



Effect of different carbon nano-fillers on rheological properties and lap shear strength of epoxy adhesive joints



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ABSTRACT

In this work, the rheological properties, thermal stability and the lap shear strength of epoxy adhesive joints reinforced with different carbon nano-fillers such as multi-walled carbon nanotubes (CNT), graphene nanoplatelets (GNP) and single-walled carbon nanohorns (CNH) have been studied. The nano-fillers were dispersed homogeneously using Brabender® Plasti-Corder®. The epoxy pre-polymer with and without the nano-fillers exhibited shear thinning behavior. The nano-filler epoxy mixtures exhibited a viscoplastic behavior which was analyzed using Casson's model. Thermo-gravimetric analysis indicated an increase in the thermal stability of the epoxy with the addition of carbon nano-fillers. Carbon nano-fillers resulted in increased lap shear strength having high Weibull modulus. The joint strength increased by 53%, 49% and 46% with the addition of 1 wt.% CNT, 0.5 wt.% GNP and 0.5 wt.% CNH, respectively. The strength of the joints having high filler content (>1 wt.%) was limited by mixed mode type of failure.

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1. Introduction

Adhesive bonding is an easy and economically alternative method applicable for joining a variety of materials. Adhesive bonding offers unique advantages such as uniform stress distribution throughout the joint and good strength to weight ratio [1,2]. Epoxy-based adhesives find vital applications in automotive, aeronautics, electronics and packaging industries due to their excellent mechanical properties [1,3]. They have outstanding adhesion to various substrates such as steel, aluminum and carbon fiber reinforced plastics [4,5]. However, the strength of the epoxy adhesive joints is low compared to bolted, riveted and welded joints and is limited by strength of the epoxy. Efforts have been made to improve the strength of epoxy joints by adding nano-fillers such as carbon nanotubes (CNT) [2,6–10], nano-SiO₂ [11,12], nano-Al₂O₃ [13–15], carbon black [16] and nano-CaCO₃ [13] with varying degrees of success. Since 1990s, carbon based nanomaterials have attracted lot of attention due to their extraordinary mechanical, electrical and thermal properties. The large ratio of the strength and elastic modulus of the carbon nanomaterials to that of

polymers makes them an excellent reinforcement [17]. Carbon nanotubes have been shown to significantly improve the mechanical, electrical, and thermal properties of polymer composites [18–20]. Recently, two-dimensional (2D) nanostructures such as graphene nanoplatelets (GNP), graphene, and graphene oxide have emerged as favorable fillers for polymer matrices [21–23]. GNP is promising reinforcement for polymer composites because of its high aspect ratio (length to thickness ratio), unique graphitized planar structure, and low manufacturing cost. Furthermore, the high surface area of GNP results in large contact area with polymer matrix, resulting in good load transfer and improvement in mechanical properties. Single-walled carbon nanohorns (CNH) are recently reported novel materials having a typical diameter of 2 nm and a length of 30–50 nm which have not yet been commercialized. The individual CNH have a tendency to couple together and form stable dahlia flower like particles with a narrow diameter distribution of 80–100 nm [24]. The strengthening derived from nano-fillers depends on their intrinsic strength as well as their shape (aspect ratio). The mechanical behavior of polymer matrix composites also depends strongly on the interface between the filler and the matrix which influences the transfer of the mechanical load and hence is dependent on the shape of the particles. It is therefore expected that CNT, GNP and CNH may not give the same properties in composites.

