Experimental investigations on fatigue life enhancement of composite (e-glass/epoxy) single lap joint with graphene oxide modified adhesive

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Abstract

With the increased use of composites in various sectors as a lightweight material exhibits high strength to weight ratio with tailor made properties. It becomes necessary to focus on various joining methods and different types of composite joints and their strength. The present study aims to improve the mechanical strength of single lap joint of composite material comprises of E-glass fibers and nano modified adhesive. Epoxy adhesive has been modified by dispersing Graphene Oxide (GO) to investigate the possibility of enhancement in the fatigue strength and fracture resistance of the single lap joint. Modified Hummer’s method has been used for synthesis of Graphene Oxide. Experimental investigations have been carried out for comparison of tensile and fatigue strength which shows significant improvement in the number of failure cycles for 0.25 wt.% and 0.75 wt.% GO concentrations respectively as compared to neat adhesive. Tension test results showed a significant increase in the fracture toughness of the joint due to addition of GO nanoparticles. There has been 33% and 19% increase in fracture toughness in 0.25 wt.% and 0.75 wt.% GO samples respectively as compared to neat adhesive. Improvement in fracture toughness, among all other nano-reinforcements has been obtained using GO, mainly because of its better capability of deviating the crack growth path to the longer path causing the final failure to retard and consequently improving mechanical properties of the adhesive for the tensile and fatigue strength parameters.

1. Introduction

The usage of composite materials in various fields of engineering has been found at its pick. Automotive and aerospace are the major domains where these materials are used. Even though having significant benefits, the use of these materials above a certain extent finds limitations. The process of joining of these Fiber Reinforced Plastic (FRP) composites unlike conventional metals becomes a point of concern. For most of the times, the adhesive bonding of these joints is preferred. Hence there is a need to investigate and enhance the bonding strength of the adhesive joints.

Graphene is one of the most researched materials recently for its peculiar mechanical and chemical properties. It has been the most preferred reinforcing agent to form nanocomposites. Un-functionalized graphene nano platelets (GNP) used as reinforcing additives in the epoxy resin showed enhancement in the Young’s modulus than the neat epoxy adhesive. Increasing the percentage of GNP concentration gradually decreased the lap shear strength of the nanocomposite because of formation of agglomerates of GNP [1]. It was observed that use of 0.25 wt.% of GNP in epoxy structural adhesive of double cantilever beam (DCB) specimen increased the fracture toughness resistance by 5 folds compared to neat adhesive [2]. Functionalized graphene oxide was used to prepare GO/epoxy nanocomposite to check for the Young’s modulus, hardness and tensile strength. It was observed that these parameters improved as compared to pure epoxy at graphene oxide’s
concentration of 1.5 vol.% [3]. Some investigations were carried out to check the strength parameters of the single lap joint and found that average shear strength and elongation to failure found to be increased by 28% and 36% respectively by addition of silica nanoparticles and multiwalled carbon nanotubes [4–6]. Allen et al [7] discussed two types of synthesis of graphene viz. mechanical exfoliation method and chemically derived graphene. Chemically obtaining graphene taking into consideration its applicability in various fields, the chemical methods proved to be better than exfoliation techniques. Camanho et al [8] suggested a damage model to simulate the spread of high cycle fatigue loading failure of delamination. The experimentally determined coefficients and the damage state are obtained depending on the conditions of loading for the establishment of Paris law for rate of crack propagation. Nikhil Koratkar et al [9] studied and reviewed the properties of graphene and functionalized graphene sheets (FGS) to be dispersed with different polymer resins forming graphene nanocomposite. It was discovered that the nanocomposite of 0.125 wt. % of FGS improved the toughness of the fracture by about 65% and the energy of the fracture by about 115%. Fuqiang Wu and WeiXing Yao [10] provided a fatigue damage model containing two material parameters based on the rule of composite stiffness degradation. Priyank et al [11] presented a simulation based approach. They used atomistic based continuum (ABC) multiscale modeling technique and found around 30% enhancement in the strength of Single lap joint using with 2% concentration CNT. Panta et al [12] characterised a high-performance epoxy-based adhesive with graphene nanoplatelets (GNPs) and phase-separated triblock copolymers (BCPs) to enhance the lap shear strength of the adhesive joints of aluminium sheets. Adding 1.0 wt% OZ-GNPs into 10 wt% SBM modified epoxy adhesive resulted in the lap shear strength increase by 129%, compared to that of the unmodified epoxy. In their research they found that the BCPs formed different nanostructures in the epoxy matrix and the resulting nanostructures activated different toughening mechanisms that were responsible for the improvement in the lap shear strength, while the OZ-GNPs increased the joint strength by stiffening the matrix.

