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Influence of adding nanomaterials on shear properties of epoxy resin at different temperatures

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A R T I C L E I N F O

Article history:
Received 12 January 2023
Accepted 6 May 2024
Available online
6 May 2024
Keywords:
Single-lap adhesive joints
Nanofiller
Epoxy resin
Nanocomposite

A B S T R A C T

Adhesive joints play a vital role in different industries owing to their advantages and ease of application compared to other joining methods. This research focuses on enhancing the mechanical properties of epoxy adhesives by incorporating graphene nanoplatelets (G) and iron-oxide nanofillers (Fe_3O_4). Single-lap adhesive joints, including both G and Fe_3O_4 nanoparticles, are fabricated at 2%, 3%, and 4% weight percentages and tested under tensile load at ambient, 45°C, and 88°C. The results reveal that adding G and Fe_3O_4 nanofillers enhances shear strength at elevated and room temperatures without altering the epoxy glass transition temperature (*Tg*). Furthermore, G nanofiller performs better in improving shear strength than Fe_3O_4. The optimal weight percentage is identified as 3 wt% for G and Fe_3O_4, as higher percentages lead to decreased shear strength due to agglomerations. This study provides insight into tailoring epoxy adhesives for improved mechanical performance under varying temperature conditions.

1. Introduction

Adhesively bonded joints are gaining significant attention across various industries due to their advantage over traditional joints. These joints offer many advantages, including exceptional strength that surpasses conventional joints, low fabrication cost, and low structural weight (Khoramishad et al., 2016). Many community orientations worldwide use lead-free adhesive materials in electronic applications due to toxic effects that are risky to human health. In addition to lead-free applications, recently, there have been motivations for finding alternative die-attached approaches for the development of silicon carbide technologies in replacement of silicon technologies. This means that silicon carbide technologies lead to good performance when working at high temperatures when powerful, and at high voltages. Many industrial applications subject electronics to high temperatures in terms of their working temperature ranges. The low-temperature joining (TLTJ) technique has been executed and can be replaced with lead-free materials from die-attached materials and high-temperature applications (Schwarzbauer, Kuhnert, 1991). Epoxy thermosets find wide-ranging applications across various industries, such as electronic packaging circuit encapsulation, construction, wind turbines, automotive, marine, and aerospace industries. These conventional epoxy thermosets offer many benefits, including lightweight properties, minimal shrinkage, and excellent chemical and corrosion resistance. The pioneers used this technique to create joints in micron-scale die attachments from silver paste in power electronics applications at the end of 1980. Since then, the technologies have attracted the attention of specialists who work in this area (Khotbehsara et al., 2020). Graphene is an analog of a giant aromatic ‘‘polymer molecule’’ possessing optical absorption properties, mechanical strength, and high electrical conductivity. It can enhance electrical conductivity and mechanical strength by incorporating into inorganic systems and polymers (Siow et al., 2012). Loh et al. (2010) and Wang et al. (2007) demonstrated that increasing the temperature of the sintering process, up to 40% of the melting temperature, can lead to improved mechanical properties. This enhancement was generally observed to be more significant than the effect of sintering curing time.

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ISSN 2291-8752 (Online) - ISSN 2291-8744 (Print)

doi: 10.5267/j.esm.2024.5.002

The duration of the curing process influences the shear strength of a joint. When longer holding times are employed at higher temperatures, adhesion is enhanced through diffusion within the joint region, resulting in improved interconnection. Consequently, increasing both the temperature and holding time positively affects the joint's shear strength. This is achieved by promoting a more effectively sintered joint with increased density and superior adhesion at the interface (Khazaka et al., 2014; Knoerr and Schletz, 2010). A faster heating rate must be employed during the processing time to achieve a denser joint and higher shear strength. Interestingly, polymer composites require lower amounts of graphene oxide (GO) fillers than traditional mineral fillers to enhance mechanical properties (Li et al., 2013; Hou et al., 2020). The reported tensile strength and modulus of multiple layers of graphite are 300 nN and 0.5 TPa, respectively (Frank et al., 2007). Adding graphene nanoplatelet to epoxy at a weight ratio of 0.1 wt.% enhances the tensile strength by 40% and the tensile modulus by 31%, as reported by Rafiee et al. (2009), in comparison to the unfilled epoxy. The thickness of individual GO sheets ranges from approximately 0.7 nm to 1.5 nm, with an average thickness of 1 nm (Suk et al., 2010; Gudarzi & Sharif, 2012; Stankovich et al., 2007). Researchers have suggested that certain approaches could be modified to produce GO at a lower cost using less sophisticated techniques, which would facilitate scale-up for mass production (Bora et al., 2013). The Fe3O4 material operates at high temperatures, typically around 60–90°C. The annealing temperature determines the material's size, shape, and thermal properties (Sánchez-Romate et al., 2022). Khoramishad et al. (2018a) studied the effect of temperature on the shear strength of MWCNT-epoxy nanocomposite adhesive joints experimentally. They reported that when the testing temperature was increased, the improving effect of MWCNTs was decreased. In another study, Khoramishad et al. (2018b) studied the impact of graphene oxide nano-platelets (GOPs) on the strength of nanocomposite adhesive joints across varying temperatures, up to the adhesive's glass transition temperature. Results showed that GOPs influenced joint strength differently depending on temperature, with their enhancing effect diminishing as temperature increased. Beyond a critical temperature, GOPs even led to decreased joint strength compared to pure adhesive, with the critical temperature varying based on the weight percentage of GOPs, ranging from 60°C for 0.1 wt% GOPs to 40°C for 0.3 wt% GOPs.

Extensive research has focused on exploring the unique electrical, thermal, optical, electronic, and mechanical properties of metal nanofillers and nanocomposites (Wilson et al., 2004; Yan et al., 2012). Among the various magnetic nanomaterials, there has been significant interest in Fe3O4 nanofillers due to their exceptional characteristics (Frounchi & Hadi, 2013). Magnetite Fe_3O_4, being a metal oxide, holds potential as an electrode material because of its easy redox reactions, cost-effectiveness, and minimal environmental footprint, as noted by Shi et al. (2011) and Liang et al. (2011). However, the low electrical conductivity of Fe_3O_4 can impede effective ion diffusion, resulting in low capacitances (Du et al., 2009). To overcome this limitation, developing hybrid nanostructured electrodes has gained attention as an effective approach to enhance conductivity. In this strategy, carbon materials with high surface area and conductivity serve as a conductive network (Wu, 2010; Chen, 2010). Fe3O4 nanofillers can be easily synthesized and integrated with carbon hosts through direct connections, enabling the fabrication of these hybrid nanostructured electrodes (Shi et al., 2011; Mu et al., 2011). The current research focuses on enhancing the mechanical properties of epoxy adhesives by incorporating graphene nanoplatelets (G) and iron-oxide nanofillers (Fe_3O_4), under elevated temperatures. The experimental work conducted in this research involves the fabrication and characterization of epoxy nanocomposite adhesives filled with varying weight percentages (2%, 3%, and 4%) of G and Fe_3O_4 nanofillers. The study assesses the impact of different nanofillers (G, Fe_3O_4) and weight percentages on the adhesive joint performance through shear tests conducted at ambient, 45 °C, and 88 °C temperatures. The fracture surfaces of the adhesive joints were examined using SEM micrographs.