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Although many studies on CNT and graphene/GNP reinforced epoxy composites are already reported [19,25–36], very limited literature is available on adhesive bonding using CNT–epoxy [2,6–10] and GNP–epoxy [37,38] composites and there is no work on CNH–epoxy composites/adhesive joints. Yu et al. have studied the strength and durability of up to 5 wt.% CNT reinforced epoxy adhesive joints using the Boeing Wedge test under water at 60 °C [2]. They observed that both the bond strength and durability increased with the addition of CNT. The 1 wt.% CNT reinforced joint showed very high fracture toughness ($7.4 \times 10^6 \text{ J m}^{-2}$) compared to pure epoxy joint (1983 J m^{-2}) [2]. The durability of CNT/epoxy adhesive was also found to be better compared to pure epoxy joints. Gerson et al. [7] have studied the effect of CNT on the curing kinetics of epoxy adhesives and found that CNT increased the cross-linking density and glass transition temperature of epoxy (T_g). Srivastava has used 3 wt.% CNT reinforced epoxy to join carbon/carbon (C/C) and carbon/carbon–silicon carbide (C/C–SiC) composites [6]. The C/C–C/C and C/C–SiC–C/C–SiC joints exhibited a 25% and 41% increase in joint strength respectively with CNT reinforcement, compared to pure epoxy joint. To prevent the agglomeration of CNT, Wolf et al. [9] attached a thermally stable protein SP1 to CNT and observed that the CNT/SP1 reinforced adhesive joints showed 50% higher peel strength and 25% higher shear strength over neat epoxy joints. Sydlik et al. have modified CNT using multiple covalent functionalization methods with different zwitterionic surfactants and studied its effect on the shear strength of the adhesive joints [8]. It was observed that the lap shear strength of 1 wt.% functionalized CNT–epoxy joints was 36% and 27% higher than that of neat epoxy joint and non-functionalized CNT–epoxy joint, respectively. Zhang et al. [10] have used silane treated CNT to reinforce epoxy joints of carbon/carbon composites and observed that the silanized CNT were dispersed uniformly and had a good interface strength. The C/C composite joints with 0.2 wt.% silanized CNT had an average shear strength of 10.4 MPa, which was 31% higher than that of the neat epoxy joint. These results indicate that functionalization of nano-fillers can further improve the strength of the joints. But it is to be noted that functionalized CNT are expensive. There have been a few reports recently on use of GNP as reinforcement. Soltannia and Taheri have investigated the effect of different carbon fillers such as CNT, GNP and carbon fibers (CNF) in epoxy adhesive [37]. It was observed that the GNP/epoxy adhesive joints showed increased strength among the three carbon fillers. It was also shown that higher strain rate testing resulted in higher adhesive strength [37]. Guadagno et al. have synthesized tensile butt joints and measured the joint strength of up to 4 wt.% GNP reinforced epoxy adhesive. They found that the 1 wt.% GNP reinforced epoxy had nearly two times strength compared to pure epoxy joint. It was observed that higher addition of GNP lead to poor dispersion leading to decrease in the joint strength [38]. It is seen from literature that the maximum strengthening is observed at an intermediate level of nano-filler addition, which is dependent on the type of nano-filler. It is to be noted that in an adhesive joint, both the cohesive strength of the adhesive and the strength of adhesion with the substrate determine the overall strength. Dispersion of carbon nano-fillers is known to be a challenge due to their large specific surface areas and high viscosity of the epoxy resin. Poor dispersion of nano-fillers is known to result in ineffective strengthening and may also affect adhesion with the substrate and cause premature failure. Several dispersion techniques such as ultrasonication (bath and probe type), shear mixing, calendaring and combination of these techniques have been used to disperse nano-fillers uniformly in polymer matrices with different degrees of success [39]. So, there is a need for novel processes which can result in improved dispersion, especially for high viscosity materials such as epoxy resin.

The objective of present work was to carry out a systematic study of the effect of different carbon nano-fillers, namely CNT, GNP and CNH, on the strength of epoxy adhesive joints. One of the prime novelty of this study is use of Brabender® Plasti-Corder®, which is high energy shear mixer generally employed for blending of thermoplastic materials under controlled temperature conditions, to mix the epoxy and nano-fillers. This is the first study employing it for dispersing carbon nano-fillers in epoxy as per the author's knowledge. It is also a first study on CNH as reinforcement. The effect of the type and nano-filler content on the rheological properties, thermal stability and the strength of the epoxy joints is presented and discussed.

2. Experimental

2.1. Materials

A di-glycidyl ether of bisphenol-A (DGEBA) based two part epoxy adhesive (EP 415, Rotex polymers, Chennai, India) was used in this study. CNT of diameter 10–20 nm and length 10–30 μm were purchased from Cheaptubes Inc. (Texas, USA). GNP having thickness between 3 and 10 nm were obtained from Redex Nano (Ghaziabad, India). The CNH were synthesized in-house by using a DC arc discharge technique. In this method, pure graphite rod of 11 mm diameter and 260 mm length was used as anode. The cathode was a rotating graphite disc of 300 mm diameter. The discharge was carried out in a water cooled stainless steel chamber in helium gas static atmosphere to reduce contamination. The experimental conditions used for synthesis of CNH were 150 A current at 32 V under the pressure of 500 Torr. With an electrode separation of 1 mm, the arc was produced which consumed the anode gradually. The CNH were collected from the inner and upper wall of the reaction chamber. The epoxy mixture and joints containing CNT, GNP and CNH are named as EP–CNT, EP–GNP and EP–CNH respectively.

2.2. Preparation of carbon nano-filler epoxy mixtures

The epoxy resin and carbon nano-fillers were taken in required quantities to prepare 0.2, 0.5, 1, and 2 wt.% nano-filler/epoxy mixture. The constituents were charged into the Brabender® Plasti-Corder® and mixed at 100 rpm for 15 min. The hardener was then added in the requisite weight ratio (100:40 by weight) and mixed manually for 10 min using a Teflon rod. This mixture was used for preparing lap shear joints of AA6061 sheets as described later.

2.3. Rheological studies

Rheological measurements were carried out for carbon nano-filler epoxy mixtures obtained from Brabender® Plasti-Corder® having different filler contents. A stress-controlled rheometer (ANTON PAAR, Physica 301, Germany) having a 25 mm parallel plate geometry and operating in steady mode was used to measure the viscosity. The measurements were carried out with a gap size of 1 mm at 25 °C over a shear rate of 0.1–100 s^{-1} .

2.4. Thermal stability studies

The thermal stability of the cured adhesives was studied using a thermo-gravimetric analyzer (TGA, SDT Q600). The samples were heated to 800 °C at a heating rate of 10 °C/min under nitrogen atmosphere. The weight change as a function of temperature was analyzed and the onset temperatures for degradation were compared.

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