Evaluation of mode III Delamination behaviour of modified Carbon/Glass Fibre Reinforced Polymer Composites with Nanoclay particles

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Abstract. Incorporating nanofillers in epoxy resin is a novel approach to improve the mechanical properties of polymer composites. Recent studies disclose that the inclusion of nanofillers such as SiC, CNT, alumina and nanoclay into epoxies at micro and nanoscale levels enhances the mechanical properties of epoxies. In this research, the improved mechanical properties of nanoclay modified carbon/glass fibre-reinforced polymer nanocomposite (FRPNC) were investigated. The neat DGEBA epoxy resin was modified with nanoclay at different wt % (0.5-2wt%) by ultrasonication process for achieving better dispersion of nanofillers. The modified polymer laminates were fabricated with unidirectional carbon/glass fibres with the stacking sequences of (0°G/0°G/0°C)S using hand lay-up process. The mechanical properties such as mode III delamination toughness, tensile strength and flexural strength were investigated using servo controlled hydraulic universal testing machine and 3-point bending test setup respectively. The highly cross linked structure between epoxy and nanoclay particles improves the mode III fracture toughness, tensile and flexural properties. The damage mechanisms of fractured specimens are characterised by SEM images.

Key words: Glass fibre, Carbon fibre, Nanoclay, Delamination, Tensile strength, Flexural strength.

1. Introduction

The carbon fibres have outstanding mechanical properties however it has low compressive/tensile ratio and lower strain-to-failure values. Conversely, the glass fibres have low tensile strength and higher strain-to-failure values. To retain the advantageous of both carbon and glass fibres, hybrid polymer laminates were developed. In the fabrication of polymer composites, thermoset epoxy resin was mostly used as matrix thanks to its outstanding mechanical properties, low shrinkage effect, good chemical resistance and higher adhesion properties. Due to these unique characteristics, the epoxy resin was widely used to fabricate composite structures as a secondary load carrying members. DGEBA epoxy resins are typically brittle material and it offers poor resistance against fracture loading. To extend the applications of polymer composites in structural field it is necessary to improve the fracture toughness of epoxy resins. Dispersion of nanofillers with DGEBA epoxy resin is a new technique to improve the electrical, mechanical and tribological characteristics of polymer composites [1-4]. The mechanical
properties of polymer nanocomposites are influenced by several factors such as type of fibres reinforced, type of filler materials incorporated, length of fibre and its orientation with respect to the stress direction [5-6]. In addition, the properties of nanocomposites could be further enhanced by increase in weight fraction nanofillers, length/diameter ratio, and arrangement of nanoparticles, uniform dispersion of nanoparticles [7-10], and size of interface zone [11]. Mechanical characterization of the fibre reinforced polymer matrix composites was greatly affected by length to diameter ratio of the fibres, higher adhesion strength between fibres and matrix material. Graphene based fillers were reinforced in thermoset and thermoplastics matrices which improves its rheological, electrical, mechanical and thermal properties [12]. Inclusion of elastomers in epoxy resin improves the fracture toughness of polymer laminates. However, this inclusion decreases the tensile strength of laminates due to lower strain values. To compensate this effect the surface modified SiO$_2$ particles in addition with elastomers were incorporated into epoxies improves the fracture toughness as well as tensile strength of laminates [13-14]. Titanium oxide (TiO$_2$) is one of the promising material has been attracted by many researchers thanks to its highly desirable mechanical properties. Incorporation of small fraction of TiO$_2$ nanoparticles into vinyl ester resin significantly improves the toughness of the polymer matrix composites [15]. The tensile properties of PU/Nanoclay nanocomposites were improved with loading of nanoclay particles up to 3 wt.%. At the same time, when the nanoclay loading is above 3 wt. %, the mechanical properties of the nanocomposites were decreased considerably [16]. Inorganic nanoparticles like nanoclay possess outstanding mechanical properties; inclusion of nanoclay in organic polymers shows greater improvements in mode-I fracture toughness values [17]. In another research, the cyclic load behaviour of epoxy based polymer matrix composites with reinforcement of glass fibres were improved by adding both multi-walled CNT/nanoclay particles into epoxy resins [18].