2 Materials and specimens' fabrication

2.1 Materials

The adhesive was fabricated using an epoxy resin provided by the UHU Endfest 300 company. The resin's viscosity is 37.000 mPa.sec, and its density is 1.12 gr/cm^3 (UHU plus). According to the resin data sheet, a weight ratio of 100:50 between the resin and hardener is used in this research. The specifications of UHU Endfest 300 adhesives are presented in **Table 1**.

Table 1. UHU plus endfest 300 adhesive specifications.

UHU endfest plus 300	
Chemical basis	Epoxy resin
Density	Binder: approx. 1.2 $\left(g/cm^3\right)$ Hardener: approx. 0.96 $\left(g/cm^3\right)$
Viscosity	Binder: 40000 $\left(mPa.sec\right)$ Hardener: 30000 $\left(mPa.sec\right)$
Working life $\left(20\,^{\circ}C\right)$	90mins
Mixing ratio (by weight)	1:1 (other mixing ratio possible)

The current research uses G and Fe_3O_4 as primary nanofillers. The G has an average particle size of 6-8 nm and a surface area of 120-150 m^2/g, a carbon content of 99.5% provided by Skyspring Nanomaterials, Inc. manufactures in the USA. Table 2 lists the properties of G. On the other hand, the Fe_3O_4 has a purity of 98%, an APS of 20-30 nm, a specific surface area SSA of 40-60 m^2/g, and a density of 4.8-5.1 g/cm^3. They also have a spherical morphology and appear as dark brown Nanopowder. Table 3 shows the compounds and properties of Fe_3O_4 particles provided by Skyspring Nanomaterials, Inc. manufactures in the USA.

Table 2. Properties of G nanofillers

specific area of surface	120-150 m^2/g
Thickness	6-8 nm
Outer diameter	15 µm
content of carbon	99.5+%

Table 3. Properties of Fe_3O_4 nanofillers

Component	Fe_3O_4	SO_4	SiO_2	Mg	Mn	Na	Ni	Cr	Pb	Ca	Cl
Wt. %	>98+	0.1	0.2	0.01	0.115	0.01	0.01	0.2	0.005	0.2	0.2

This research utilizes Aluminum 7075 as the substrate material. The Modulus of Elasticity of the material is 72 GPa, and the ultimate tensile strength ranges from 462 to 538 MPa. The substrates were cut from an Aluminum 7075 sheet measuring 76.1mm × 25.4mm × 1.7mm. Good adhesion of the substrates is crucial, and to achieve this, the substrates must be well-prepared by washing them with liquid soap and water and then placing them in the orthitinol solution for 30 minutes. Finally, the substrates should be washed with lukewarm water and dried with a cotton cloth. Table 4 shows the chemical composition of Aluminum 7075.

Table 4. Chemical composition of AL7075

Component	Si	Fe	Mg	Al	Zn	Ti	Cr	Cu
Wt. %	40	0.5	2.1-2.9	remain	5.1-6.1	0.2	0.18-0.28	1.2-2

2.2 Specimen fabrication

To study the effect of nanofillers on the mechanical and electrical properties of adhesive joints, single-lap joints (SLJs) were fabricated with nanocomposite adhesives. Two types of nanocomposite adhesives, G and Fe_3O_4 nanofillers, were dispersed into the epoxy adhesive material to prepare nanocomposite adhesives. The dispersing process of nanofillers in the epoxy adhesive involved multiple steps. First, to reduce the high viscosity of the resin at 37.000 mPa.sec, the resin was heated to 50°C before adding the nanofillers. After introducing nanofillers to the epoxy adhesive at varying weight percentages of 2%, 3%, and 4%, the mixture was subjected to mechanical mixing at a speed of 180 rpm for 30 minutes. To further enhance the dispersion of the nanofillers, an ultrasonic process was carried out using an ultrasonic solicitor for 1 hour. The ultrasonic solicitor operated at a power of 250 watts with a 5-second on/off cycle to minimize heat generation during sonication. The mixture was placed in a water and ice mixture.

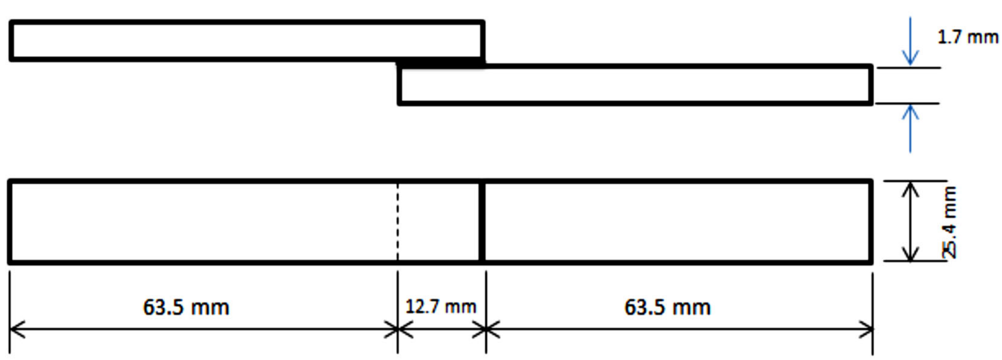

Fig. 1. Single-lap joint specimen dimension according to ASTM-D1002

Following the ultrasonic treatment, the mixture underwent a vacuum process for 30 minutes to eliminate any trapped air bubbles. Subsequently, the curing agent was added to the mixture at a weight ratio of 100:50 and mechanically mixed for 15 minutes at 180 rpm. Once again, any remaining air bubbles were removed by subjecting the mixture to a vacuum. The reinforced adhesive was applied onto aluminum substrates and the specimens were placed in a manufacturing fixture to ensure proper alignment and sufficient pressure during the curing process. The curing took place in an oven at 45°C for 157 minutes. The aluminum substrates, made of aluminum 7075 and cut from a 1.7 mm thick aluminum sheet, underwent surface preparation to promote strong adhesion with the adhesive. This process involved washing the substrates with liquid soap and water, immersing them in an ethanol solution for 30 minutes, rinsing them thoroughly with warm water, and drying them. The dimensions of SLJ are shown in **Fig. 1**. The dimensions and testing conditions of SLJs were considered according to standard ASTM D1002.

3. Tests

3.1 Glass Transition Temperature Tg

The glass transition temperature (*Tg*) test is performed using a Differential Scanning Calorimeter DSC-63 device according to ASTM E1356 (ASTM E1356, 2014). The *Tg* of the pure epoxy samples and epoxy mixed with graphene nanofiller and iron oxide at 4 wt% weight percentage are obtained.

