INFLUENCE OF GRAPHENE NANOPLATELETS ON MECHANICAL AND THERMO-MECHANICAL PROPRTIES OF GLASS/EPOXY COMPOSITES

VPLIV GRAFENSKIH NANOPLOŠČIC NA MEHANSKE IN TERMO-MEHANSKE LASTNOSTI KOMPOZITOV NA OSNOVI STEKLA IN EPOKSIDNIH SMOL

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The current work focuses on the influence of graphene nanoplatelets (GPs) on the mechanical and thermo-mechanical properties of unidirectional glass/epoxy (GE) composites. GE and GP-GE composites are fabricated using the hand lay-up method followed by compression molding. Different percentages of GPs used in the fabrication of GP-GE composites are (0.1, 0.3, 0.5, and 0.7) w/% by weight of the epoxy. Tests are conducted to experimentally evaluate the uniaxial compressive properties and the coefficient of linear thermal expansion (CLTE) in the direction of fibers. Tests are also conducted to study the thermo-mechanical properties such as storage modulus (E) and loss modulus (E). The variation in the storage modulus and loss modulus is measured from 15 °C to 125 °C, using a dynamic mechanical analyzer (DMA) while the CLTE is measured using a dialatometer. The results indicate that the compressive as well as thermo-mechanical properties increase with the addition of GPs up to 0.5 % and then they decrease. The addition of GPs has no effect on the CLTE of the GE composites in the fiber direction.

Keywords: graphene nanoplatelets, glass/epoxy composites, thermo-mechanical properties, compression, coefficient of thermal expansion

V članku je opisan vpliv dodatka grafenskih nanoploščic (GP) na mehanske in termomehanske lastnosti usmerjenih kompozitov na osnovi steklenih vlaken in epoksida (GE). Kompozite tipa GE in GP-GE so izdelali z ročno tehniko nalaganja, nato je sledilo oblikovanje pod tlakom. Izbrane količine dodanih grafenskih ploščic v kompozitih GP-GE so bile (0,1, 0,3, 0,5 in 0,7) w/g glede na maso epoksija. Po izdelavi kompozitov so določili njihove mehanske lastnosti (tlačno trdnosti) in koeficiente linearnega toplotnega rastezanja (CLTE; angl.: coefficient of linear thermal expansion) v smeri vlaken. Pri testih termomehanskih lastnosti sta bila določena tudi modul shranjevanja (E') in modul izgub (E''). Oba parametra so določili v temperaturnem območju med 15 °C in 125 °C v dinamičnem mehanskem analizatorju (DMA). CLTE so določili s pomočjo dilatometra. Rezultati preiskav so pokazali, da se tlačne kot tudi termomehanske lastnosti kompozitov izboljšujejo do dodatka 0,5 % grafenskih ploščic, nato pa se postopoma slabšajo. Dodatek grafenskih ploščic kompozitom GE ne vpliva na CLTE v smeri vlaken.

Ključne besede: grafenske nanoploščice, kompoziti steklo-epoksidna smola, termomehanske lastnosti, stiskanje, koeficient linearnega toplotnega rastezanja

1 INTRODUCTION

Fiber-reinforced polymer (FRP) composites, due to their superior properties such as high strength-to-weight ratio, high stiffness-to-weight ratio, corrosion resistance and thermal resistance, have been widely used in different industries such as aerospace, marine, construction and automobile industries, and nuclear power plants. Additions of various nanofillers such as carbon nanotubes (CNTs), nanoclay and GPs tend to improve the performance of polymer nanocomposites and fiber-reinforced epoxy composites under different loading conditions. Also, the properties of FRP composites are susceptible to temperature changes.

Graphene oxide, when added to epoxy resin, improved the tensile strength by 11.5 % at 0.3 % and the

