Interlaminar fracture toughness in Glass – Cellulose Reinforced Epoxy hybrid composites

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Abstract: Laminates of fibre reinforced composites are weak in through thickness but strong in fibre direction, this lead to development of hybridization concept in polymer composites. In this work a new method of disperssing cellulose micro particles on uni-directional (UD) Glass fibre epoxy composite using semi-automated draw down coating technique was adopted to enhance fracture toughness. Test results show that by adding cellulose increases the load carrying competency by 32% in mode-I as compare to Glass-Epoxy composite samples. Improvement in interlaminar critical energy release rates (\(G_{IC}\) and \(G_{IIc}\)) up to 55% in Mode –I and 19 % in Mode –II respectively was also observed. This enhancement in fracture toughness is due to the amount of fiber bridging seen during crack initiation and propagation.

Keywords: Cellulose, Glass Epoxy, Critical energy, Fracture toughness and Fiber bridging

1. Introduction

In recent years, hybrid polymer composites are largely used in aerospace, automotive and civil engineering applications. Hybrid composites are the materials that are formed by combining different fibers and matrices together to form a single structured composite. In general these composites are exposed to delamination significantly. Various methods to improve their resistance to delamination exists which adds substantial cost and intricacy to the production procedures of the composite materials. Therefore techniques to increase the fracture toughness of these high performance composite materials are crucial in order to use these materials in practical applications.
The addition of epoxy Terminated Butadiene Nitrilerubber (ETBN) liquid rubber in the form of solution on to glass epoxy pre-pregs brought about substantial improvements in fracture toughness under mode I and mode II loading by adopting an optimum coating density of 27.77 g/m². Also the addition of ETBN liquid on to carbon epoxy pre-pregs increased the fracture toughness under mode I and mode II loadings with a coating density of 22.66 g/m²[1-2]. Combination of multi walled carbon nano tubes (MWCNT’s) and spherical silica or rubber particles gives high performance epoxy composites showing balanced stiffness, strength and fracture toughness along with superior electrical conductivity[3]. The interleaved E-glass layer contributes only a small amount to the energy absorption under mode I fracture [4-5]. Z-pinning is another method to improve the fracture toughness under mode I loading by varying the z-pin content [6]. The effect of MWCNT’s in carbon epoxy composites shown increased mode II fracture toughness by 30% and reduced size of internal delamination developed due to impact damage [7]. By incorporating CTBN rubber modifier in Glass reinforced hot cured epoxy composites was observed increase in the fracture toughness [8]. The tapered geometry steel fibers in epoxy matrices showed that pull out of tapered fiber dissipates up to 27 times more energy than straight fibers[9]. Increasing the cotton fibers up to 0.5 wt % showed increases in the flexural strength, flexural modulus and fracture toughness of the composites [10]. Twist flax yarn structure and the rough surface of flax fibers led to remarkable fiber bridging between flax, flax yarn and glass fibers [11] resulted in improved interlaminar properties of hybrid fiber reinforced polymer composites containing flax/glass fibers.

In this work investigated the effect of cellulose micro particles in UD Glass fiber reinforced epoxy composite on mode I and mode II fracture toughness. Dispersion of cellulose on UD Glass fiber by the aid of semi-automatic draw down coating technique followed by hand layup method of moulding composite was adopted. The fracture toughness for glass- epoxy and cellulose coated samples are evaluated.

2. EXPERIMENTAL

2.1. Materials and Methods

UD Glass fiber was used in this work supplied by Marktech Pvt. LTD, Bangalore. The matrix system consist of a medium viscosity epoxy resin (LAPOX L-12) and a room temperature curing polyamine hardener (K-6) Supplied by Atual industries LTD, Gujarat, India. Cellulose particles are used as filler material supplied by Maple biotech Pvt. LTD, Pune, India. To make the cellulose solutions for draw down coating, Cellulose is manually mixed with 50ml of Sodium Hydroxide (NaOH) to get a clear solution. Then, measured amount of cellulose and NaOH solution is poured and draw bar is moved on UD Glass fiber to get uniform distribution/coating using portable laboratory made semi-automated coating machine as shown in Figure 1. This machine can coat woven fiber/pre-preg of size 350 mm length and 130mm wide.