3.2 Shear Strength Test

To investigate the mutual effect of the nanofillers and temperature on the shear strength of the epoxy adhesive tensile tests are performed according to ASTM –D1002 using a universal SANTAM (BONGSHIN) STM-150 test machine at a loading rate of 5 mm/min. End tabs, cut from the same aluminum sheet, are bonded to the specimen using adhesive to eliminate load path eccentricity, out-of-plane bending moments, and non-uniform shear stresses in the adhesive layer. Tensile tests are conducted at three ambient temperatures, 45 °C and 88 °C. **Fig. 2a** and **Fig. 2b** show the tensile test machine and the test specimens.

The shear strength of the adhesive joint is calculated using the following equation.

$$\tau = \frac{p}{lb}, \tag{1}$$

where P is the applied load, b is the joint width, and l is the overlap length.

Fig. 2. Tensile test machine (a) and single-lap adhesive joint test samples (b)

4 Results and discussion

4.1 DCS results of adhesive

Fig. 3 shows the Differential Scanning Calorimeter (DCS) results for the epoxy adhesive with 4%wt Fe_3O_4 nanofiller. In the current research, *Tg* of the pure epoxy, as well as the epoxy filled with 4%wt of graphene and Fe3O4 nanofillers, are extracted. *Tg* for the pure epoxy is 87.9 °C and for the epoxy mixed with graphene and iron oxide were 87.1 °C and 88.88 °C, respectively. It was observed that the obtained *Tg* values for the pure epoxy and the filled epoxy are close to each other. This could be due to the low filler percentage used in the current research, which does not affect the pure epoxy *Tg* properties.

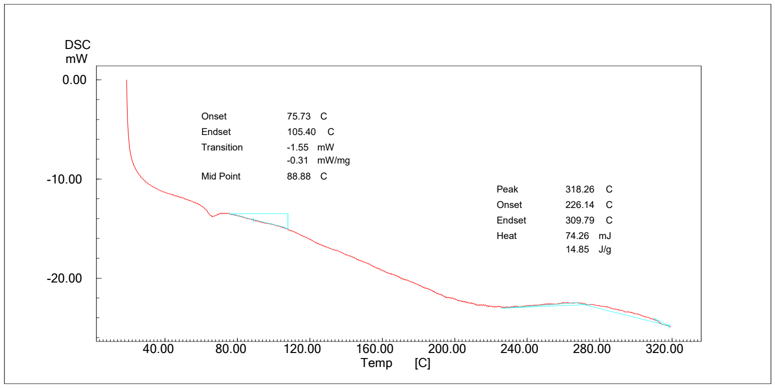

Fig. 3. *Tg* of the epoxy resin filled with 4 wt%, Fe_3O_4 nanofillers

4.2 Shear Stress Results

Fig. 4 shows the pure and filled epoxy resin test results with 2, 3, and 4 wt % of the graphene nanofillers at room temperature. The strength and stiffness of the pure resin increased by adding nanofillers. The maximum strength belonged to the 3 wt% samples was 15.9 MPa. The strength value for the samples with 4 wt % fillers was reduced to 15.13 MPa, possibly due to the agglomeration of the nanofillers at higher filler percentages. The strength for the pure resin and resin with 2 wt% were obtained 10.9 MPa and 15.75 MPa, respectively.

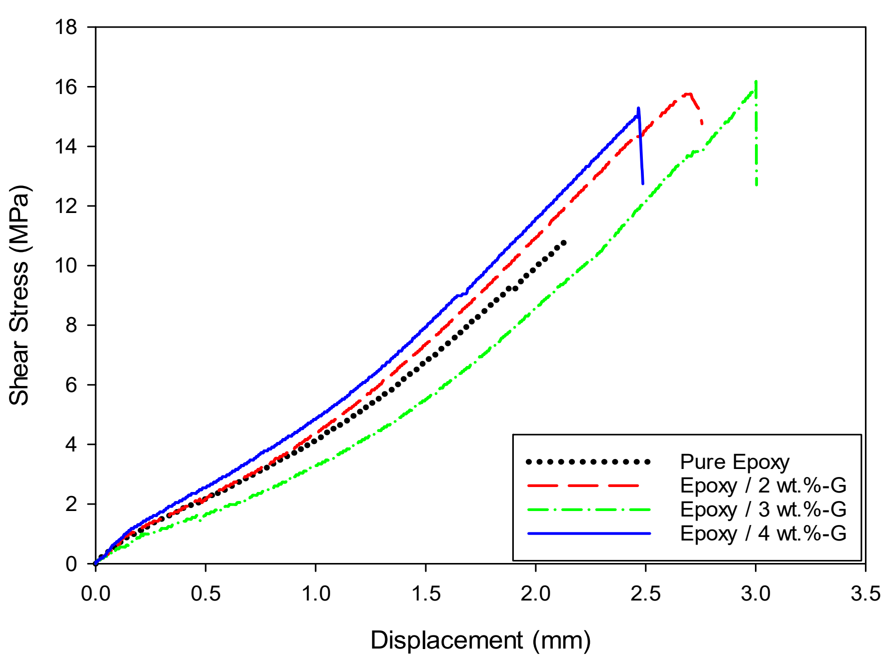

Fig. 4. Stress-Displacement curve of the Epoxy-Graphene composite with different weight fractions at 25°C.

Fig. 5 presents the test results of the graphene/epoxy samples at 45°C. Samples containing 3 wt% nanofillers exhibited the highest strength, measuring 16 MPa, while pure epoxy measured 8.9 MPa. The samples with 2 wt % and 4 wt % of nanofiller had shear strengths of 15.87 MPa and 15.56 MPa, respectively.

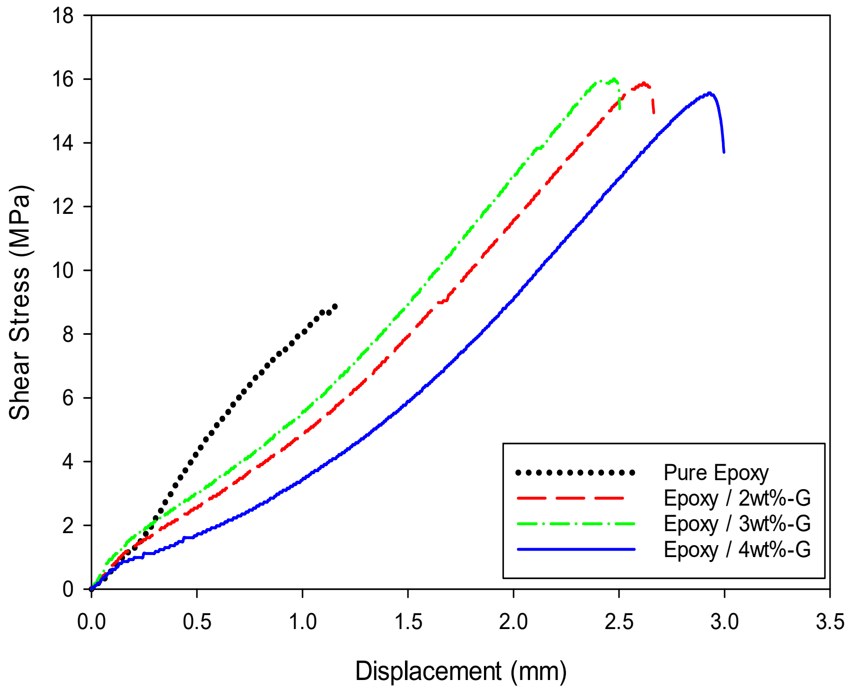

Fig. 5. Stress-Displacement curve of the Epoxy-Graphene composite with different weight fractions at 45°C.

