



Comparison of mechanical, electrical and thermal properties in graphene oxide and reduced graphene oxide filled epoxy nanocomposite adhesives



Ruchi Aradhana ^{a,*}, Smita Mohanty ^{a,b}, Sanjay Kumar Nayak ^{a,b}

^a Central Institute of Plastics Engineering & Technology (CIPET), Chennai, Tamil Nadu, India

^b Laboratory for Advanced Research in Polymeric Materials (LARPM), Central Institute of Plastics Engineering & Technology (CIPET), Bhubaneswar, Odisha, India

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ABSTRACT

Reduced graphene oxide (rGO) was synthesized from graphite flakes utilizing improved Hummers method via graphene oxide (GO) precursor. The oxidation of graphite and subsequent reduction of GO were confirmed by using FTIR, XRD, Raman spectroscopy and TEM techniques. The nanocomposite adhesives were developed using GO and rGO with different loading of 0–1.0 wt%. The adhesive strength was studied by conducting lap shear test and it was noticed that, Ep-0.5 GO adhesive exhibited the highest strength among all adhesive formulations and showed ~50% increment than pristine epoxy. Impact strength evidenced noticeable enhancement with decrease in notch depth from 2.54 to 0.5 mm for all adhesive systems clearly indicating brittle to ductile transition due to the declining stress concentration area. The fracture analysis of impact samples and extent of fillers dispersion were visualized by SEM. SEM micrographs evidenced clear surface contrast of samples based on variable notch depth. The electrical resistivity in Ep-rGO systems showed 93% decrease with 1 wt% rGO loading, while thermal conductivity of Ep-0.5 GO revealed 211% increment as compared to pristine epoxy.

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1. Introduction

In the last decades, the polymer conductive adhesives become the promising candidate for rapid development of electronic packaging in modern electronics. Modern electronic components and devices requisite two major factors such as efficient heat dissipation to raise reliability and hinder premature failure [1] and/or effective electron conduction to improve electrical conductivity and provide finer pitch capability [2]. The electrically or thermally conductive adhesives has yielded a novel prospective for electrical interconnection such as flip-chip interconnection, chip-on-board, chip-on-flex, multi-chip modules [3], etc due to the following advantages: lower processing temperatures [2], environment friendliness, easy processing steps, and cost-effectiveness [4], etc. Epoxy resin a significant thermoset material has wide spread usage as conductive adhesive in various

applications i.e. aerospace, die attach, printed circuit boards (PCBs), light emitting devices, solderless interconnections [1], etc owing to their magnificent properties such as excellent adhesion, good chemical and heat resistance, excellent mechanical properties [3] and compatibility with wide range of substrates and additives [5], etc. Although epoxy resin exhibits aforementioned properties, low electrical conductivity (around 10^{-7} to 10^{-14} S/m) and thermal conductivity (T_c) (around 0.1–0.3 W/mK) [1] limit their potential applications in miscellaneous electronic industries. To overcome these limitations, various conductive fillers such as carbon nanotubes (CNTs) [6–8], graphene [1,9,10] and other carbon nanomaterials [11–13] have been employed as powerful reinforcement in epoxy resin.

Graphene a two-dimensional atomically thin layer of sp^2 -bonded carbon atoms considered as an emerging nano-reinforcement for conductive adhesive owing to their large specific surface area, high electrical conductivity ~6000 S/m [14] and high T_c around 4840–5300 W/mK [15]. The synthesis of graphene by chemical reduction method using graphene oxide (GO) as a precursor is the most commonly researched route due to the scalability [16] and large-scale production [17]. Graphene oxide

* Corresponding author. Central Institute of Plastics Engineering & Technology (CIPET), TVK Industrial Estate, Guindy, Chennai, Tamil Nadu, 600032, India.

E-mail address: aradhanaruchi@yahoo.com (R. Aradhana).

(GO) exhibits oxygen functionalities at its edges (carboxyl groups) and on basal plane (epoxide and hydroxyl groups) which disrupts its sp^2 conjugated backbone resulting in lower electrical conductivity. To restore its electrical conductivity, GO can be partially reduced by reacting with reducing agents (i.e. hydrazine and its derivatives) to synthesize reduced graphene oxide (rGO) by extracting oxygen functional groups [9,18,19]. GO and rGO are derivatives of graphene with structural defects caused due to the oxidation and/or reduction processes [17]. These nanomaterials seize attention of scientific researchers as promising reinforcements of epoxy resin for improving thermal and electrical conductivities. Kim et al. [20] investigated the effect of functionalized graphene nanosheets on thermal conductivity of epoxy composites and reported two times increment in conductivity of GO/epoxy composite as compared to the pristine epoxy with 3 wt% GO loading. Shan et al. [21] prepared graphene oxide (GO) reinforced epoxy insulation nanocomposite and studied their thermal conductivity. The authors reported that with addition of 0.5 wt% GO, the thermal conductivity increased from 9.82×10^{-3} to 54.63×10^{-3} W/mK at 6K temperature as compared to virgin epoxy. In addition, Olowojoba et al. [22] examined thermal conductivity of thermally reduced graphene oxide/epoxy composites and 2 wt% rGO inclusion resulted in maximum conductivity (i.e. 0.264 W/mK) at 60 °C whilst pristine epoxy exhibited T_c value of 0.190 W/mK. Various researchers studied the electrical conductivity of rGO reinforced epoxy composites and found tremendous increase even with inclusion of low content loading. Yosefi et al. [19] inspected electrical properties of self-aligned in-situ reduced graphene oxide/epoxy nanocomposites and found an enormous increase with only 0.26 wt% loading of rGO. Sharmila et al. [16] prepared epoxy nanocomposites by incorporating microwave exfoliated reduced graphene oxide (MERGO) and inspected their ac electrical conductivity. The authors reported that epoxy nanocomposite attained 10^{-5} S/m ac conductivity which is adequate for electronics applications. Pokharel et al. [23] studied the electrical percolation threshold of multi-step microwave reduced graphite oxide (RGO)/epoxy composites and found massive increment in electrical conductivity with 0.3 wt% of RGO.