The above literature review effectively discloses that, to efficiently transfer the applied load to the fibre reinforced, the matrix material should be tougher. The toughening mechanism of polymer laminates are critically influenced by many factors and greatly influenced by the behavior of matrix material. The dispersion of nanofillers into epoxy resins makes matrix material tougher due to better adhesive strength between nanofillers and epoxy matrix. This interaction increased the load transfer capacity of matrix results improved fracture toughness, tensile and compressive strength of modified epoxy based polymer composites. Numerous experimental works has been reported in investigating the effect of incorporation of various nanoparticles in epoxy based polymer composites. However, effect of addition of nanoclay on delamination characteristics of carbon/glass fibre reinforced epoxy based composites (FRPC) was not investigated to date. In this work, pure mode III interlaminar fracture behaviour of nanoclay dispersed epoxy based polymer composite is investigated. To analyse the influence of nanoclay on fracture and other mechanical properties, epoxy modified with different concentrations of nanoclay (0.5wt%, 1wt%, 1.5wt%, 2wt %). The laminates were fabricated with alternate sequence of unidirectional carbon and glass fibers.

2. Materials and specimen preparations

2.1. Materials

Unidirectional e-glass fibre strands (ASTMD578-00) with tensile strength of 40GPa, and unidirectional carbon fibres with tensile strength of 70GPa was used as fibre materials. Highly cross linked thermoset epoxy resin used as a matrix material. Surface modified nanoclay (Cloisite 15A) used as nanofiller material. Details of chemical structure of constituent’s materials such as epoxy resin, curing agent and other incorporated nanoparticles were illustrated in Table.1

2.2. Modified epoxy

To investigate the influence of nanofillers, thermoset epoxy resin is modified by different weight percentage of nanoclay i.e. 0.5 wt.%, 1wt% , 1.5wt% and 2wt.% and pristine epoxy. Further addition of nanoparticles (i.e. above 2 wt %) leads to the agglomeration of nanoparticles. To attain improved mechanical properties, uniform distributions of nanoclay particles are required within epoxy material. To achieve uniform dispersion, nanoclay particles were mixed into epoxy resin using mechanical
stirrer and sonication process. The formation of intercalated structure between matrix and nanoparticles interface region (figure 1) provides the modulus and strength to the modified composites. At the interface, nanoparticles create near-molecular blend with matrix which enhances strength, thermal and tribological properties.

### Table 1. Material chemical structure

| Name                              | Chemical structure               |
|-----------------------------------|----------------------------------|
| DGEBA                             | ![Diethylenglycol Bisphenol A](https://example.com/image1.png) |
| Triethylenetetramine (TETA)       | ![Triethylenetetramine](https://example.com/image2.png) |
| Surface modified Nanoclay(cloisite 15A) | ![Surface modified Nanoclay](https://example.com/image3.png) |

**Figure 1.** Molecular structure of Nanoclay-epoxy matrix

2.3. **Preparations of laminates**

Five set of symmetrical laminates with (0°G/0°G/0°C)₅ orientation of carbon/glass fibre were prepared using hand layup techniques (Fig.2). Teflon film is introduced at the mid plane of the laminates for create an initial crack in ECT specimen. Laminates were fabricated with different weight percentages of nanoclay and categorized as CGFRP1 (0 wt. %), CGFRP2 (0.5 wt. %), CGFRP3 (1 wt. %), CGFRP4 (1.5 wt. %), CGFRP5 (2 wt. %) based on the inclusion of nanoclay content. Each laminates having six ply with thickness of 4-5mm.

**Figure 2.** Symmetric Laminates
3. Mechanical characterization

3.1. Mode III Delamination fracture test

The mode III interlaminar fracture toughness of modified polymer laminates were calculated based on the experimental values of Edge Crack Torsion (ECT) test [19-24]. Testing specimens were characterized by typical ECT configuration setup as shown in Fig. 3.