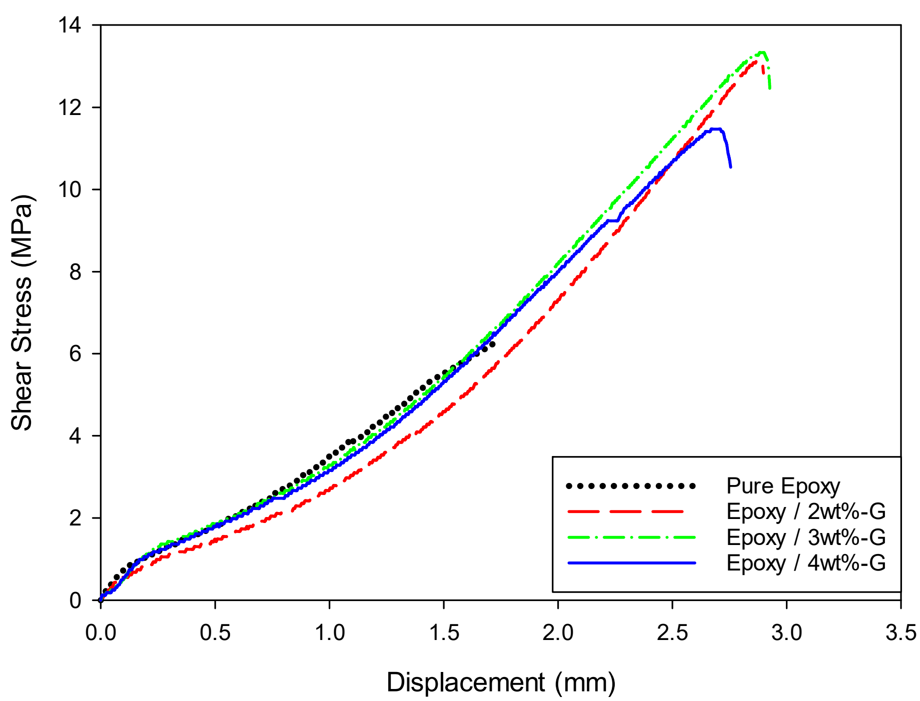

Fig. 6. Stress-Displacement curve of the Epoxy-Graphene composite with different weight fractions at 88°C.

Fig. 6 shows the results of the tests conducted on the samples filled with G at 88 °C. The strength of the pure epoxy at this temperature decreased to 6.3 MPa. Among the specimens tested at this temperature, those containing 3 wt% nanofillers exhibited the highest shear strength of 13.3 MPa. Specimens containing 2 wt% and 4 wt% nanofillers showed strengths of 13.14 MPa and 11.47 MPa, respectively.

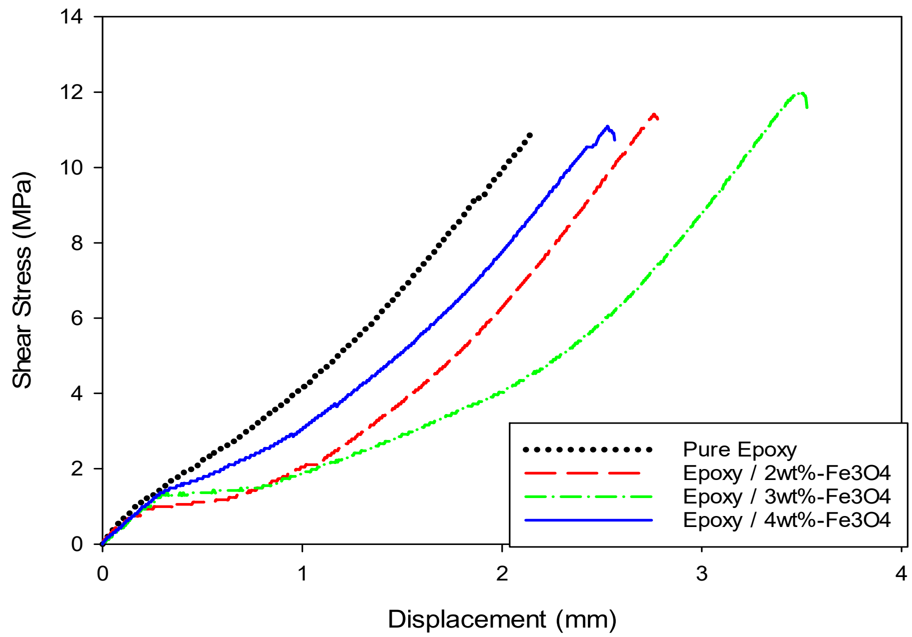

Fig. 7. Stress-Displacement curve of the Epoxy-Fe_3O_4 composite with different weight fractions of Fe_3O_4 nanofillers at 25 °C

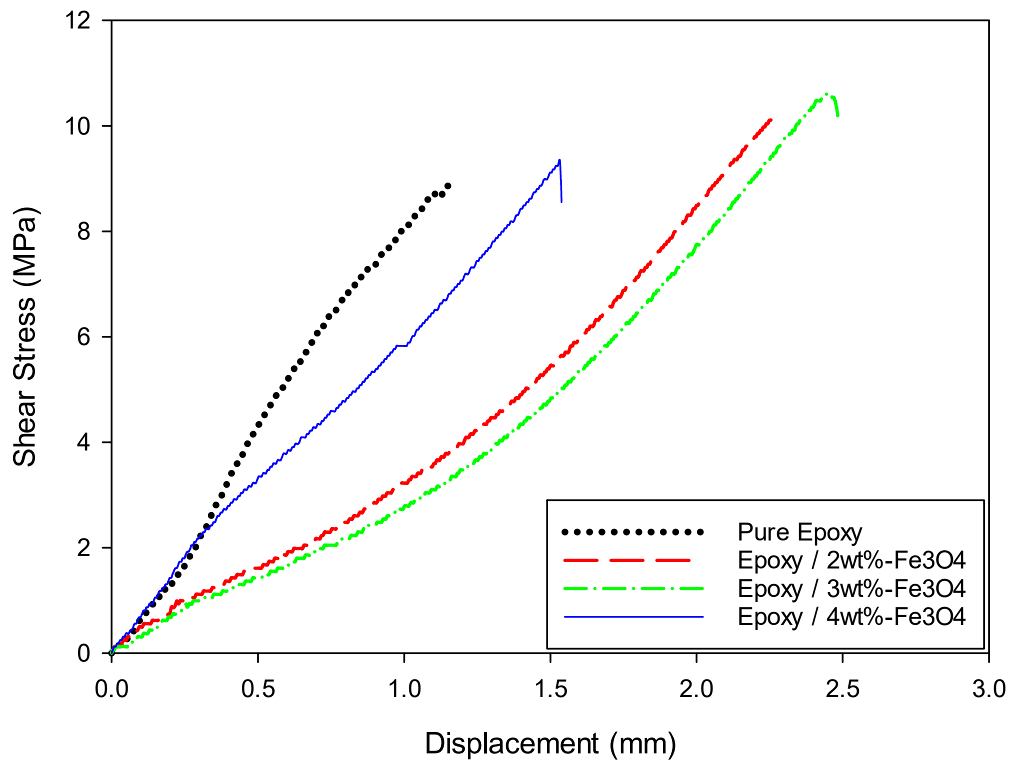

Fig. 8. Stress-Displacement curve of the Epoxy-Fe_3O_4 composite with different weight fractions of Fe_3O_4 nanofillers at 45 °C.

