



Synergistic effect of carbon nanotubes and n-butyl glycidyl ether on matrix modification for improvement of tensile performance of glass fiber/epoxy composites



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ABSTRACT

In this work, an epoxy resin was modified by stiff multi-walled carbon nanotubes (MWCNTs) and soft n-butyl glycidyl ether (BGE) and the modified epoxy resins were employed as the matrices for improvement of the tensile performance of glass fiber reinforced composites. The results indicated that the simultaneous addition of MWCNTs and BGE led to the greatest enhancements in the composite strength and modulus, demonstrating that their simultaneous introduction yielded a synergistic effect on the matrix modification for the improvement of the tensile performance of the GF/epoxy composites. The scanning electron microscope observations showed that the modified epoxy resins were substantially coated on the surfaces of GFs while the un-modified matrix was lightly attached on the GFs. The enhancements in the composite tensile performance were attributed to two aspects of enhanced GF strength/modulus and improved interfacial adhesion for the modified matrix compared to the un-modified case.

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1. Introduction

Glass fiber reinforced polymer composites are very attractive materials for many industrial applications because of their excellent mechanical performance/cost ratio [1–4]. The mechanical properties such as strength and modulus of the glass fiber reinforced composites are mainly determined by the mechanical properties of the glass fibers (GFs). Unfortunately, the mechanical properties of the GFs are significantly influenced by the intrinsic defects on the surfaces of GFs that are readily created during the manufacturing and handling [5–11]. Traditionally, a sizing agent is often used to modify the GF surface properties to promote the GF/polymer interfacial adhesion and protect the GFs to some degree [5,10,11]. Recently, Maeder et al. [6–9] had applied carbon nanotube (CNT)/epoxy nanocomposites as coatings on the surfaces of the GFs to heal the defected GF surfaces. Indeed, a significant improvement in the GF mechanical properties was noted because of the healing effect of the nanocomposites [6–9]. The improvement was attributed to the CNTs in the nanocomposite coating acting as “bridges” at the defect tips on the fiber surfaces. In this

methodology, one extra processing step of coating on GFs is needed before GFs are incorporated into polymer matrices.

Recently, modified polymers by nanofillers were employed as matrices to fabricate glass fiber reinforced composites [12–15]. The effects of nanoclay content on the mechanical properties of chopped strand glass fiber reinforced epoxy composites were investigated [12]. At the 0.5 wt% nanoclay loading, the improvements in the interlaminar shear strength, flexural strength and flexural stiffness were about 5%, 8% and 12%, respectively. The epoxy matrix was modified with multi-walled carbon nanotubes to improve the composite mechanical properties [13]. The compressive and interlaminar shear strengths were increased by 39% and 15%, respectively for the woven glass fiber/epoxy composites. Carbon nanofibers (CNFs) were dispersed in an epoxy resin and the resultant nanocomposite was used as the matrix for carbon-fiber reinforced epoxy composites [14]. The addition of the CNFs at about 0.7 wt% significantly enhanced the strength and modulus of both quasi-isotropic and unidirectional composites. This indicated that the matrix modification by direct introduction of nanofillers could lead to the enhancements in composite mechanical properties. In our recent work [15], the modification of an epoxy resin by both multiwalled carbon nanotubes (MWCNTs) at 0.5 wt% and n-butyl glycidyl ether (BGE) at 10 wt% led to a significant enhancement in the interlaminar shear strength of woven GF/epoxy composites. CNTs are stiff modifier which may act as bridges

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between epoxy resin and GFs [9] while BGE is soft modifier which may reduce internal residual stresses created during processing in epoxy resin and hence enhance the mechanical properties of epoxy resins [16]. It may thus be inferred that simultaneous introduction of CNTs and BGE into an epoxy resin may yield a synergistic effect on matrix modification for improvements in composite mechanical properties. However, no report has been presented yet on the possible synergistic effect of the CNTs and BGE on matrix modification for enhancements in tensile performance of GF/epoxy composites.

In the present work, matrix modification was conducted for diglycidyl ether of bisphenol-F epoxy resin by both stiff MWCNTs and soft BGE. Then, unidirectional continuous glass fiber reinforced epoxy composites were prepared based on the modified epoxy matrices for investigating the synergistic effect of MWCNTs and BGE on matrix modification for enhancing the composite mechanical properties. The fracture surfaces of the samples after tensile testing were examined by scanning electron microscopy. It was observed that the mirror size on the fracture surface of GFs was related to the composite strength and hence to the fiber strength. And the relationship between the fiber strength and the mirror size was established.

