Investigation on pseudo-ductility to improve mechanical behavior in glass-cellulose epoxy composites

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Abstract: Nowadays composite materials exhibit sudden and catastrophic failure, which is undesirable for several applications. A new class of hybrid laminates was prepared using semi-automated draw down coating method with varying surface coating densities on unidirectional (UD) Glass fiber. Cellulose particles were coated on UD Glass fiber to investigate the effect of pseudo-ductility to improve mechanical behavior. Glass Cellulose epoxy hybrid laminate was fabricated with 5%, 7.5% and 10% of cellulose. Coating with 5% Cellulose produces a coating density of 319.08 g/m² and exhibits the appreciable pseudo ductile tensile stress–strain behavior with a non-linear variation at second part followed by linear variation at initial region. The response of tensile stress had shown 27% improvement in tensile modulus (330MPa) as compared to neat glass epoxy laminate with 0.04% of pseudo ductile strain. Further, flexural strength and inter-laminar shear strength of each specimen configuration were calculated and found good improvement in flexural strength with cellulose coated samples as compared to Glass-Epoxy laminate.

Keywords: Epoxy, Pseudo ductility, Cellulose particles

1. Introduction
Composite materials are becoming more imperative in the field of aerospace development, military and sports due to its high specific stiffness and strength, fatigue and corrosion resistance. Improvement in the field of materials science and innovation has led to manufacture high strength materials. In the latest years, composite materials assume a key part in aeronautic trade, car industry and other building applications as they show remarkable quality with high modulus to weight proportion.
Carbon fiber reinforced polymer (CFRP) composites consist of excessive stiffness and strength. They are, however, constrained via unexpected final failure. Experimental reviews have proven that it is possible to provide non-linear stress-strain response with carbon/epoxy angle laminates allowing the fibers to rotate under monotonic tension loading. Laminates made from such plies demonstrated remarkable resistance to micro cracking and delamination, creating additional strain and pseudo-ductility [1]. Suppressing delamination exhibited by angle ply laminate is the main predominant cause of premature failure provides amount of strains to failure and pseudo-ductile strain to develop. Concept of “excess length” obtained by fiber re-orientation in tested laminates is capable of existing pseudo-ductility in CFRP. Several angles between 15° and 45° are investigated. A good range of thin ply carbon/epoxy laminate stacks have achieved 1.23% pseudo-ductile strain & 927 MPa of maximum stress. For [±30] laminates, pseudo-ductile strain is greater than doubled to 2.88%, but strength was reduced through only 25%.

In prior experiences probably the most basic techniques to create pseudo-ductility is hybridization of fibers which will improve the failure mode of traditional composites. The novel hybrid architecture offered pseudo-ductile tensile stress-strain responses with a linear initial phase followed by way of a large plateau [2-4]. This result in 60% growth in modulus as compared to pure glass with the value of plateau stress 860MPa followed by 2% pseudo-ductile strain. Varying proportions of Glass Carbon hybrid configuration were used and showed [5] pseudo-ductile strains of 0.86% with a final failure strain of 1300 MPa. Thin CFRP angle-ply laminates and concluded that laminates consisting of symmetric pairs of ±26 plies with 0° layers on the mid plane have shown high performance pseudo ductile results when quasi-statically loaded in tension. This results in yield stress plateau of 700MPa and 2.22% pseudo-ductile strain [5]. A stable, pseudo-ductile failure form showing tremendous warning and margin earlier than ultimate failure, which has been verified with the developed using different lengths of S-Glass carbon UD sandwich hybrid structure [4, 6].

Michael R. Wisdom et al. [7] have studied three principal mechanisms to achieve pseudo-ductile namely, fiber reorientation, hybridization and mode II interfacial slip in discontinuous composites. With these mechanisms an unexpected catastrophic failure of conventional composites was reduced in tensile loading.

Natural fibers provide superior mechanical properties compared to synthetic fibers. Due to its superior mechanical property, the amount of energy required for manufacture greater than half of the required amount of synthetic fibers. Because of low energy consumption, non-irritating, nontoxic and biodegradable properties of eco-friendly natural fibers are great emerging composite fibers. Natural fiber composites prepared using jute, flax and wheat straw fibers reinforced with polypropylene matrix characterized chemically and have high cellulose content, 60-70 percent. These composites are characterized mechanically and exhibit efficient reinforcement fibers which increases fiber stiffness and strength values [8, 9].

K. Alagarraja et al. [10] has developed sisal-glass FRP composites and their mechanical properties are demonstrated. The results shown that by adding sisal fiber with Glass-Epoxy can improve mechanical properties. Also, they suggested that it can be used as alternative material for GRP composites.