It was observed that an increase in temperature to 88 °C caused a decrease in the strength of the specimens. The strength reduction percentage was as follows: 42.2% for pure, 16% for 2 wt%, and 16.1% and 25.4% for 3 wt% and 4 wt%, respectively, when compared to the strength at room temperature. The decrease in adhesive joint bond strength with rising testing temperature can be attributed to the interaction between viscoelastic properties and crosslinking reactions. Elevated temperatures alter the viscoelastic properties, influencing viscosity, damping characteristics, and molecular mobility, potentially leading to increased deformability and reduced mechanical properties. The temperature dependence of crosslinking reactions is also critical, as higher temperatures accelerate reaction kinetics but may also create weaker regions within the material. **Fig. 8** shows the stress-strain response curve for the epoxy-filled resin at a temperature of 45 °C. At a filler weight ratio of 3%, the maximum shear strength achieved was 10.6 MPa. Meanwhile, the shear strength achieved at 2% and 4% weight ratios of Fe_3O_4 nanofillers was 10.17 MPa and 9.3 MPa, respectively.

Figs. 7-9 present the stress-strain response curve for the epoxy resin filled with Fe_3O_4 nanofillers at different temperatures of 25°C, 45°C, and 88 °C. According to Fig. 7, the specimens containing 3 wt% nanofillers have a maximum strength of 12 MPa. The other samples with 2% and 4% of the Fe_3O_4 nanofillers show shear strength values of 11.4 MPa and 11.1 MPa, respectively. **Fig. 9** shows that the epoxy specimens filled with 3 wt % of the nanofiller at 88 °C have the highest shear strength of 10.1 MPa. The shear strength of the samples filled with 2% and 4% of the nanofiller are 9.42 and 7.87 MPa, respectively.

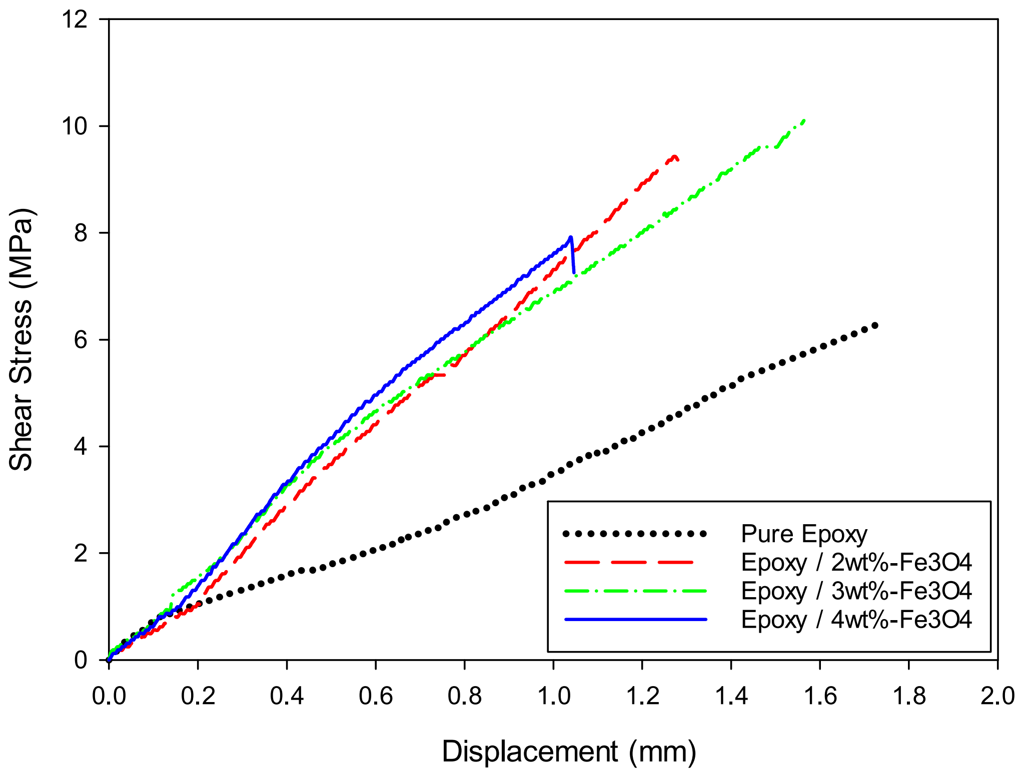

Fig. 9. Stress-Strain curve of the Epoxy-Fe_3O_4 composite with different weight fractions of Fe_3O_4 nanofillers at temperature test 88 °C.

We note that at an elevated temperature of 88 °C, the strength was reduced by 42.2% for pure,17.4 % for 2 wt% and 16 %, and 29.2% for 3 wt% %and 4wt%, respectively, compared to the room temperature strength in the specimens. This reduction observed in the strength of adhesive joints as the testing temperature increases can be explained by the interaction between viscoelastic behavior and crosslinking reactions. When exposed to elevated temperatures, the viscoelastic properties of the adhesive material are altered, affecting its viscosity, damping characteristics, and molecular mobility. These changes can result in increased deformability and a decrease in mechanical properties. The temperature dependence of crosslinking reactions is another important factor to consider. At higher temperatures, the reaction kinetics are accelerated, which can lead to faster crosslinking. However, it is also possible for weaker regions to form within the material due to the intense heat. Therefore, the overall strength of the adhesive joint can be influenced by the changes in viscoelastic behavior and the temperature-dependent crosslinking reactions. Variations of the shear strength for the epoxy adhesive filled with graphene nanofillers at 2, 3, and 4 wt% as a function of test temperature is depicted in Fig. 10; the graph indicates that the rate of strength reduction in pure epoxy is higher than that of filled epoxy. This confirms that temperature has a more harmful impact on pure epoxy than on filled epoxy. This could be attributed to the rapid heat distribution in the epoxy resin containing conductive nanofillers.