Panta et al [13] reviewed different type of nano fillers such as carbon nanotubes (CNTs), graphene nanoplatelets (GNPs), nanoclay, nano-silica (nano-SiO2) and nano-alumina (nano-Al2O3). From the literature it found that adhesion strength can be improved upto 70% as per the type of nano filler used in the epoxy. Further it is found that spherical nanoparticles are less effective than nanoparticles with plate like morphology.

Wei et al [14] focused on the methods of processing of nanocomposites and their mechanical, electrical and thermal properties. Improper mixing results in inhomogeneous dispersion and decrease of the nanocomposite’s mechanical and thermal characteristics. And proper mixing leads to improved multi-functional properties of graphene. Fernandez et al [15], performed experimental analysis of composite laminate of carbon/epoxy to check for fatigue behaviour at mode I loading conditions. The goal was to obtain the energy release rate by using different data reduction methods. Banea et al [16] conducted a survey to investigate composite structures of adhesively bonded joints of fibre-reinforced plastics (FRP). The impacts of surface preparing, bonding configuration, adhesion characteristics and environmental variables on bonding conduct are briefly outlined with respect to the adhesive-bonded FRP composite structure. Rajaganapathy [17] et al used graphene based Aluminium Matrix Composites reinforced with Titanium (Ti) particles and investigated the tribological properties using Pin on Disc set up. They have reported improvement in tribological properties using Graphene with Titanium reinforcement upto 3% along with 1% of Silver nano particles. They also found beyond 3% reinforcement of Graphene and Titanium results in decrease in both strengths. Krishna Gouda et al [18] studied thermomechanical behavior of micro bamboo filler/epoxy hybrid composite with reinforcement of graphene nanoplatelet (GNPs) of different percentages. They have found increase in thermal conductivity with increased weight percentage of GNP from 0.1% to 1% with marginal rise in corrosion rate. Arukali et al [19] worked on synthesis of magnetic iron oxide/graphene oxide (Fe3O4/GO) nanocomposites and studied their tribological properties under magnetic field. The friction and wear performance has been improved upto 35.5% by increasing the GO concentration in nanocomposite under magnetic field. Other than engineering applications of GO, Valentina et al [20] used it in bone 3D printing to obtain patient-tailored scaffolds. They have found that these materials can be dispersed in the ceramics and polymers to characterize biocomposites which can be used for medical applications. Suneev Anil Bansal [21] used wet chemical oxidation process for the reinforcement of GO in the epoxy matrix. They have reported considerable improvement in the visco-elastic properties of the mixture. Further they reported around 10% improvement in the hardness of E-GO mixture at 0.5 wt% of GO reinforcement.

Many researchers worked on the dispersion of nano composites such as Carbon Nano Tubes (CNT), Carbon Nano Fibers (CNF) and Graphene Nano Platelets (GNP) etc. But very few have worked on the Graphene Oxide (GO) for improving the fatigue life of composite adhesive lap joints. There is need to further explore the dispersion of nanomaterials into adhesive and its effect on the fatigue strength of the composite lap joint. This concept has been proposed, developed, analysed, and tested in the current research work by investigating 0.25% and 0.75% concentration of GO in the epoxy resin. The tensile strength is improved by 33% and 19% respectively. Whereas the fatigue strength enhanced very significantly by 400% and 600% respectively.
2. Materials and methods

The selection of the materials to fabricate the single lap joint can be divided into three parts viz. selection of composite material for the adherend, selection of the adhesive to be used to join the adherend parts and the selection of nanomaterial to be dispersed into the adhesive.

2.1. Fabrication of composite laminate plate

E-Glass fibers HinFabTM HGU500 make with 500 gsm and Epoxy matrix were selected as composite material. The orientation of the fibers in the laminate was chosen to be $[0/45/-45/90]$. This symmetric configuration was selected as it provided marginal strength in all directions in the laminate to increase. The laminate was fabricated by using hand lay-up process and the plates of dimension of 100 mm $\times$ 25 mm $\times$ 3 mm were used for the purpose of single lap joint preparation. After the matrix became tacky, it was press cured in oven at 70 $^\circ$C for 3 h and then was allowed to cool at normal room temperature.

