Visual Abstract

Influence of matrix modification on interlaminar fracture toughness of glass epoxy laminates using nano and micro fillers

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ABSTRACT. Fiber-reinforced polymer (FRP) is a composite material made of a polymer matrix reinforced with fibers. Hybrid composites are referred to as high-performance FRP materials used in safety-critical structural applications. Generally, FRP composite laminates are very weak in their out-of-plane properties, to address this issue unidirectional (UD) Glass laminates are prepared by modifying the matrix using plasma-treated multi-walled carbon nanotubes (MWCNTs) in epoxy matrix and compared fracture toughness characteristics with low-cost micro fillers like Aluminum oxide (Al_2O_3) and Sodium Carbonate (Na_2CO_3). These Nano and Micro fillers are loaded with 0.5wt%, 1wt% and 2wt% in the epoxy matrix while making FRP laminates. Adding these fillers into the epoxy matrix was caused a significant increase in

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the out-of-plane load-bearing capacity of the composites as compared to plain Glass epoxy laminates. Thus, the fracture toughness was enhanced by 20-26% and 14-17% under mode I and mode II loading respectively. Further, a Scanning electron microscopic analysis was also done on delaminated glass laminates to understand the failure mechanisms.

KEYWORDS. Fracture Toughness, Delamination, Hybrid matrix, MWCNTs, Laminate

INTRODUCTION

In a variety of industries, including automotive, aerospace, marine, and even civil engineering applications, fiber-reinforced polymer (FRP) composites have drawn attention for their exceptional qualities, including mechanical properties, impact resistance, design flexibility, and lightweight nature [1]. However, the through-thickness properties of the composites like delamination resistance [1-2] are very poor as compared to other directions. This is due to the poor performance of the matrix phase in the interlaminar region. The most common method for enhancing interlaminar fracture toughness is to modify the resin system using different fillers like graphene, CNT, MWCNT, ETBN, Al_2O_3, SiC, Graphene Nanoplatelets (GnPs), and Carbon Blacks (CBs), etc. [1-14]. The mode I fracture toughness of CNT-incorporated carbon epoxy prepreg with a loading range of CNT from 0 g/m^2 to 4 g/m^2 by an increment of 0.5 g/m^2 is examined. The results revealed that the incorporation of 1g/m^2 of CNT showed a 32% increase in fracture toughness [3]. The change in the failure sequence and increase in the mode I and mode II fracture toughness by 25% and 10% was reported for composites with 3wt% incorporation of CNT in CFRP laminates [2]. The influence of MWCNT on Carbon-Epoxy prepreg by varying the concentration of MWCNT in the range of 0.05 to 0.5 wt% on mode I and mode II fracture toughness was investigated and it was found that the concentration of 0.05wt% of MWCNT was optimum to enhance the mode I and mode II fracture toughness [4]. The addition of 1 wt% of MWCNT exhibited enhancement in mode I and mode II fracture toughness of Carbon fiber-reinforced epoxy composites [5]. Many researchers have concluded that the attributes of optimum weight percentage of reinforcements, length, aspect ratio, and orientation of reinforcements and/or fillers will play a significant role in enhancing fracture toughness [1-4,12-15]. Otherwise, agglomeration of particles leads to the development of the micro-cracks in the resin phase which is the cause of reduced fracture toughness in the interface region of the composites. Fiber bridging [6-7] is the key aspect in slowing down the rate of random propagation of cracks. The effect of milled Glass fiber (2.5, 5, 7.5, and 10% by weight of epoxy matrix) in Glass Epoxy composite under mode I and mode II were studied and found that good fiber/matrix interface, fiber bridging leads to improvement in the mode I and mode II fracture toughness significantly by 102% and 175%, respectively for 5 wt.% addition of milled glass fibers [8]. The comparative study on the effect of 1 g/m^2 incorporation of MWCNT and multilayer graphene (mG) in Carbon Epoxy fiber composite under mode I loading was studied and found an increase in fracture toughness of 12.3% by the addition of MWCNTs and 101.4% by the addition of mG in comparison to pristine Carbon Epoxy composite [9]. The effects of integrating micro-Al_2O_3 on to carbon fiber surface and its contents on the mechanical properties of carbon fiber-reinforced polymer composites were investigated. The results revealed that mode II interlaminar fracture toughness, impact strength, flexural properties, and initial modulus all increased with an increase in areal density until 15 g/m^2. The mode II interlaminar fracture toughness was 522 J/m^2, and the impact strength and flexural properties reached the maximum values [10]. The influence of Aluminum Oxide filler on fracture toughness properties of Chopped Strand Mat (CSM) E-Glass fiber reinforced epoxy resin matrix composites was evaluated and it was found that the fracture toughness of the 4wt% of Alumina in epoxy matrix-Glass fiber composite was the highest [11]. An effort was made to suppress the growth of crack by incorporating MWCNT, Al_2O_3, and SiC Micro fillers at the interface of unidirectional (UD) Glass fiber lamina, using a draw-down coating technique. The results revealed that 0.5wt% of Al_2O_3 in epoxy resin exhibited a 54% increase in interlaminar radial stress [12]. The effect of Al_2O_3 powder on the physical and mechanical properties of the polymer hybrid composites based on unsaturated polyester resin reinforced with carbon and glass fibers was assessed. The higher values of impact strength, fracture toughness, and flexural strength were obtained at 5wt% of Al_2O_3 in polyester resin for both fibers [13]. The influence of the addition of nanoparticles like multi-walled carbon nanotubes (MWCNTs), graphene nanoplatelets (GnPs), and carbon black (CB) in carbon fiber-reinforced polymer composites was assessed on mode I and mode II interlaminar fracture toughness. The results showed that the addition of MWCNTs and GnPs resulted in enhanced mode I and mode II fracture

