNTs-T300/EP composites have good interface bonding performance, low porosity and uniform fiber distribution. Interfacial friction and resin toughening are the main mechanisms for the interface enhancement of MWCNTs-T300/EP composites.

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1. Introduction

Carbon fiber (CF) reinforced epoxy composites have attracted wide attention in academia and engineering fields for their good mechanical and economic performance [1]. However, under the harsh working conditions of high load and strong impact, the carbon fiber reinforced epoxy composites still have many disadvantages, such as low strength, low toughness and high environmental sensitivity. The mechanical properties of carbon fiber reinforced epoxy composites are not only affected by the inherent characteristics of CF and epoxy resin (EP), but also affected by the interfacial interaction of CF and EP [2]. The interface between CF and EP is the key element in the performance of the composite [3].

In recent years, to improve the mechanical properties of composites, carbon nanotubes (CNTs) have been introduced into the traditional continuous fiber reinforced polymer composites to build hierarchical reinforcing structure, which is currently an important research topic [4]. Because of their good mechanical

properties, CNTs are widely used as additives to improve the mechanical properties of carbon fiber reinforced plastics (CFRP) [5,6]. Researches have shown significant improvements in glass fiber and carbon fiber composites by using multi-scale reinforcements, especially the fiber-matrix interfacial properties such as interfacial shear strength (IFSS) [2,7–10], interlaminar fracture toughness [11,12] and glass transition temperature (T_g) [13]. Sager et al. reported that the interfacial shear strength of the radially aligned MWCNTs deposited carbon fiber specimens was increased by 11% than that of the untreated, non-sizing carbon fibers [8]. CNTs can effectively enhance the interaction of the CF-EP interface by mechanical interlocking of the EP matrix and CF surface [14]. In general, the mechanical properties of CFRP were improved after the addition of CNTs, due to the strengthening of the fiber-matrix interface [15]. Iwahori et al. studied the mechanical properties of CNT modified CF/EP laminated composites, and the flexural strength of CF/EP composite increased by 2.7% due to the addition of CNTs [16]. The reason for the low increase in flexural strength is that the CNTs were directly dispersed in the epoxy matrix rather than connected to the carbon fiber surface.

Dispersing CNT into the composite matrix and directly connecting CNT to the fiber surface are two strategies for forming

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CNT-based hierarchical composite. To further improve the interfacial properties between fiber and resin, many researchers have focused on attaching CNTs directly to fiber bundles and fabrics. So far, there have been four methods for directly attaching CNTs to fiber surface: (1) chemical reaction between functionalized CNTs and fiber [17,18], (2) electrophoretic deposition of CNTs on the fiber surface, (3) the growth of CNTs on the fiber by chemical vapor deposition (CVD), (4) depositing CNTs comprising coating the fibers [19] with a CNT-containing sizing (a thin polymer coating) and spraying the CNT-containing solution onto the fibrous fabric. Among the above methods, the chemical reaction method and electrophoretic deposition method all require complex chemical treatment and complicate equipment. The CVD method not only requires expensive equipment and complex processing technology, but also the high processing temperature, which will lead to surface damage of carbon fiber, and affecting the mechanical properties of carbon fiber. For example, the tensile test performed by Sager et al. showed that CVD treatment significantly reduced the tensile properties of carbon fibers, and the tensile strength and tensile modulus were reduced by 30% and 13%, respectively, in the case of random orientation of MWCNTs [8]. For the depositing CNTs method, it is the most simple and effective, and will not cause damage to the fibers (even repairing surface defects and improving fiber tensile properties) [19–21]. In addition, the process does not require the removal of industrial sizing and can be used for various types of carbon fiber reinforcements, such as fiber bundles, fabrics and preforms. To improve the interface performance of carbon fiber composite, some researches have adopted the method of depositing CNTs. For example, Kwon and colleagues achieved an increase in interlaminar fracture toughness of CF/vinyl ester laminate composites by depositing CNTs, with a maximum increase of about 30%. However, the current researches on depositing CNTs method is mainly focused on improving one single mechanical properties of composites, which cannot achieve the improvement of comprehensive mechanical properties, and the study on its strengthening mechanism is still at the level of qualitative analysis. There is a lack of quantitative analysis of the interfacial properties between carbon fiber and epoxy resin, and the influence of interfacial properties on the mechanical properties of carbon fiber/epoxy composites.