The main focus of this investigation is to introduce pseudo ductile strain in UD glass epoxy composite by coating Cellulose micro particles on unidirectional (UD) Glass fiber. Cellulose particles are dispersed on UD glass lamina using improvised semi-automated draw down coating technique which was by earlier researchers [11, 12]. This is a new kind of technique presented in this work to coat cellulose particles on glass fiber instead of direct mixing in epoxy[13] for making hybrid laminates.
Glass cellulose hybrid laminates were made with 5%, 7.5% and 10% using hand layup method under room temperature curing hardener. Details of coating done on UD Glass fiber and test results were presented to reveal the pseudo ductility to improve the mechanical properties is discussed under the following sections.

2. Materials, Processing and Experimental techniques

2.1 Materials used
In the present work, UD E-glass fiber reinforcement was used and is supplied by Mark Tech Composites Pvt. Ltd. Bangalore, India. Epoxy resin is mixed with the hardener in the ratio 10:1 by weight. The matrix system consists of a medium viscosity epoxy resin (LAPOX L-12) and cured at room temperature. Polyamine hardener (K-6) supplied by ATUL, India Ltd, Gujarat, India. Cellulose Micro particles are supplied by Maple Biotech Pvt. Ltd. Pune, India. Details of cellulose micro particles, epoxy and hardener are shown in Table 1. Mechanical properties of E-glass fabric are listed in Table 2.

Table 1. Physical properties of epoxy resin, hardener and cellulose micro particles

| Material                  | Trade name and chemical name                        | Density, kg/m³ |
|---------------------------|-----------------------------------------------------|----------------|
| Cellulose micro particles | Consist of glucose residue linked with linear polymer | 1150           |
| Epoxy                     | LAPOX L-12 Diecydyl Ether of Biphenyl A (DGEBA)     | 1162           |
| Hardener                  | K-6 TriethyleneTetro amine (TETA)                    | 954            |

Table 2. Mechanical properties of Glass fiber and Epoxy resin

| Property                  | Glass fiber | Epoxy resin |
|---------------------------|-------------|-------------|
| Tensile modulus, GPa      | 72          | 4           |
| Tensile strength, GPa     | 3.5         | 0.1         |
| Poisson’s ratio           | 0.2         | 0.3         |
| Strain to failure         | 4.7         | 4.5         |

2.2. Preparation of cellulose solution for draw down coating
Physical appearances of Cellulose particles are shown in Figure 1(a). Initially particles are dissolved in organic solvents such as Acetone, DMF (Dimethylformamide), and Ethanol and observed settling time. But, it is seen that cellulose particles are settled in very short time. Hence Sodium Hydroxide (NaOH) was chosen to prepare solution for coating. The pure NaOH is a white crystalline solid available in the form of pallets shown in Figure 1(b).

Figure 1. Showing physical appearance of (a) Cellulose micro particles and (b) Sodium Hydroxide pallets
It is hygroscopic in nature and readily dissolves in water. To make 1 N (normal) solution of NaOH, it required 40g of Sodium Hydroxide dissolved in distilled water to make volume of 1 liter. Accordingly 50g NaOH solution was made and then Cellulose are dispersed and which gives better dispersion with higher settling time compared to solvents (Table 4). Settling time of cellulose particles in solvents as well as in NaOH solution were shown in Figure 2. The detailed properties of cellulose listed in Table 3.

Table 3. Properties of Powdered cellulose

| Description          | Powdered cellulose                                                                 |
|----------------------|-------------------------------------------------------------------------------------|
| Chemical name        | Glucose residue linked linear polymer in 1:4                                        |
| Chemical formula     | (C\textsubscript{12}H\textsubscript{20}O\textsubscript{10})\textsubscript{n}         |
| Density              | 0.0015 g/mm\textsuperscript{3}                                                    |
| Characteristics      | Solubility: A white or close to white, best or granular powder, almost              |
|                      | insoluble in water, in acetone, in ethanol, in toluene, in dilute acids and in      |
|                      | most natural solvents, fairly soluble in a 50 g/l solution of sodium hydroxide      |
| Description          | Purified, routinely disintegrated cellulose prepared by means of processing         |
|                      | alpha cellulose bought as a pulp from fibrous plant substances; happens as a        |
|                      | white, odorless substance consisting of fibrous particles                           |

Figure 2. Settling time of cellulose particles in solvents as well as in NaOH solution

Table 4. Settling time of cellulose in different organic solvent and NaOH solution

| Organic solvents (in ml) | Cellulose (in grams) | Time taken to settle down (in minutes) |
|--------------------------|----------------------|---------------------------------------|
| Acetone                  | 2                    | 5                                     |
| Ethanol                  | 2                    | 6                                     |
| DMF                      | 2                    | 15                                    |
| NaOH solution            | 2                    | 25                                    |
2.3 Methods
2.3.1. Coating Cellulose on UD Glass lamina
A semi-automated draw bar coating mechanism shown in Figure 3 was employed to coat cellulose particles on UD Glass lamina. This mechanism operates on a simple hand lever (Figure 3), giving precise weight on metering or coating rod and also prevents rotation of rod during coating stroke.