toughness due to strong interaction between nanoparticles whereas the CBs showed lower fracture toughness due to lesser surface area and modulus of elasticity [14]. The effect of aspect ratio (AR) and specific surface area (SSA) of Graphene Nano-Platelets (GNPs) on the interlaminar fracture behavior of carbon fiber-reinforced polymer (CFRP) composites was studied. The results showed that the interlaminar fracture properties improved with the addition of different types of GNPs. The highest improvement of both Mode I and II fracture toughness was attained with the addition of GNPs [15]. The impact of nano-additives like Al_2O_3 and TiO_2 at 1 wt% on the mechanical, wear, and water sorption/solubility performance of commercially available restorative materials used in dental therapy (Nexcomp Flow A2). The results exhibited that the Al_2O_3 nanomaterials, which have greater hardness, showed higher wear resistance than the neat RC composites. Incorporating nano-additives into dental composites improves diametrical strength because stress is transferred between matrix and particle. The composite with Al_2O_3 nanoparticles has the best wear resistance and tensile strength compared to TiO_2 nanoparticles [16]. The nano-Aluminum oxide (Al_2O_3), nano-Silicon Carbide (SiC), or a hybrid of them were mixed into epoxy resin with an ultrasonic system with various weight percentage ratios of the nanoparticles. The results indicated that the addition of Al_2O_3 nanoparticles to the epoxy resin resulted in higher resistance to wear than when SiC or hybrid nanoparticles were added [17]. Recently many studies have found that epoxy nanocomposites as a friction material for mechanical and tribological applications still face some challenges, such as agglomeration and porosity, which has reportedly led to their mechanical property degradation and also affects their tribological performance [18].
This investigation aims to assess the effect of micro and nanofiller i.e Aluminum oxide Sodium Carbonate and MWCNT on mode I and mode II fracture toughness of Glass Epoxy composites. The incorporation and proper dispersion of fillers in epoxy resin were achieved with a magnetic stirrer. The hand layup technique was adopted to manufacture the composite laminates. The mode I and mode II test were performed as per ASTM D 5528 standard [19] and ASTM D 7905 standard [20] respectively. The SEM examination was used to describe the fiber-matrix interface and failure mechanisms.

EXPERIMENTATIONS

Materials

The reinforcement used in this study is unidirectional (UD) Glass fiber supplied by Mark Tech Pvt. Ltd, Bangalore, India. The Epoxy resin (Lapox L12) and Polyamine Hardener (K-6) used are supplied by Atul Industries Pvt. Ltd, Gujarat, India. The additives MWCNTs used are supplied by Bayer Material Science C150P, USA. The micro fillers Aluminium Oxide (Al_2O_3) and Sodium Carbonate (Na_2CO_3) were supplied by Venkateshwar Chemicals, Dharwad, India. The properties of epoxy resin and Glass fiber reinforcements are depicted in Tab. 1 and Tab. 2 respectively.