![Figure 3. ECT Specimen configuration](image)

In this test configuration, the ECT samples with an initial crack length of 15 mm were tested. All the samples were tested under torsional loading conditions by providing three loading support computer-controlled Instron 5567 testing machine. The out of plane loading condition produces a torque that twists the laminates. Crack-growth characteristic of the specimens were measured by a parameter strain energy release rate ($G_{IIIC}$). The fracture toughness values are estimated by the compliance values using laminated plate theory with the equations (1) and (2).

\[
C = \frac{\delta}{p} = \frac{W^2L}{4B(1-2S)a(D66)I} \tag{1}
\]

\[
G_{IIIC} = \frac{P_c^2C(1-2S)}{2LB(1-(1-2S)\frac{a}{B})} \tag{2}
\]

Where, $W$ - applied load, $L$ - selective length, $B$ - sample specimen breadth, $a$ - crack length dimension, $P_c$ – maximum or critical load, $(D66)_i$ and $(D66)_u$ – torsional stiffness values of uncracked and half cracked specimens.

In carbon/glass fibre reinforced modified epoxy based laminates, reinforced fibre carries an applied load and nanoclay added epoxy matrix was effectively transfers the load to the fibres. The experimental testing results are summarised in figure 4. It is observed that mode III delamination fracture toughness value of CGFRP laminates were mostly influenced by matrix material and this could be improved by addition of nanofillers in the epoxy matrix. The interlaminar fracture toughness of CGFRP1-CGFRP5 laminates is in the range of 112 J/m$^2$ to 144 J/m$^2$. The maximum fracture toughness value of 144 J/m$^2$ is observed for CGFRP4 specimen at 1.5 wt. % addition of nanoparticles. Incorporation of nanoparticles in epoxy resin up to 1.5 wt. % increases the fracture energy, but further addition of nanoparticles decreases its fracture strength due to low interfacial adhesion strength. Inclusion of nanoclay particles in matrix material enhances the fracture toughness of modified laminates thanks to matrix/particles adhesion, interlocking and plasticizing effect. The other influential parameters are stacking sequences and orientation of the fibres. The experimental results show that, stacking of carbon fibres in the middle layer enhances the interlaminar strength [25-26]. In addition, the nanoparticles inclusion in epoxy matrix restricts the intercalated chains to break which increases
the resistance against fracture loading. Further addition of nanoparticles i.e. 2.0 wt% in epoxy decreases the fracture toughness of composites to 133 kJ/m² due to weaker interaction between matrix and nanoparticles.

3.2. Ultimate Tensile Strength
The tensile strength values are obtained from the UTM at a load cell capacity of 100 kN. The displacement values are accurately measured by an extensometer. At least five specimens were tested for each laminate and also for pristine epoxy specimen. Test is repeated thrice and the average of all the values are summarised in Table 2. The effects of addition of nanoclay on tensile load of carbon and glass fibre reinforced laminates were shown in Figure 5.

The tensile stress of CGFRP specimens are increasing linearly from 221 MPa to 260 MPa. The maximum tensile stress of 260 MPa was observed for CGFRP4 specimen at 1.5wt% which is 17% higher than tensile stress (249 MPa) of pristine epoxy based specimen. This results show that the additions of nanoclay makes strengthen the polymer composites and modified matrix accepts large load and safely transfer to the reinforced fibers which increases the load withstanding capacity, increases the percentage of elongation at break results higher tensile strength [27].
### Table 2. Tensile values of tested specimen

| Specimen label | Maximum Load (kN) | Elongation at break (%) | Tensile strength (MPa) |
|----------------|-------------------|-------------------------|-----------------------|
| CGFRP1         | 7.18              | 8.00                    | 221                   |
| CGFRP2         | 7.94              | 8.20                    | 252                   |
| CGFRP3         | 7.96              | 8.40                    | 256                   |
| CGFRP4         | 8.34              | 8.43                    | 260                   |
| CGFRP5         | 7.54              | 7.49                    | 249                   |