![Schematic of Semi-automated coating machine](image)

1. Base plate
2. Guide way
3. Hand lever
4. Fibre layer
5. Coating rod
6. Glass plate

![Coating Bar specifications](image)

- Total rod length = 160 mm
- Working rod length = 130 mm
- Rod diameter = 10 mm
- Wire diameter = 1 mm
- Wire material = Chrome plating stainless steel
- Film thickness = 0.1 mm

![Figure 4. Coating Bar specifications](image)

NaOH-Cellulose liquid solution was coated on UD Glass layer by the aid of semi-automatic draw bar coating method as shown in Figure 3. To get uniform surface coating density of NaOH-Cellulose on UD Glass surface a draw bar wound with wire gauge diameter of 1 mm is shown in Figure 4. Solution was poured on fiber layer surface using a syringe and was drawn through the draw bar by rotating hand lever. Un-coated and coated UD Glass fibers are shown in Figure 5. The procedure is repeated until coating is done on required number of fiber layers.

It is simple, cost effective and controlled technique to get uniform surface coating density with reduced fiber waviness and fiber misalignment during coating. The cellulose coated UD Glass laminas were staked one over the other to get required thickness using epoxy as matrix curing 24 hours under room temperature.
Figure 5. Images showing cellulose coated and uncoated fiber surface (a) Glass fiber before coating, (b) 5% cellulose with coating density 319.08 g/mm² (c) 7.5% cellulose with coating density 333.33 g/mm² and (d) 10% cellulose with coating density 353.27 g/mm².

After curing the specimens were cut as per the required size by referring ASTM standards. The specimens are coded according to the cellulose concentration and surface coating densities and listed in Table 5.

Table 5. Specimen codes with measured coating density with respect to cellulose concentration

| Specimen code | Cellulose content in % | Bar dia in mm | Weight before coating (grams) | Weight after coating (grams) | Surface coating density (g/m²) | Amount of Epoxy (grams) | No. of fiber layers |
|---------------|------------------------|---------------|-------------------------------|-----------------------------|-----------------------------|------------------------|-------------------|
| GEC 5         | 5                      | 1             | 10.53                         | 11.20                       | 319.08                      | 81.57                  | 10                |
| GEC 7.5       | 7.5                    | 1             | 10.53                         | 11.70                       | 333.33                      | 81.57                  | 9                 |
| GEC 10        | 10                     | 1             | 10.53                         | 12.4                        | 353.27                      | 81.57                  | 8                 |
| GE            | -                      | -             | -                             | -                           | -                           | 81.57                  | 11                |
| GEC           | -                      | -             | -                             | -                           | -                           | 81.57                  | 10                |

2.3.2. Tensile test

Tensile test determines the in-plane tensile properties of fiber reinforced polymer matrix composite as per ASTM standard D 5083[14] using uni-axial tensile test machine. Hybrid composite laminates were fabricated using hand layup method. The specimens were cut into required dimension shown in Figure 6, using a band saw cutter, the edges are finished by sand paper. The prepared specimen was
placed in a tensile machine and applied to tensile load until it breaks. The load extension values were recorded.

![Figure 6. Dimensions of the tensile test specimen](image)

2.3.3. Flexural test

Flexural test determines the flexural stiffness and strength properties of reinforced plastics. Test were conducted as per the ASTM D7264 [14] standard test procedure. This test method utilizes a three-point loading configuration using tensile machine. According to ASTM D7264 procedure a bar of rectangular cross section with 128mm in length, 13mm width and 4 mm thick rested on two supports and is loaded by means of loading nose midway between the supports is shown in Figure 7. The load extension values are recorded to determine the flexural properties (including strength, stiffness and load/deflection behaviour) of polymer matrix composite. The flexural stress can be calculated at any point on the load-deflection curve by following equations (1). [15]

![Figure 7. Dimension of the flexural test specimen](image)

\[
\sigma = \frac{3PL}{2bh^2}
\]  
(1)

Where: \( \sigma \) = stress at the outer surface at mid-span, MPa, \( P \) = applied force, N, \( L \) = support span, mm, \( b \) = width of beam, mm, and \( h \) = thickness of beam, mm.

Composite materials also undergo failure due to inter-laminar shear strength which consists of the determination of the resistance to delamination under shear forces parallel to the layers of the laminate.