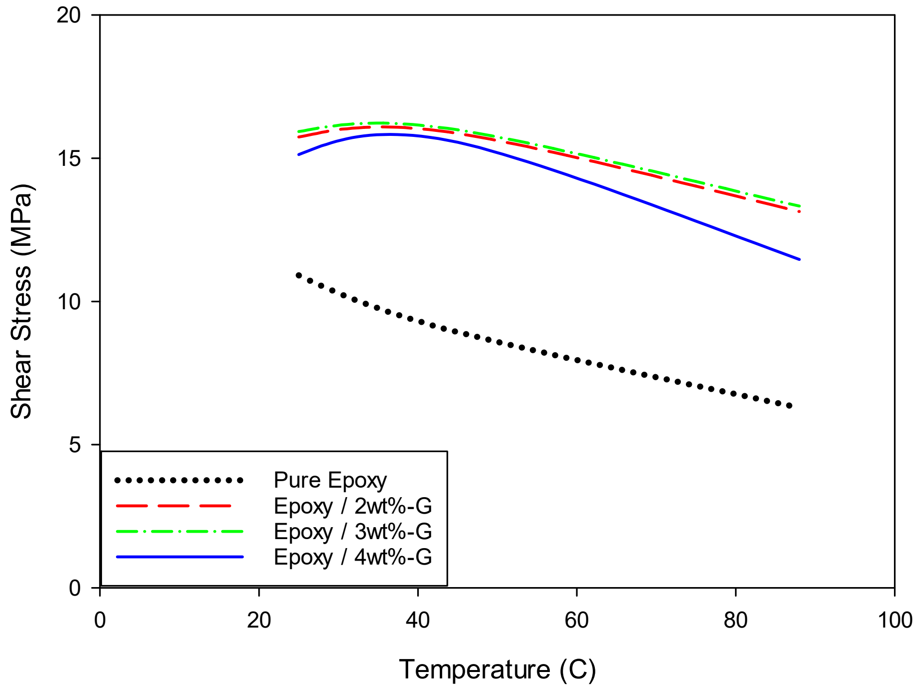

Fig. 10. Shear Strength of the Epoxy-Graphene Composite under various temperatures

Fig. 11 illustrates the impact of Fe_3O_4 nanofillers on the shear strength of epoxy resin at different temperatures. The strength of the samples deteriorates due to temperature, but the degradation rate is lower in nanofilled specimens than in pure specimens. This can be attributed to adding thermally conductive nanofillers to the specimens.

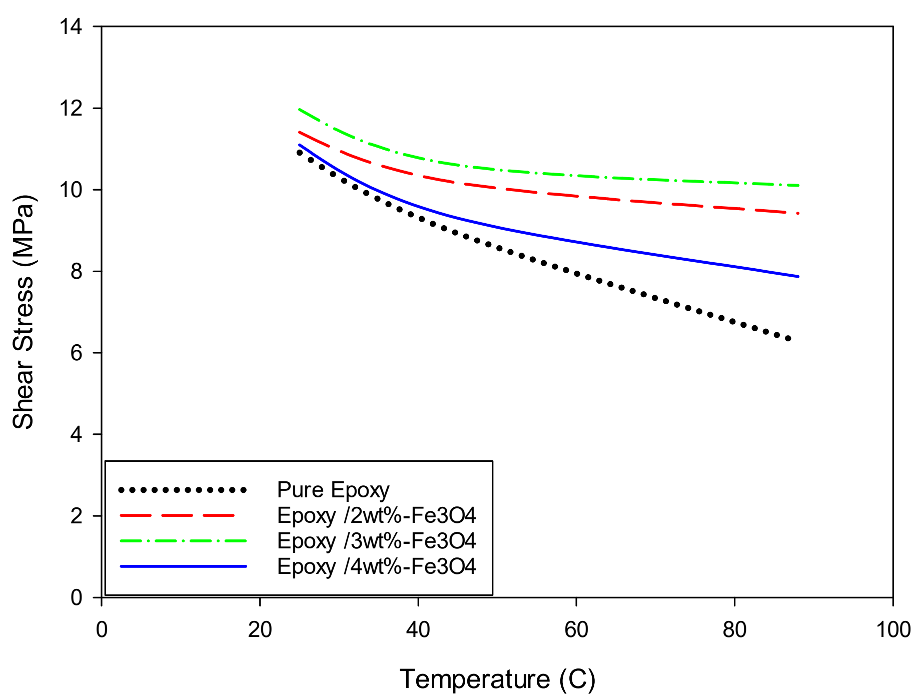

Fig. 11. Shear Strength of the Epoxy-Fe_3O_4 Composite under various temperatures

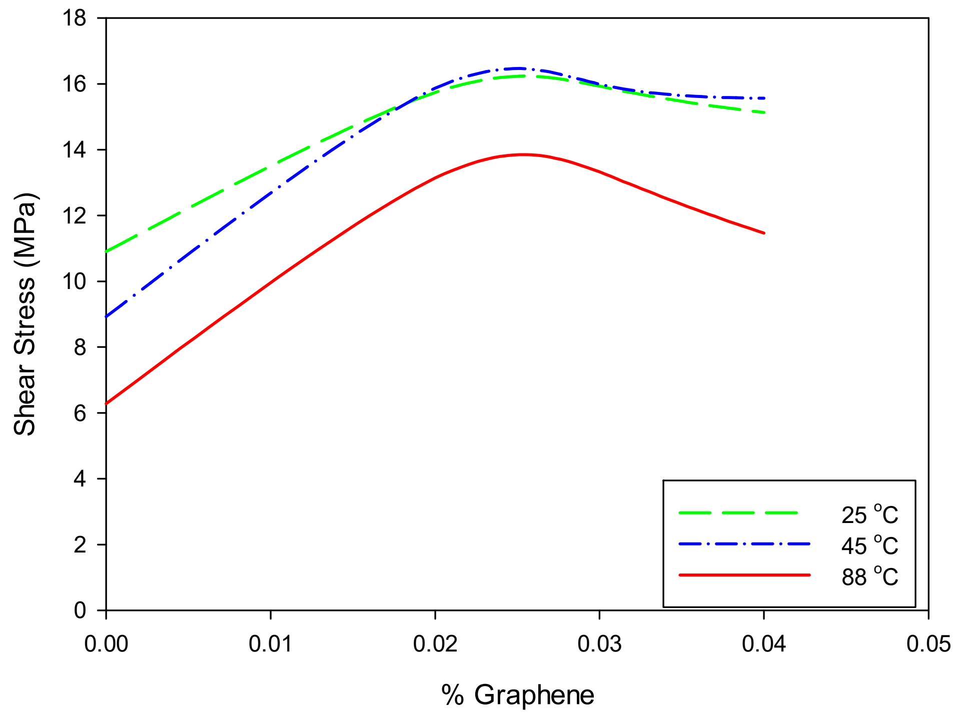

Fig. 12. Shear Stress of Epoxy-Graphene Composite with different weight fractions of G at different temperature tests