In the current study, reduced graphene oxide (rGO) was synthesized from commercially obtainable graphite flakes by using improved Hummers method through an intermediate product (i.e. graphene oxide (GO)). The synthesis confirmation and structure of GO and rGO were studied by using Fourier transform infra-red spectroscopy (FTIR), X-ray diffraction (XRD), Raman spectroscopy and Transmission electron microscopy (TEM). Thereafter, epoxy nanocomposite adhesives incorporated with GO and rGO, i.e. Ep-GO and Ep-rGO were developed at different loading levels of 0–1 wt%. The adhesive strength of epoxy based nanocomposite adhesives was examined by conducting lap shear test using aluminum substrates. Mechanical properties of all epoxy based adhesives were studied in terms of tensile test and izod impact test. The impact test was examined elaborately by altering the notch depth in the range of 0.5–2.54 mm. The fracture analysis after impact test and nanomaterials dispersion were visualized using Scanning electron microscopy (SEM) technique. The volume, surface resistivity and thermal conductivity of epoxy nanocomposite adhesives were measured by using megaohmmeter and guarded hot plate method. Dynamic Mechanical Analysis (DMA) and Thermo-gravimetric analysis (TGA) techniques were also utilized to study viscoelastic behavior and thermal stability of epoxy-based adhesives.

2. Materials and methods

2.1. Materials

Epoxy resin based on medium viscous diglycidyl ether of bisphenol A (DGEBA) (Araldite GY250) with epoxy equivalent: 170–190 g/kG and low viscous triethylenetetramine (TETA) (Aradur HY951) with specific gravity 0.98 g/cm^3 were acquired from Huntsman Int Pvt. Ltd (Mumbai, India). The chemicals utilized for synthesis of reduced graphene oxide (rGO) such as graphite flakes (282863) with $<20 \mu\text{m}$ size, potassium permanganate (KMnO_4 , 223468, M.W 158, purity ~99%), phosphoric acid (H_3PO_4 , W290017, 85 wt% in H_2O), hydrazine hydrate (N_2H_4 , 225819, 50–60% N_2H_4 , reagent grade) were purchased from M/s Sigma Aldrich Pvt. Ltd. India. Hydrogen peroxide (H_2O_2 , 7722841, 30% in H_2O , ACS reagent) and dichloromethane (CH_2Cl_2 , 75092, purity $>99\%$) were procured from TCI Chemicals Pvt. Ltd. India. Some common chemicals such as conc. sulfuric acid (H_2SO_4 , 98%), hydrochloric acid (HCl, 98%) and acetone were procured from Merck specialists Pvt. Ltd. India. Aluminum substrates with $101.6 \times 25.4 \times 1.62 \text{ mm}$ dimension were used for lap shear testing.

2.2. Synthesis of graphene oxide (GO)

An improved Hummers method was adopted for synthesis of graphene oxide (GO) from graphite flakes ($>20 \mu\text{m}$) [24]. First, graphite flakes (3 g) was added in a mixture of conc. sulfuric acid and phosphoric acid ($\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$) in 9:1 ratio (360:40 ml). Under constant stirring at 0 °C, potassium permanganate (KMnO_4) (18 g) was added slowly to avoid overheating of the mixture. After the addition of KMnO_4 , mixture color turned from black to dark green and the ratio of graphite and KMnO_4 was maintained in the order of 1:6. Then, the mixture was stirred for 12 h at 50 °C and the color changed to dark brown. After the mixture was cooled down to room temperature, it was poured into a mixture of deionized (DI) water (400 ml) and 30% H_2O_2 (3 ml) placed in an ice bath to inhibit the oxidation reaction and the color turned to yellowish brown. The graphite oxide was obtained using centrifugation technique (REMI R-4C, M/s REMI Laboratory Equipments, Mumbai) with 2000 rpm speed for 5 min. The resulting product was then washed with DI water and 30% HCl to maintain a pH of 4–5 and dried overnight at 25 °C. The structure of graphene oxide (GO) is depicted in Fig. 1(a).

2.3. Synthesis of reduced graphene oxide (rGO)

Reduced graphene oxide (rGO) was synthesized from graphene oxide (GO) by using chemical reduction method where hydrazine hydrate (N_2H_4) used as a reducing agent [10]. Initially, GO (500 mg) was dispersed and exfoliated within DI water (500 ml) by using ultrasonication technique (6.5 L capacity, M/s Darsh Technologies, India) for 30 min. After dispersion, hydrazine hydrate (5 ml) was added to the mixture and then subjected to reflux at 100 °C for 10 h. The suspension was diluted with DI water (2000 ml) and dried completely in oven at 120 °C through evaporation. The resulting product was washed in dichloromethane (DCM) by the help of ultrasonication for 5 min and this washing process was repeated for 5 times and the product was then dried at 80 °C for 1 day. The structure of reduced graphene oxide (rGO) after removal of certain oxygen functionalities is presented in Fig. 1(b).

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