The inter-laminar shear strength can be determined by following equation (2). [15]

\[
\sigma_{\text{int}} = \frac{3PL}{2bh^2}
\]  
(2)
\[ \tau = \frac{3P}{4bh} \]

Where: \( \tau \) = apparent inter-laminar shear strength, MPa, \( P \) = is the maximum load at the moment of first failure, \( b \) = width of beam, mm, and \( h \) = thickness of beam, mm

3. Results and discussion
The use of composite within the distinctive field is increasing day by day because of their superior mechanical properties. Engineering and scientists are working collectively since many years to seek out substitute solution for various materials which provides light weight, high strength to weight ratio. Within the present learn natural fibers are delivered to the glass fiber reinforced composite substances and their effect on mechanical properties is evaluated.

3.1 Tensile properties
The main aim of the tensile test is to determine the pseudo-ductile strain in Glass cellulose epoxy composites. Five specimens were prepared with the variation of cellulose content. These specimens are loaded in the universal testing machine (UTM). Stress-strain curve was plotted to display pseudo-ductile strain behavior in Glass cellulose composite.

Specimens were cut according to ASTM standard. Figure 6 shows the overall stress-strain graphs obtained for varied cellulose percentage and surface coating density in UD Glass-Epoxy laminates. Arrows and numbers in bracket indicate the beginning of pseudo ductile strain in Glass cellulose composite specimens.

![Stress Vs Strain](image)

**Figure 8.** Stress versus stain plots for pristine and cellulose coated specimens (Glass-E-Cellulose)

The numbers 1 to 1’ and up to 5 to 5’ shown in Figure 8 where the plot deviates from linear, was determined for each specimen type using intersection point of vertical line on the horizontal axis of the graph.

Final strain values were determined at a drop in stress towards end of failure process. This was necessary to compare the result obtained from various specimen types showing various failure modes. The stress plateau has shown by specimen GEC 5 exhibits failure strain 0.22% at stress value of 330 MPa which shows small amount increment in pseudo ductile strain between the region 1 and 1’. But for GE specimen showing no appreciable amount of nonlinear response between the region 4 and 4’. This indicates that by adding 5% Cellulose on UD Glass fiber gave 0.04% pseudo ductile strain in glass epoxy laminates. The enhanced pseudo ductile strain in Carbon epoxy laminates can be
achieved by coating optimized amount of Cellulose particles instead of combining glass in carbon fibers showed by J. D. Fuller et.al [5]. Further, the Specimen GEC 7.5 exists at 0.21% strain and stress value 220 MPa which is lower than specimen GEC 5, GEC 10, GE and GEC. Due to poor set up in the machine and material defect, failure for specimen GEC 10 occurs at stress never exceeding the level of the first load drop.

3.2 Flexural properties
The flexural test results of the tested specimen series are discussed here in detail. Figure 9 (a) shows the variation cellulose percentage with respect to flexural strength for Glass-Epoxy-Cellulose samples. The flexural strength obtained for GEC 5 sample shows higher stress values compared to samples with GEC 7.5, 10 and GE. GEC 5 exhibits maximum flexural strength of 1938 MPa as compared to GEC 7.5. It was observed that increase in cellulose content from 7.5% to 10% there was a decrease in the flexural strength of 17.36%.

![Figure 9. (a) Flexural strength values and (b) Inter-laminar shear strength values](image)

The most common test used for measuring shear delamination is short beam test where specimen is loaded in three point bending until delamination forms. Figure 9 (b) shows variation of cellulose with respect to inter-laminar shear strength (ILSS) for Glass-Epoxy-Cellulose samples. It was seen that the maximum inter-laminar shear strength of 30 MPa in GEC 5 as compared to GEC 7.5, GEC 10. It was estimated that 36% increase in ILSS for GEC 5 with respect to GE samples. Further, increase in cellulose content from 7.5% to 10% there is decrease in ILSS was observed.

4. Conclusion
New method of coating cellulose particles on Glass fiber was successfully made using semi-automated draw down coating technique. This method is simple, controlled and accurate measurement of surface coating density for varied cellulose percentage in UD Glass fiber lamina.

Tensile test results have shown that the coating of Cellulose on UD Glass fiber improves pseudo ductile strain up to 0.04% and a 26% improvement in flexural strength using Cellulose in case of a surface coating density of 319 g/m² with respect un coated glass laminates.

Enhancement in Inter laminar shear strength (ILSS) of 36% was found with surface coating density of 319 g/m². However it was observed that increase in cellulose content resulted in decreased ILSS.

5. Future scope
- The present study provides wide range of scope for future investigation and can extend it to fabricate new kind of composite by changing one of the phase material and angle orientation.
• Incorporation of cellulose with Glass-Epoxy fiber has been less studied area. Many other studies like effect of fiber orientation, pattern of loading can be made.

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