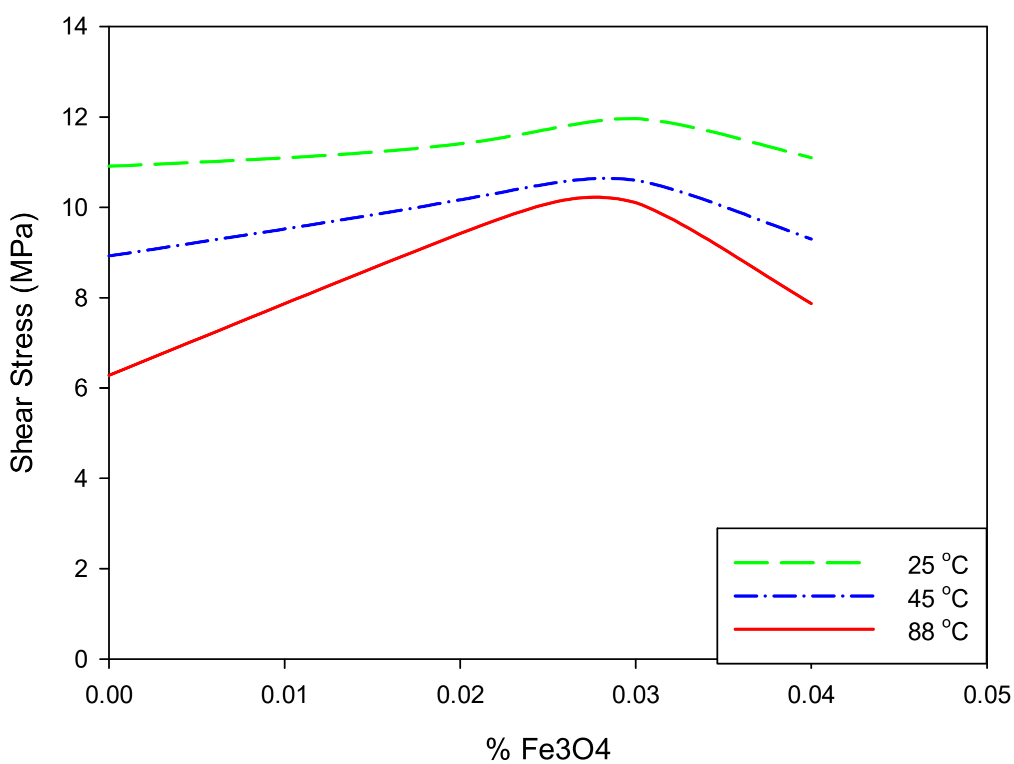

Fig. 13. Shear Strength of the Epoxy-Fe_3O_4 composite with Fe_3O_4 content

As can be seen in **Fig. 10** and **Fig. 11**, by increasing the temperature from 25 ˚C to 88 ˚C a temperature close to the adhesive glass transition temperature) the strength was considerably reduced. As the testing temperature approaches and surpasses the *Tg*, the epoxy transitions to a rubbery state. This transition is associated with increased molecular mobility and decreased material properties. The transition from a glassy to a rubbery state reduces material strength and modulus. Epoxy is characterized by a rigid, three-dimensional network of crosslinked polymer chains in its glassy state. The chains become more mobile as the *Tg* is surpassed, leading to a less organized and mechanically robust structure. **Fig. 12** represents the relationship between the shear strength and the weight ratio of graphene at different temperatures. We note that the highest shear stress (16 MPa) is at a weight ratio of 3 wt% at a temperature of 45 °C. We also note that there is an increase in load with an increase in the weight ratio of graphene up to 2 wt%, then it decreases. The strength reduction after 3 wt% is attributed to the agglomeration of the nanofillers in the epoxy resin. Fig. 13 represents the relationship between the shear strength and the weight ratio of Fe_3O_4 at different temperatures. It is worth noting that the highest shear strength (12 MPa) is at a weight ratio of 3 wt % at a temperature of 25 °C. Additionally, we observe that there is an increase in load with an increase in the weight ratio of graphene up to 2 wt%, then it decreases due to the filler agglomeration.

4.3 Morphology Results

Incorporating nanofillers, such as graphene nanoplatelets, into epoxy adhesives significantly enhances adhesive joint bond strength. These nanoscale reinforcements provide exceptional mechanical properties, increase bonding sites, and introduce intrinsic toughening mechanisms. However, maintaining proportions to prevent agglomeration was crucial for preserving optimal mechanical performance.

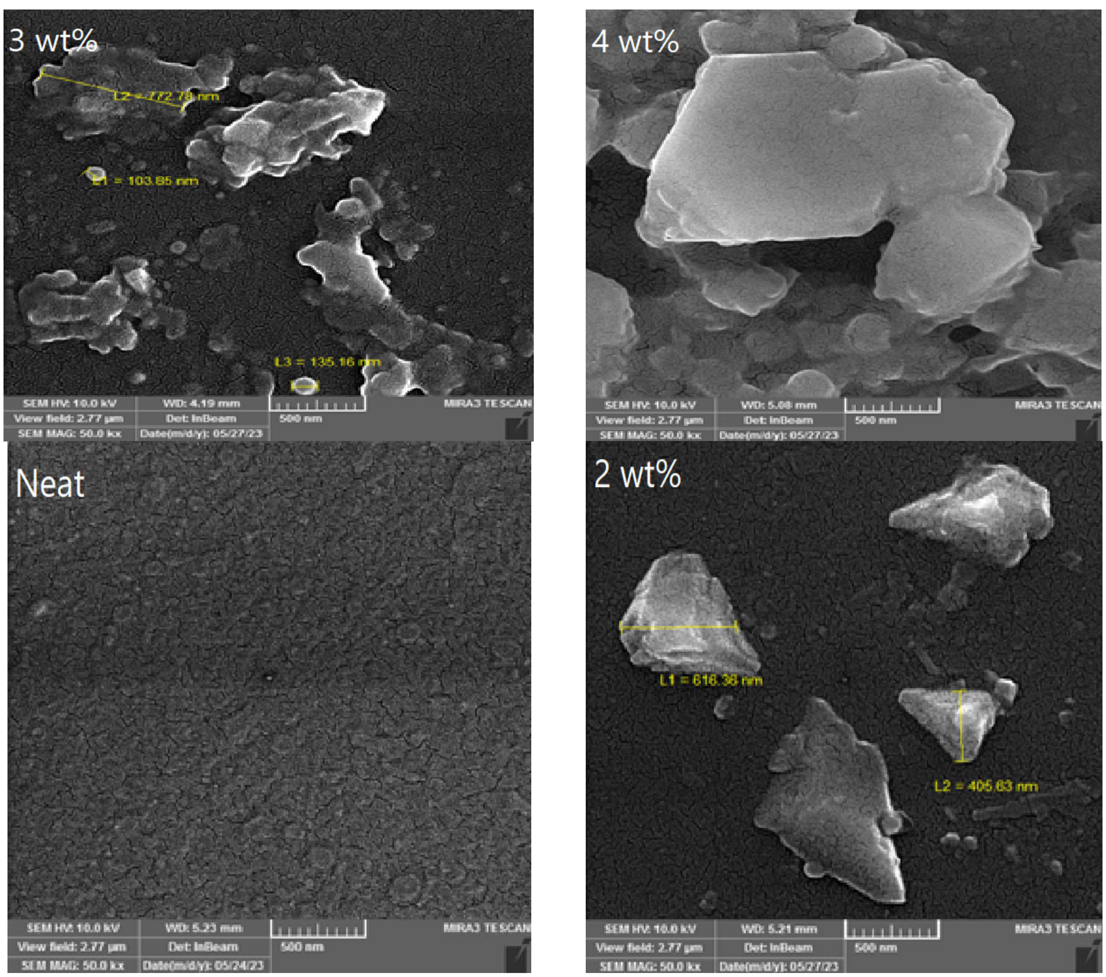

Fig. 14. The SEM micrographs of distribution of Graphene nanoplatelets nanofillers mixing with epoxy.