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flexural strength by 5.8 % at 0.5 % by weight of the epoxy resin.1 An addition of GPs of less than 0.25 % of the volume of the epoxy resin caused better tensile modulus, lap shear and energy release rate as compared to CNTs, whereas CNTs were found to be effective at relatively higher volume fractions.² The effect of an inclusion of nonfunctionalized or amine-functionalized reduced graphene oxide on the mechanical, thermal and electrical properties of epoxy nanocomposites is reported in reference.³ No significant difference in the performance of the two types of nanofillers was reported. However, an increment in properties with the addition of nanofillers compared to neat epoxy resin was reported. The effect of GPs on the CTE of polyetherimide thermoplastic composites is shown in reference.⁴ It is reported that the GPs are very effective in reducing the CTE in the three orthogonal directions. The effects of additions of GPs with different surface areas (300, 500 and 750) m²/g on the

mechanical and thermal performance of mixed silicate with different ratios of alkali silicates were studied.⁵ The GPs varied from (1, 5 and 10) w/% of the mixed silicate. It was reported that the lap shear strength increased by 25.6 % with the addition of 10 % of GPs with a surface area of 750 m²/g. It was also reported that the increase in the surface area of the GPs changed the mixed silicate from brittle to ductile, leading to a reduction in the hardness compared to the polymer without GPs.

The polypropylene reinforced with short glass fibers and GPs, either individually or in combination, resulted in an increment in the mechanical and thermal performances.⁶ However, the hybrid composite with a simultaneous addition of both fillers performed better than the composites with individual ones. The addition of different nanofillers such as CNTs, GPs and the combination of CNTs and boron nitride nanosheets or CNTs and boron nitride nanotubes were shown to improve the tensile and the fracture performance of GE composites.7 Thermal properties of GE composites were measured experimentally to study the effect of the addition of different volume fractions of various carbon-based nanofillers such as graphene oxide, reduced graphene oxide, GPs and CNTs.8 Enhancements in the thermal conductivity by 8.8 % with 0.3 % MWCNTs, 12.6 % with 1 % GPs, 8.2~% with 2 % graphene oxide, and 4.1 % with 0.042 %reduced graphene oxide were observed. However, with the same volume fraction of nanofillers, the GPs resulted in the highest improvement in the properties. Glass/vinyl ester composites with CNTs were produced using a high-pressure injection and the addition of 3 % nanofillers was shown to improve the thermal conductivity by 1.5 times.⁹ It can be found in the literature that the mechanical, thermal and thermo-mechanical data available on the GP-GE composites at different GPs content are limited. In this context, the current study focuses on experimental work to investigate the impact of GPs on the CLTE, compressive and thermomechanical properties such storage modulus and loss modulus of GE composites.

2 EXPERIMENTAL STUDY

To experimentally evaluate the effect of an addition of GPs on the mechanical, thermal and thermo-mechanical properties, GE and GP-GE composites were fabricated using the hand lay-up method followed by compression molding. Commercially available epoxy LY556, hardener Araldite HY951 and glass fibers having an area density of 740 g/mm² were used for the fabrication. Four layers of glass-fiber fabric were used for each laminate. The GPs were procured from PlasmaChem GmbH, having a purity of 91 %, a thickness of 1–4 nm, a width of up to 2 μ m, and a specific surface area of 700–800 m²/gm.

To prepare a blend of epoxy/GPs, the GPs were initially added to ethanol and sonicated for one hour. The mixture was then transferred to a stirrer and shear mixed at 500 min⁻¹ for one hour. This ensured the dispersion of the GPs into ethanol. The epoxy resin was then added to the ethanol/GPs mixture, which was shear mixed for 6 h on a hot platform at 70 °C before being utilized in the fabrication of GP-GE composites using the hand lay-up method followed by compression molding. The thickness of each laminate fabricated was around 3.2 mm. The composite laminates were then cured at room temperature for 72 h before testing. Specimens of the required dimensions for various tests were cut using a waterjet cutting machine. The sizes of compressive, CLTE and DMA test specimens were (142×14) mm, (40×10) mm and (55×10) mm, respectively. Thermo-mechanical tests were performed using a dynamic mechanical analyzer (DMA) from NETZSCH, Germany. Uniaxial compression tests were performed using a test fixture on a universal testing machine with a capacity of 400 kN, according to ASTM D6641, whereas CLTE tests were performed in a dialatometer.