2.2. Adhesive

Epoxy EPOFINE 740–1 make was used as matrix, and also as the adhesive for preparing single lap joint. Hardener was mixed with epoxy resin in the ratio 4:25 by volume. Variations in the amount of hardener found to affect the curing time and consequently affected its strength.
2.3. Nanomaterial
Graphene Oxide (GO) was used as a reinforcing agent in the epoxy adhesive. It consists of functional groups of oxygen. The synthesis of GO was done using Modified Hummer’s method. Dispersion of nanoparticles is one of the most important parameters in enhancing and improving the mechanical properties of the nanocomposite. This is because the load applied can be effectively transferred through the epoxy/GO interfacial interactions. Proper dispersion of GO particles ensures the availability of maximum area of surface of filler, which affects the adjacent chains of polymer and consequently the properties of whole nanocomposite. Samples of Epoxy/GO nanocomposites as shown in figure 1 were prepared for the purpose of experimental investigation. 0.25 wt.% and 0.75 wt.% of GO in epoxy resin were prepared. Initially, GO was dispersed in acetone at a concentration of 2 mg ml$^{-1}$ of acetone. The mixture of acetone and GO was kept in ultrasonic bath sonicator for 1.5 h for GO particles to get properly mixed with acetone. This mixture was then poured into the measured quantity of epoxy contained in the beaker as shown in figure 1(a).

This complete mixture was then kept on magnetic stirrer at 1500 rpm for 3 h. Initially, the highly viscous epoxy resin loses its viscosity because of acetone. Evaporation of acetone allowed the epoxy to regain its viscosity. This process of properly mixing of acetone with epoxy also helped the GO particles present in the acetone to homogeneously disperse in epoxy resin. Mixture was kept on the magnetic stirrer till the acetone was completely evaporated. The GO/Epoxy nanocomposite was found with its viscosity regained.

2.4. Optical microscopy of GO/Epoxy nanocomposite
After the preparation of GO/Epoxy nanocomposite adhesive, the dispersion of GO in epoxy resin was checked by obtaining the optical images of both 0.25 wt.% GO and 0.75 wt.% GO samples as shown in figure 1(c). Optical images were obtained using Carl Zeiss optical microscope. The particles were found to be well dispersed with no agglomeration. The uniform dispersion of the GO particles in the adhesive is clearly seen in the optical microscopic images for both concentrations as shown in figure 1(c). Variation in the density of GO particles for different concentrations has been clearly seen in the optical image.

3. Preparation of single lap joint specimen (SLJ)
The geometrical dimensions were made in accordance with ASTM D5868 standards for testing of materials. The geometry of the single lap joint specimen is as shown in the figure 2(a). The finished single lap joint specimens are shown in figure 2(b). A single adherend is composed of 8 lamina layers of glass fibers in [0/45/−45/90], orientation.

Prior to the preparation of the joint, the adherend was surface treated by using tool bit so as to increase the surface roughness of the area of overlap. This pre-treatment of the surface shows enhancement in the mechanical performance of the joint, improved the durability of the joint and improved the adhesion phenomenon. After surface preparation, epoxy adhesive was applied on surface. The specimen was held in
C-Clamp to ensure proper bonding. The joint was kept at room temperature till the epoxy became tacky, later it was kept in oven at 120°C for 30 min. The same procedure was adopted to prepare all the three types of joints namely (neat adhesive, 0.25 wt.% GO and 0.75 wt.% GO).

4. Experimental investigations

4.1. Tensile testing
ASTM D5868 standard were followed for the tension test of these FRP single lap joint specimens. Load versus displacement (cross head travel-CHT) was obtained for the comparison of effect of Epoxy/GO nanocomposite on the variations of fracture toughness. The testing has been done on universal Testing Machine (UTM) of 100 kN capacity for the better accuracy. The specimen with end tabs as shown in fig. has been axially loaded as per guidelines given in the standard with a cross head travel rate of 3 mm min⁻¹ for all the specimens.

There is no significant variation in the magnitude of the load at the failure of the joint as seen from figure 3(c). But there is a variation in the behavior of the joint towards fracture. As seen from figure 3(c), the effective area covered by the curve for 0.25 wt.% GO specimens seems the highest. Also, the effective area of the curve for 0.75 wt.% GO specimens is higher than that of neat adhesive specimens. The fracture toughness was found to be increased by 33% and 19% respectively for 0.25 wt.% GO and 0.75 wt.% GO.

4.2. Fatigue testing
Fatigue testing has been performed in accordance with ASTM D 5868 standard. All the three specimens were tested for 90% of their ultimate tensile load. The R ratio was kept 0.1 for all the tests. Frequency of 3 Hz was set and number of cycles to failure was observed.

Debonding was observed from the edges where stress concentration was high. Crack was initiated and propagated gradually with increase in cycle. Debonding can be observed by naked eyes as shown in figure 4 with
reduced lap area by yellow colored arrows. The gap keeps on decreasing as number of cycles and delamination increases. It was noted that debonding began from ends of the overlapped bonded region and propagated to the midpoint of the joint. The reason behind that was peel stress.