Properties	Epoxy (Lapox L12)
Density (kg/m^3)	1150-1200
Viscosity at 20°C (m Pa.s)	9000-12000
Storage temperature (°C)	4-45
Tensile strength (N/mm^2)	50-60
Compressive strength (N/mm^2)	110-120
Flexural strength (N/mm^2)	130-150
Impact strength (kJ/m^2)	17-20
Modulus of elasticity (N/mm^2)	4400-4600
Coefficient of linear Thermal expansion (10-6/°C)	64-68
Thermal conductivity (kcal/mh°C)	0.211
Water absorption at 20°C /10days (%w/w)	0.5-0.6

Table 1: Properties of DGEBA epoxy resin (Lapox L12).

Properties	E-Glass fiber (220 gsm)
Weave type	Plain
Density (kg/m^3)	2500
Thickness (mm)	0.19
Young's modulus (GPa)	33
Tensile strength (MPa)	630
Compression strength (MPa)	510
Flexural strength (MPa)	810

Table 2: Properties of Glass fiber reinforcements.

Matrix modification

The epoxy matrix used is Lapox L12 which is modified by the inclusion of additives like CNTs, Aluminum Oxide (Al_2O_3), and Sodium Carbonate (Na_2CO_3). The additives were added to the matrix in different concentrations namely 0.5wt%,1wt%, and 2wt%. The required quantity of additives is taken and mixed in a known quantity of epoxy resin with the aid of a mechanical stirrer. Further, the modified matrix is applied to the glass fiber laminas manually to make the composite laminates for Mode I and Mode II tests.

Fabrication of composites

The UD Glass Fiber with Modified Epoxy Composites is manufactured using the hand layup technique for mode I and mode II tests. The required thickness of the composite laminates is attained by maintaining an equal number of UD Glass fibers at the top and bottom of the pre-crack. The pre-crack is achieved by using a separation film of 14-micron thickness at the mid-plane of the laminates. The UD Glass fibers are laid one over the other and modified epoxy resin is coated and dead weight is placed on the composite and cured. The curing of composite laminates is done at room temperature. The cured laminates are trimmed for sizing in conformance with the ASTM D 5528 standard [19] for Mode I loading and ASTM D 7905 standard [20] for Mode II loading. For each arrangement, six samples were fabricated. Tab. 3. gives the laminate codes of Modified Epoxy/UD Glass Fiber Composites and their compositions.

Laminate Codes	Matrix Modifier	Glass Fiber (wt%)	Modifier (wt%)	Epoxy (wt%)
GE	--	50	--	50
MWCNT/GE 1	MWCNT	50	0.5	49.5
MWCNT/GE 2	MWCNT	50	1	49
MWCNT/GE 3	MWCNT	50	2	48
Al_2O_3/GE 1	Al_2O_3	50	0.5	49.5
Al_2O_3/GE 2	Al_2O_3	50	1	49
Al_2O_3/GE 3	Al_2O_3	50	2	48
Na_2CO_3/GE 1	Na_2CO_3	50	0.5	49.5
Na_2CO_3/GE 2	Na_2CO_3	50	1	49
Na_2CO_3/GE 3	Na_2CO_3	50	2	48

Table 3: Laminate Codes of Modified Epoxy Glass Fiber Composites and Their Compositions.

TEST METHODS

Mode-I Test

Double Mode I fracture test is performed in accordance with ASTM D 5528 standard [19]. The standard Double Cantilever Beam (DCB) specimens are prepared. The pre-crack in the laminates is maintained using a thin film of 14-micron thickness. The specimens are attached with piano hinges on the top and bottom surfaces in order to subject them to mode I loading. The uniaxial testing machine of 10kN capacity was used for the test. The cross-head displacement speed of 2mm/min is maintained. A total of five samples for each type were tested. A high-definition digital camera of SONY make with a resolution of about ±0.5mm was used to record the growth of crack during the test. The concept of modified beam theory (MBT) is used to determine the critical strain energy rate in accordance with ASTM standards using Eqn. 1.

$$G_{IC} = \frac{3P\delta}{2B\left(a+|\Delta|\right)} \tag{1}$$

where P is the load or force, δ is the load point displacement, B is the specimen width, a is the delamination length, and Δ is the correction factor rotation that may occur at the delamination front. Δ is calculated empirically using a least-squares plot of the cube root of compliance (C ^ (1/3)) as a function of delamination length.