![Figure 1. Schematic of semi-automated coating machine](image-url)
Cellulose coated UD Glass fiber layers are allowed for drying about 2 minutes and then stacked one over the other to get a required thickness. As per the ASTM D 5528 initial crack/pre crack was made using 14 micron film separation at the middle plane of the specimen between 5th and 6th. Then the composite is cured under room temperature curing hardener using hand moulding method. Details of cellulose content used in tests are shown in Table 1.

| Sample code | Cellulose in % | Surface coating density (g/m²) |
|-------------|----------------|-------------------------|
| GE          | -              | -                       |
| CGEC-1      | 5              | 319.08                  |
| CGEC-2      | 7.5            | 333.33                  |
| CGEC-3      | 10             | 353.27                  |

3. Fracture Test

3.1 Mode – I fracture test

Mode I experiments had been conducted in accordance with ASTM-D5528. A thin film of thickness 13μm was introduced at the mid plane of laminates while preparing. Geometry with its dimensions of the double cantilever beam (DCB) specimen is shown in Figure 2. Calibrated uni-axial tensile machine of 40kN capacity was used to conduct mode I test. The specimen loading and its attachments are shown in Figure 3. Load was applied to the DCB specimens with displacement rate of 0.01mm/sec. In each coating configuration a batch of five DCB specimens were tested.

For the DCB test, the edges of all specimens were coated with white paint for the better visualization of crack propagation. A paper scale is attached on edge of specimen for measuring crack length during propagation. A light source and SONY 60X optical zoom digital camera was employed for monitoring crack growth in DCB specimen. The resulting load (P) and elongation (δ) were recorded for every 1mm increment.
up to 5mm and then continued crack propagation was measured for every 5mm incremental until the crack length reached to 90mm.

Figure 3. Specimen loaded in UTM machine.

Mode-I interlaminar fracture toughness for initiation and propagation were calculated using [12] equation (1)

\[
G_{IC} = \frac{3P\delta}{2B(\alpha + |A|)}
\]

Where \(P\): the load, \(\delta\): the load point displacement, \(B\): the specimen width, \(\alpha\): the delamination length, and \(\Delta\): correction factor rotation may occurs at the delamination front. \(\Delta\) is determined by experimentally with the aid of generating a least square plot of the cube root of compliance (\(C^{(1/3)}\)) as function of delamination length.

3.2 Mode II fracture test
Mode II End notch flexural (ENF) experiment had been performed in correspondence with the Japanese Industrial Standard (JIS K 7086) for UD Glass composite laminates.
A film inserted of 5 mm wide and 20 micron thick at crack location under the support to slide one wedge over another during loading. ENF test samples were loaded as per the diagram shown in Figure 4 with constant displacement rate of 0.01mm/sec. The specimens were positioned such that the initial crack length \( a_0 \) was 25mm from the left end of roller support.

Mode –II interlaminar fracture toughness was calculated using [13] equations (2), (3) and (4)

\[
\mathcal{G}_{IIc} = \frac{9a_1^2P_C^2C_1}{2B(2L^3 + 3a_1^2)}
\]

Where

\[
a_1 = \left( \frac{C_1}{a_0} - \frac{2}{3} \frac{C_1}{C_0} - 1 \right) \frac{L^3}{L^3 + 3a_1^2} \right)^{1/3}
\]

\[
a_0 = \frac{a_{0L} + a_{0C} + a_{0R}}{3}
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\[
a_0 = \frac{a_{0L} + a_{0C} + a_{0R}}{3}
\]

Where \( a_1 \) is the crack length for initial critical load calculated using equation(3), \( a_0 \) is the average value of crack length calculated using equation (4), \( a_{0L} \) is initial crack length of left side of test sample, \( a_{0C} \) is initial crack length of center part of test sample, \( a_{0R} \) is initial crack length of right side of test sample, \( P_C \) is the initial critical load, \( C_1 \) and \( C_0 \) are the load point compliances at the initial elastic and initial critical loads(mm/N), respectively. ‘B’ is the width of the test sample (mm) and ‘L’ is the distance between supporting point and loading point (mm).