The fatigue tests were carried out keeping the parameters viz. R ratio and frequency same. Five specimens of each concentrations were tested to obtain the best average of the results. Increase in % of GO in the epoxy resin, Figure 4. Fatigue damage when loaded at 90% of ultimate strength (R = 0.1) (a) 500 cycles (b) 1500 cycles (c) 2500 cycles (d) 3500 cycles (e) 5000 cycles.

Table 1. Fatigue Test Results.

| Sr no. | Specimen type          | Sample no. | 90% Load (kN) | No. of cycles to failure | Average No. of cycles to failure |
|--------|------------------------|------------|---------------|--------------------------|---------------------------------|
| 1      | Neat Adhesive 0% GO/Epoxy | 1          | 3.281         | 4926                     | 3171                            |
|        |                        | 2          | 3.281         | 4963                     |                                 |
|        |                        | 3          | 3.281         | 2570                     |                                 |
|        |                        | 4          | 3.281         | 1348                     |                                 |
|        |                        | 5          | 3.281         | 2049                     |                                 |
| 2      | 0.25% wt. GO/Epoxy     | 1          | 3.244         | 11563                    | 12745                           |
|        |                        | 2          | 3.244         | 12095                    |                                 |
|        |                        | 3          | 3.244         | 12956                    |                                 |
|        |                        | 4          | 3.244         | 13934                    |                                 |
|        |                        | 5          | 3.244         | 13176                    |                                 |
| 3      | 0.75% wt. GO/Epoxy     | 1          | 3.046         | 19752                    | 19904                           |
|        |                        | 2          | 3.046         | 20970                    |                                 |
|        |                        | 3          | 3.046         | 18549                    |                                 |
|        |                        | 4          | 3.046         | 19577                    |                                 |
|        |                        | 5          | 3.046         | 20672                    |                                 |
increases the average number of cycles to failure as indicated in figure 3(d) and tabulated in the table 1. The graphs shows variation of number of cycles to failure with respect to number of specimens tested.

5. Failure modes of tested specimens

Scanning Electron Microscopy (SEM) was performed on the fatigue tested specimens to investigate the failure modes of the debonded specimens. The area of overlap was the region of interest. Mode of failure can be understood only by analysing the surface of failed specimen. The surface of the tested specimen being composite laminate is non-conducting. So, prior to SEM, the samples were gold coated to make the surface conducting.

As per ASTM D5573-99, there are seven modes of failure of fiber reinforced plastics bonded joints. Figure 5(a) shows the SEM images of the 0.25 wt.% GO specimen tested for fatigue failure. Figure (a) and (b) represents the adhesive failure of the single lap joint whereas Figure (c) shows fiber failure and (d) shows the mixed mode failure as there was significant breakage of the fibers, Similarly, figure 5(b) represents the SEM images of 0.75 wt.% GO specimens tested for fatigue failure. It can be carefully observed that, in Figure (a) and (b) the layer of epoxy adhesive above the fibers has been distorted, so it is adhesive failure and that in Figure (c) and (d) it clearly shows the breakage of fibers.

Figure 5. SEM Characterization of debonded specimen ((a) 0.25 wt.% GO specimen, (b) 0.75 wt.% GO specimen).
6. Conclusion

The tensile testing of the specimens showed significant variation in the behaviour of the adhesive material due to the inclusion of Graphene Oxide nanoparticles. Fracture toughness was found to be increased by 33% in 0.25 wt. % GO and 19% in 0.75 wt.% GO in comparison to neat adhesive or 0 wt.% GO. It can also be concluded that there was prominent increase in the fatigue strength of the composite joint. As compared to neat adhesive, the cycles in 0.25 wt.% GO specimens were found to be increased by 9574 cycles and that cycles in 0.75 wt.% GO specimens increased by 16733 cycles. Increase in the number of cycles to failure with increase in the percentage of the graphene oxide nanoparticles indicated the major outcome that graphene oxide increases the bonding strength of the joint in fluctuating load conditions thereby increasing the fracture toughness resistance.

Dispersion of GO nanoparticles in the epoxy adhesive indicated interestingly the deviation of the crack growth path to a notified longer path due to its unique ability to fill in micro cracks which may form within the material under loading conditions. This results in the final failure to retard and slow down and ultimately improved the mechanical characteristics. The modified adhesive have indicated sufficient bonding strength so as to adhere to the surface and resulted in mixed failure according to ASTM D5573–99 standards. Overall GO/Epoxy nanocomposite has significantly and successfully enhanced the mechanical characteristics of the composite single lap joint.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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