3 RESULTS

The DMA was used to determine the storage modulus (E') and loss modulus (E'') of the GE and GP-GE



Figure 1: Storage modulus (E^{*}) and loss modulus (E^{*}) of GE and GP-GE composites

Table	1:	Com	pressive	strength	and	modulus	of	GE and	GP-GE	composites
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	Neat	0.1 % GPs	0.3 % GPs	0.5 % GPs	0.7 % GPs
Strength (MPa)	203 ± 6.6	225 ± 9.4	241 ± 5.6	254 ± 8.5	245 ± 5.9
Modulus (GPa)	21.9 ± 0.22	23.3 ± 0.49	25.7 ± 0.78	28.2 ± 0.92	26.6 ± 0.24

composites that are given in Figures 1a and 1b. The analysis was performed under three-point bending from 15 °C to 125 °C at a rate of 5 °C/min and a frequency of 5 Hz. It can be seen from Figure 1a that the storage modulus increased with the addition of GPs up to 0.5 %and then decreased with further addition. A similar behavior can be observed at the peak of the loss-modulus curves shown in Figure 1b. The storage modulus of the GP-GE composites in the pre-glass transition stage (below 70 °C) is higher than that of the GE composites; however, it is lower for the GP-GE composites in the post-transition (beyond 80 °C) stage, with 0.1 % and 0.3 % GPs content. The storage modulus at 0.5 % GNs is 30.2 % higher than that of the GE composites at 15 °C. The glass-transition temperature (T_g) of the material can be found from the E' vs T curve. The glass-transition temperature is the temperature at which composites change from a glassy to a rubbery phase. The $T_{\rm g}$ of the GE composites is found to be 70 °C. The $T_{\rm g}$ value increased with the addition of GPs from 0.1 % to 0.5 %and then decreased. However, a slight increment in the $T_{\rm g}$ value can be observed for the GP-GE composites compared to GE composites.

The stress-strain curves for the GE and GP-GE composites under a compressive load are presented in **Figure 3**. The compressive strength is calculated from the peak load carried by the specimen, whereas the modulus is obtained from the slope of the stress-strain curve. The compressive strength and modulus of the GE and GP-GE composites are given in **Table 1**. It can be observed that the compressive strength and modulus of the composites increase with an increase in the GPs content up to 0.5 % and then decrease with further addition of GPs. The



Figure 2: Stress-strain curves for GE and GP-GE composites under compression

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compressive strength of the GE composites is found to be 203 MPa. The strength of the GE composites increases by (10.8, 18.7, 25.1 and 20.7) % with the additions of (0.1, 0.3, 0.5 and 0.7) % GPs, respectively. The compressive modulus of the GE composites is 21.9 GPa. The modulus of the GE composites increases by (6.4, 17.4, 28.8 and 21.5) % with the additions of (0.1, 0.3, 0.5 and 0.7) % GPs, respectively. The reduction in the compressive properties at 0.7 % GPs is due to the agglomeration of nanoparticles at higher contents.

Tests were performed on the dialatometer to study the effect of the GPs content on the CLTE of the GE composites in the fiber direction from room temperature to 70 °C. The size of the specimens used for measuring the CLTE was (40 × 10) mm. The CLTE value is dominated by the property of the glass fibers in the longitudinal direction. Hence, the addition of GPs did not affect the thermal expansion property of the GE composites in the fiber direction. The CLTE for the GE and GP-GE composites measured in the fiber direction is found to be 9×10^{-6} /°C.

4 DISCUSSION

As GPs allow superior properties, the effects of different weight fractions of GPs on various properties such as mechanical, thermal and thermo-mechanical properties of GE composites are analyzed in the current study. The properties evaluated are the compressive strength, modulus, CLTE and thermo-mechanical properties in the flexural mode. The amounts of GPs considered in the study are (0.1, 0.3, 0.5 and 0.7) % of the weight of the epoxy resin. GE and GP-GE composites are fabricated using the hand lay-up method followed by compression molding. The variation in the storage modulus and loss modulus is measured from 15 °C to 125 °C at a rate of 5 °C/min and a frequency of 5 Hz. Uniaxial compression tests are carried out to measure the compressive strength and modulus of the GE and GP-GE composites. The CLTE of the GE composites in the fiber direction is measured by conducting tests on the dilatometer.

5 CONCLUSIONS

The effects of the incorporation of GPs on different properties such mechanical, thermal and thermo-mechanical properties of the GE composites fabricated using the hand lay-up method followed by compression molding are studied. The addition of GPs increases the storage modulus and loss modulus of the GE composites up to 0.5 % before decreasing. The increase in the properties is a result of a good distribution of GPs in the GE composites. The agglomeration of GPs at higher weight fractions resultes in a reduction in the properties. A similar behavior is observed for the strength and modulus of the composites under compression. The addition of GPs has no effect on the CLTE of the GE composites in the fiber direction.

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