To improve the interfacial properties of carbon fiber and epoxy resin, and further improve the mechanical properties of carbon fiber/epoxy composite, carbon fibers (T300) were coated with MWCNT using aqueous suspension deposition method. Carbon fiber/epoxy laminated composites were prepared by molding process. The surface morphology of MWCNT deposited carbon fiber (MWCNTs-T300) and the wettability and interfacial properties between MWCNTs-T300 and EP were studied. The mechanical properties of carbon fiber/epoxy laminated composites were tested. The tensile and bending fracture morphology were analyzed. The mechanisms of interface strengthening are discussed, which provides the theoretical basis and technical support for the preparation of carbon fiber/epoxy composites with excellent performance and their application in engineering field.

2. Experimental

2.1. Materials

T300 carbon fiber monodirectional fabric is provided by Nanjing Fiberglass Research & Design Institute Co., Ltd. MWCNT: S-MWNT-1020, length 0.5–2 μm , diameter 10–20 nm, and purity greater than 97%, produced by Shenzhen Nanotech Port Co., Ltd. The E-44 epoxy resin with an epoxy value of 0.44 mol/100 g is provided by Taizhou Huili Electronic Material Co., Ltd. Curing agent: D-230

polyether amine (PEA), provided by Suzhou Chanco Industrial Co., Ltd.

2.2. Preparation of samples

MWCNTs were dispersed in deionized water by ultrasonic (ultrasonic generator, sonic power 125 W, ultrasonic frequency 80 kHz) for 3 h prior to the preparation of MWCNT deposited CF to obtain a stable suspension at a concentration of 0.1 wt%. Then, the carbon fiber fabric was immersed in the suspension for 20 min, followed by drying at 70 °C for 2 h and drying at 120 °C for 2 h.

The original CF laminated composite samples and MWCNT deposited CF laminated composite samples were prepared by compression molding using E-44 resin as the matrix and T300 carbon fiber as the reinforcing material. The size of the tensile samples and bent samples are 230 × 25 × 2 mm and 110 × 10 × 4 mm, respectively. The thickness and fiber content of the two kinds of samples are the same. The fiber volume fraction of the tensile samples is about 50%, and the fraction of the bent samples is about 60%. The main steps of the preparation are: tailoring the carbon fiber fabric, cleaning the mold, coating the mold with release agent and laying a layer of mold release paper. Then, the resin impregnated carbon fiber fabrics are stacked in the mold, close mold and pressurize, curing at 70 °C for 24 h. The samples are obtained after mold unloading.

2.3. Characterization

After the MWCNT deposition, the CF fabric was torn to expose the inside of the fabric, and then the carbon fiber bundle was examined by the Hitachi S4800 scanning electron microscope (SEM) to evaluate the adhesion state of MWCNT on the carbon fiber surface. The tensile strength and modulus of the composites were measured by the electronic universal testing machine (CMT 5105) using rectangular specimens made according to the American Society for Testing and Materials (ASTM) standard D3039. Three-point bending test was carried out on the bend specimens by CMT 5105 electronic universal testing machine. The test standard is ASTM D790-2015 and the bending speed is 2 mm/min. In addition, interlaminar shear strength (ILSS) was tested according to ASTM standard D2344.

The test specimens were placed in a plastic bag and pour into distilled water, seal and hold at 50 °C for 24 h. The test specimens were dried at 50 °C and then test the mechanical properties of the composite (test standard is the same as room temperature mechanical properties). At least 5 specimens were tested for each set of conditions.

The heat resistance was measured by DSC (Mettler-Toledo, DSC823e) (N_2 atmosphere, the test temperature is from room temperature to 250 °C, scanning rate of 10 °C/min, mass of 20 mg). The micromorphology of the sample fracture surface was detected by the Hitachi S4800 scanning electron microscope.

3. Results and discussion

3.1. Surface characteristics of MWCNTs deposited carbon fiber

The deposition state of MWCNTs on carbon fiber surface after deposition process was observed by SEM. Fig. 1a is the surface morphology of the original carbon fiber, indicating that the surface of the original carbon fiber is smooth. The typical SEM images of carbon fibers after MWCNT deposition are shown in Fig. 1b–d. In Fig. 1b, due to the low concentration of MWCNTs suspension (0.05 wt%), the MWCNTs deposited to the carbon fiber surface is

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