4. Results and discussion

4.1 Mode –I Interlaminar fracture Toughness

The illustrative load versus elongation curves for DCB samples with and without cellulose coatings are plotted in Figure 5. The cellulose coated samplesCGEC-1, CGEC-2, and CGEC-3 show an enhancement
of 61%, 27 %, and 8% respectively in load carrying capacity at peak condition compared to the Glass Epoxy (GE) samples. The specimen CGEC-1 shown very prominent enhancement in peak loads of 61% compare to CGEC-2 coated with 333 g/m² and GE samples. The enhancement in fracture toughness CGEC-1 due to uniform distribution of cellulose over UD Glass fiber and good amount of fiber bridging as seen in Figure 8(b) during initiation propagation. Increase in cellulose content resulted in a thick cellulose layer is formed on fiber surface, due to this improper wetting on fiber surface during layup in CGEC 2 and 3 samples. Hence reduced fracture toughness was observed in CGEC 2 and CGEC 3 as compared to CGEC 1 sample.

![Figure 5. Load versus Elongation curves for GE and CGEC samples.](image)

Interlaminar fracture toughness versus crack length for each samples plotted in Figure (6). There is an increase in fracture toughness of the cellulose coated samples. CGEC-2 and CGEC-1 Samples shows good improvement of 57% and 46% respectively during crack initiation. CGEC-1 indicates the enhancement of fracture toughness by 9.43% in crack propagation compare to GE samples. It is higher fracture toughness observed in all CGEC samples due to better fiber bridging (shown in Figure 8(a,b,c& d)).

![Figure 6. Mode I interlaminar fracture toughness (G_{IC}) values versus crack length.](image)
The results summary of all DCB samples is shown in Figure 7. $G_{IC}$ values indicated in Figure 7(a) is significantly higher with a value of 33.976kJ/m² for the CGEC 1 samples compared GE samples having $G_{IC}$ of 28kJ/m². Similarly, propagation strain energy release rate ($G_{IP}$) was found to be 52kJ/m² for GE samples. However, the $G_{IP}$ value was reduced by 15% to 36% in all cellulose coated samples as shown in Figure 7(b).

The samples CGEC1 gave higher $G_{IC}$ values as compared to GE samples, but the $G_{IP}$ values are lower than the GE samples. This reduction of $G_{IP}$ values due low stiffness cellulose particle in low stiffer UD Glass fibers.

**Figure 7.** Mode I interlaminar fracture toughness (a) $G_{IC}$ values and (b) $G_{IP}$ values.

**Figure 8.** Fiber bridging in DCB (a) low fiber bridging in GE, (b) Higher fiber bridging, (c) Moderate fiber bridging and (d) low fiber bridging
4.2 Mode –II interlaminar fracture Toughness

The illustrative load versus elongation curves for Mode-II test samples with and without cellulose coatings are plotted in Figure 9. The cellulose coated sample CGEC-3 show an enhancement of 10% in load carrying capacity at its peak as compared to the GE sample. This is due to higher coating density and property of cellulose which were elongated in shear loading.

![Figure 9. Load versus Elongation curves for mode –II fracture test.](image)

Mode II interlaminar fracture toughness versus crack length for each samples plotted in Figure 10. There was increase in fracture toughness by 19% for CGEC-3 samples with respect to GE sample, further decrease in cellulose content resulted reduced Mode II fracture toughness in case of CGEC-1 and CGEC-2. Overall, there was much appreciable enhancement is not found in mode-II fracture toughness due low stiffness cellulose particle are incorporated in UD Glass fiber.

![Figure 10. Mode II fracture toughness (G_{IIc}) for GE and CGEC samples.](image)
5. Conclusion
Cellulose micro particles were successfully incorporated at its fiber level using semi-automated draw down coating technique followed by hand moulding method under room temperature cured of UD Glass epoxy composite laminates.

UD Glass fiber coated with cellulose, showed improvement in both mode-I initiation fracture toughness (GIc) and steady state propagation fracture toughness (GIIc) of up to 20% for CGEC-1 samples. A similar enhancement in mode-II interlaminar fracture toughness (GIIc) by 20% for CGEC 3 samples was observed. However decrease in cellulose coating density from 333 to 320 g/m², was resulted in reduced mode-II toughness. Overall, the enhancement in fracture toughness was witnessed by good amount of fiber bridging during delamination process. Hence the optimum cellulose coating surface density of 320 g/m² is the good compromise to get improved fracture toughness for mode 1 and CGEC-3 for mode II loadings is recommended from these investigations.

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