2. Experimental section

2.1. Materials

Plain un-sized woven E-glass fiber (GF) clothes with a diameter of about 11 μm for single fibers were purchased from Feihsangtongda Co., Ltd., China. The epoxy resin used in this work was diglycidyl ether of bisphenol-F (DGEBF, D.E.R.354, Dow Chemical Co., USA) with the epoxide weight equivalence in the range of 167–174. The curing agent was diethyl toluene diamine (DET D, Kunshan Chemical Material Co., Ltd., China), which is a mixture of the two DETD isomers (74–80 wt% 2,4-isomer and 18–24 wt% 2,6-isomer). n-Butyl glycidyl ether (BGE, Changshu Jiafa Chemistry Co., Ltd., China) was used as a soft modifier for epoxy resin. The chemical structures of DGEBF, DETD and BGE are shown in Fig. 1. Multi-walled carbon nanotubes (MWCNTs) made by CVD (purity >95 wt%, Chengdu Organic Chemicals Co., Ltd., China) were employed as a stiff modifier for epoxy resins [17,18]. Typical transmission electron microscope (TEM, JEOL JEM-2100F) images of the

CNTs are shown in Fig. 2. The diameters of the CNTs were in the range of 30–50 nm and the length was between 10 and 20 μm [17,18].

2.2. Preparation of GF/epoxy composites

In this work, GF/epoxy composites were synthesized from fiber bundles extracted from plain woven fabrics. The yarn fineness of the GF bundles is 220 tex. One pure epoxy and three modified epoxy matrices were prepared in terms of the formulations listed in Table 1. Then the matrices were used to impregnate the GF bundles. The matrices were heated to 50 $^{\circ}\text{C}$ to lower its viscosity so that the epoxy resins could easily penetrate into the GF bundles. The GF bundles were impregnated in the matrices for about 10 min and the GFs could be coated by the matrices thoroughly. Finally, the composites were cured at 80 $^{\circ}\text{C}$ for 8 h, then 130 $^{\circ}\text{C}$ for 10 h. The as-prepared composites were shown in Fig. 3. The GFs are well immersed by the unmodified and modified epoxy resins. The diameter of the small composites was about 0.45 mm. The fiber volume fraction (V_f) was determined by weight method according to the following equation [19]:

$$V_f = (d - d_1)/(d_2 - d_1) \quad (1)$$

where d_1 , d_2 and d were the density of the matrix, fiber and composites, respectively. The density of the matrix and fiber was 1.20 g cm^{-3} and 2.54 g cm^{-3} , respectively. The measured composite density was about 1.95 g cm^{-3} . According to Eq. (1), the fiber volume fraction in the composites was estimated to be about $56 \pm 3 \text{ vol.}\%$.

2.3. Characterization

An Instron 5882 universal machine with a 500 N load cell was used to perform the tensile testing. The crosshead speed was 2 mm/min under the 35% relative humidity at the temperature of 20 $^{\circ}\text{C}$. The specimens were placed between the rubber faced grips. The gauge length was 80 mm. Measurement of strain during tensile testing was specially carried out using non-contact video extensometers that were suitable for precisely measuring displacements of small specimens. Strain was measured by tracking contrasting gauge marks placed on the specimens.

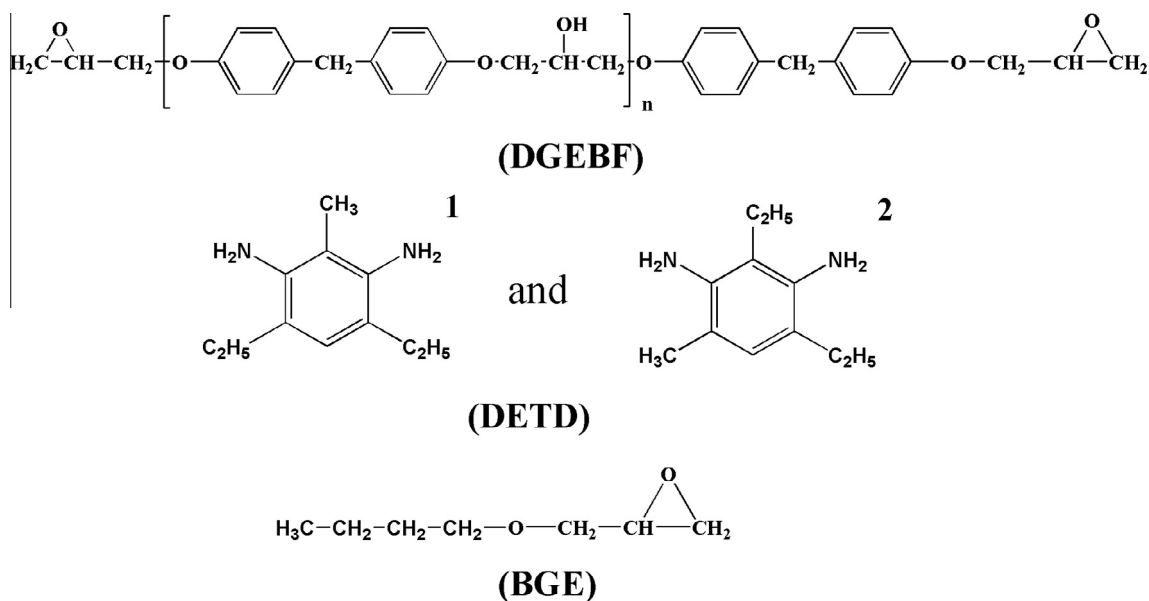


Fig. 1. The chemical structures of DGEBF, DETD and BGE.

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