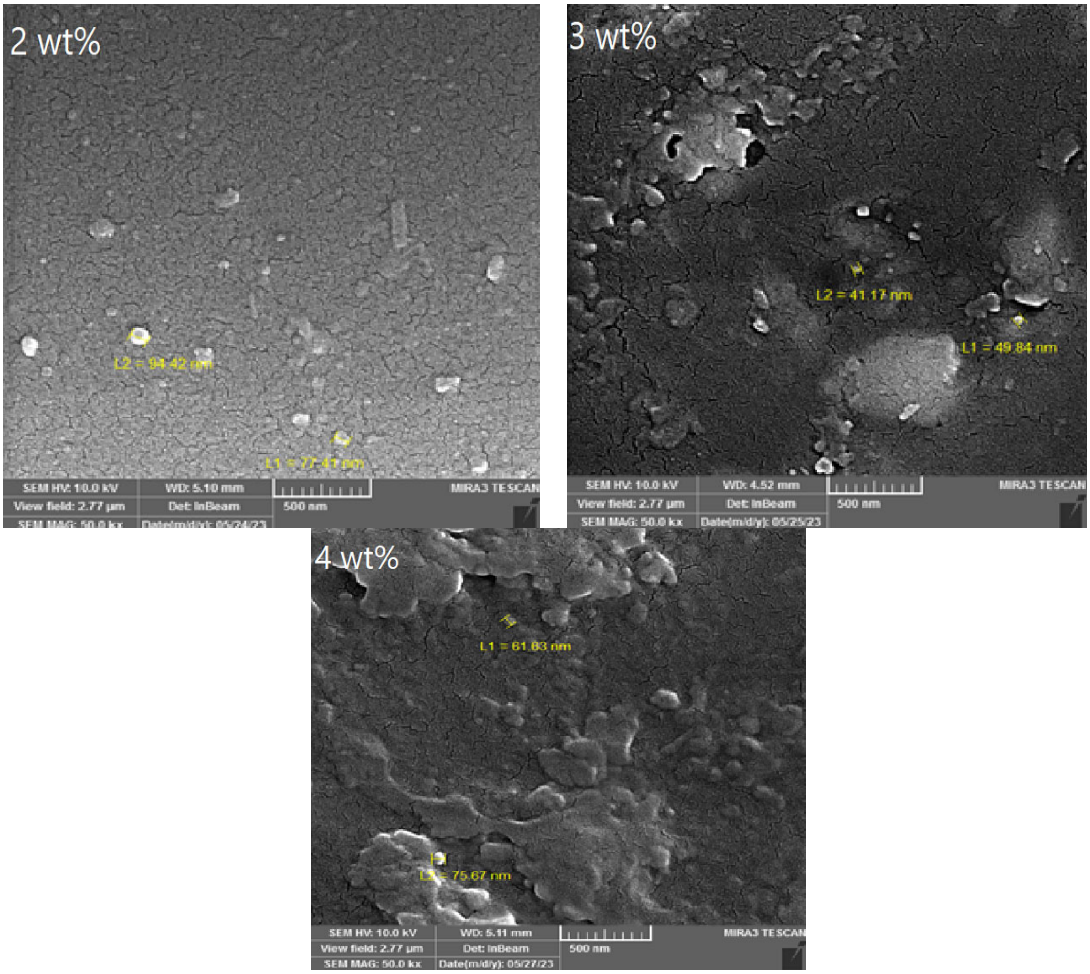

Fig. 15. The SEM micrographs of distribution of Fe_3O_4 nanofillers mixing with epoxy.

SEM images, as depicted in **Fig. 14** and **Fig. 15** show the morphological characteristics of the fracture surfaces of the pure and nanocomposite adhesive joints containing different weight fractions of graphene nanoplatelets and Fe_3O_4 nanofillers. Upon closer examination of the SEM images, it became apparent that adding graphene nanoplatelets and Fe_3O_4 nanofillers resulted in notably rougher fracture surfaces compared to the pure adhesive. This rougher surface served as an indicator of higher fracture toughness in the adhesive joint. In samples with 3 wt% of graphene nanoplatelets and Fe_3O_4 nanofillers, a homogeneous distribution within the adhesive area was observed, contributing to the enhancement of mechanical properties. However, adding 4 wt% of graphene nanoplatelets and Fe_3O_4 nanofillers led to evident agglomerates during the mixing process. This nanofillers agglomeration was the underlying reason for the degradation in shear properties observed in adhesive joints with higher nanofillers contents of 4 wt%.

5. Conclusions

This paper studied the effect of incorporating nanofillers on the mechanical properties of adhesive joints. It was found that the influence of nanofillers on the shear strength of adhesive joints varied depending on the type and weight percentage of the added nanofillers and the testing temperature. The increase in nanographene percentages of 2 wt %, 3 wt %, and 4 wt% at 88 ^{o}C led to a rise in shear strength by 112.2%, 112.6%, and 82.5%, respectively. The same trend was observed for Fe_3O_4 nanofillers, as the addition of 2 wt%, 3 wt%, and 4 wt% of Fe_3O_4 nanofillers at 88 ^{o}C caused 50%, 60.8%, and 25.3% enhancement in the adhesive joint strength, respectively. Therefore, the study demonstrated that increasing the nanofiller content from 2 wt% to 3 wt% improved the shear strength of adhesive joints. However, further increasing the nanofiller's contents to 4 wt% degraded the adhesive joint bond strength due to the nanofiller's agglomeration. Moreover, adding nano graphene fillers resulted in a general increase in joint strength greater than Fe_3O_4 nanofillers. The shear tests were conducted on the pure and nanofillers-reinforced adhesive joints at ambient, 45 °C, and 88 °C. It was found that increasing the testing temperature from ambient to higher temperatures decreased the shear strength of adhesive joints. Differential scanning calorimeter examination was conducted on the pure and nanofiller-reinforced adhesives to investigate the dispersion quality of the nanoparticles. The study demonstrated that adding a low amount of nanographene and Fe_3O_4 nanofillers to the epoxy adhesive did not considerably alter the adhesive glass transition temperature.

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