### 3.3. Flexural strength

To understand the flexural behavior 3-point flexural test (Figure 6) is conducted at room temperature. Six specimens were prepared and tested for each composite laminates, in accordance with ASTM: D790-02 standard [28]. Figure 6 shows the flexural strength values of nanoclay dispersed epoxy based carbon/glass fibre reinforced laminates with different wt.% inclusion of nanoparticles. The tests revealed that, the flexural strength of laminates for CGFRP1, CGFRP2, CGFRP3, CGFRP4 and CGFRP5 were 354 MPa, 421 MPa, 444 MPa, 464 MPa and 468 MPa respectively. The flexural strength value increases with increased wt.% of addition of nanoclay. The flexural strength got enhanced by 19% (0.5 wt.%), 25% (1 wt.%), 31% (1.5 wt.%), and 32% (2 wt.%) as compared with pristine epoxy based composites. This improved flexural strength behavior discloses that higher surface energy of nanoclay and exfoliated nanoclay bonding with matrix materials increased the bending resistance of the laminates.

![Figure 6. Flexural strength results](image_url)

### 3.4. Comparison of mechanical properties

In this work, nanoclay modified carbon/glass fibre reinforced polymer laminates were prepared with the stacking sequence of (0°G/0°G/0°C)S with different wt.% (0.5 – 2 wt.%) of nanoclay particles. The fracture toughness, tensile and flexural stress values were increased as compared with pristine epoxy specimens due to presence of better adhesive strength of nanoclay with matrix material. The comparison of mechanical properties of modified laminates are summarised in Table 3. The strain energy release rate $G_{inc}$, tensile stress, and flexural stress values are plotted in Figure 7. Incorporation of nanoclay with epoxy resin improves the delamination characteristics under mode III loading.
conditions up to 1.5 wt.%. Further addition of nanoclay (2 wt.%) decreases the mechanical properties due to decreasing chain length of modified epoxy matrix. However, these properties are higher than neat epoxy based composite properties. The flexural modulus values reaches to maximum of 468 MPa at 2wt%. The existences of van der walls bond between the chains are responsible for the improved mechanical properties.

Table 3. Comparison of properties

| Specimen   | G_{	ext{IC}} (J/m²) | Tensile stress (MPa) | Flexural stress (MPa) |
|------------|----------------------|----------------------|-----------------------|
| CGFRP1     | 112                  | 221                  | 354                   |
| CGFRP 2    | 121                  | 252                  | 421                   |
| CGFRP 3    | 125                  | 256                  | 444                   |
| CGFRP 4    | 144                  | 260                  | 464                   |
| CGFRP 5    | 133                  | 249                  | 468                   |

Figure 7. Mechanical properties of modified laminates

4. Morphology
The morphology study of the tested specimen was analysed by the Scanning Electron Microscopic (SEM) images. This SEM images shown in Figure 8 discloses that toughening mechanism of modified laminates. Matrix cracking, fibre cracking, fibre pull-out and particle debonding mechanisms were identified through the SEM images. The static adhesion strength at ply interfaces increases the interlaminar fracture toughness, tensile stiffness and flexural strength of modified nanocomposites. The interaction between nanoclay and matrix is poor; the particles were unable to carry the maximum load as a result the strength of composites will be decreased
8. SEM images of modified laminates

5. Results and discussions
In view of improving the fracture toughness of carbon/glass fibres reinforced polymer composites, small wt.% of nanoclay was incorporated into epoxy resin by sonication process. The carbon fibres were reinforced in the mid plane and the glass fibres are reinforced as the exterior surface of the laminates. Five sets of laminate with (0°G/0°G/0°C)_5 orientation and stacking sequences were prepared using hand lay-up technique. All the laminates were characterised by the experimental testing in accordance with ASTM standard. Superior mechanical properties of nanoclay enhance the properties of laminates due to the high specific surface area and greater matrix/particles adhesion and interlocking. The mode III interlaminar fracture behaviour of carbon/glass fibres reinforced polymer composites increases with increasing the wt. % addition of nanofillers. The maximum G_{IIIc} of 144 J/m^2 was observed at 1.5 wt.% of nanoclay content. The higher interaction between the fillers and matrix improves the fracture toughness of polymer composites. With increase in wt % of nanofillers, the tensile strength of laminates reaches to maximum of 260 MPa at 1.5 wt.% of nanoclay addition and further addition of nanoparticles the property value is decreased. The flexural strength of modified laminates increases with the addition of
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