Mode-II Test

The mode II fracture test is performed in line with ASTM D 7905 standard [20]. The Three Point End Notched Flexural (ENF) specimens are fabricated. Tinius Olsen UTM with a 10 kN capacity and a 1 mm/min crosshead speed was used for the tests. The span length of each specimen was held constant during testing at 100 mm in accordance with ASTM D 7905 standard. The compliance calibration method was used to calculate the G_{IIC} values for non-pre-cracked specimens in accordance with the procedures given in the standard. The relationship between the beam compliance and the fracture length can be seen in Eqn. 2.

$$C = A + ma^3 \tag{2}$$

where A and m are the Crack Compliance coefficients, C – Compliance of beam, a – Crack length
G_{IIC} can be determined using Eqn. 3

$$G_{IIC} = \frac{3ma_0^2P_{max}^2}{2b} \tag{3}$$

where a_0 – Delamination length for ENF test, P_{max} – Maximum load value in a load-displacement curve, b - Width of the specimen

Fracture Morphology

The delaminated fracture surface of composite materials at the interface with the fiber matrix was examined using a Zeiss GeminiSEM-300 operating at 30 keV. In order to fit inside the vacuum chamber of the scanning electron microscope (SEM), the samples were chopped to a size of 10mm x 10mm.

RESULTS AND DISCUSSIONS

Mode I Interlaminar Fracture Toughness

Fig.1.a depicts the load versus displacement plots of different matrix modified with MWCNT/Glass fiber epoxy along with plain Glass epoxy DCB composite specimens. The matrix modified with 1% MWCNT-Glass epoxy composites showed higher load-bearing capacity as compared to 0.5% MWCNT-Glass epoxy composites and 2% MWCNT-

Glass epoxy composites. The pristine Glass-Epoxy composites have lower load-bearing capacity. The addition of MWCNT in optimum quantity as a filler to modify the matrix has increased the stiffness of unidirectional GE composites.

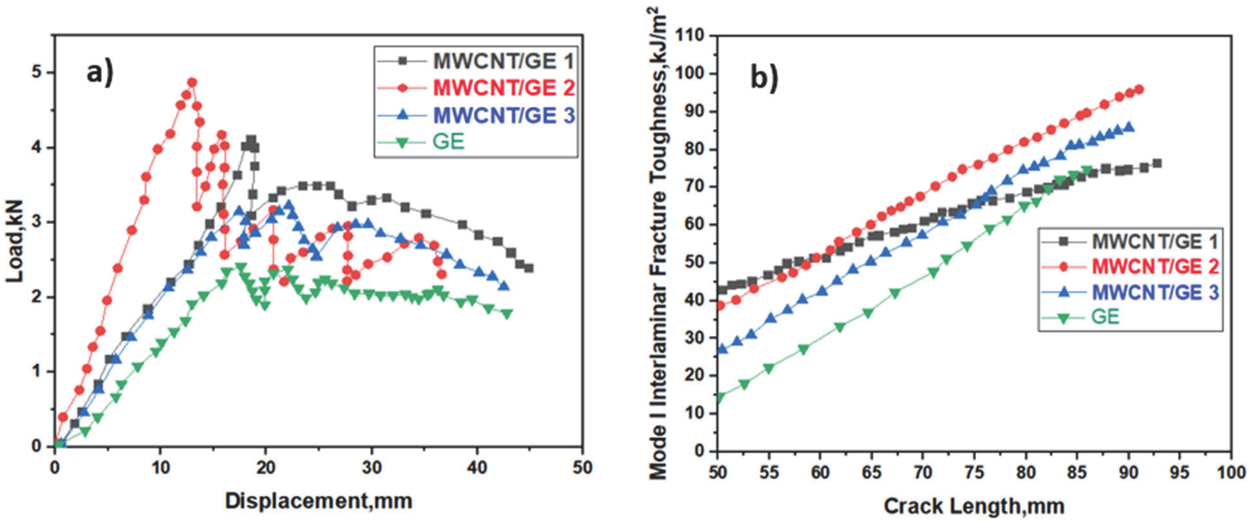

Figure 1: Load Mode I test of MWCNT modified matrix/Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode I Interlaminar Fracture Toughness Values versus Crack Length.

Fig. 1b depicts the delamination resistance curves (R-curves) of all the concentrations of matrix modified with MWCNT-Glass epoxy and pristine Glass epoxy composites According to Fig. 1b the composite with 1% MWCNT matrix modified-Glass epoxy composite showed a massive increase in fracture toughness as the crack propagated. This increase in fracture toughness can be attributed to fiber bridging as seen in Fig. 2(d). However, the composite with 0.5% MWCNT-Glass epoxy composite displayed higher toughness at the initiation. The mode I interlaminar fracture toughness of pristine glass epoxy composites was observed to be lower as compared to all other matrix-modified Glass epoxy composites. The amount of fiber bridging during crack propagation and these toughness values are well in accord. The plot of initiation and propagation fracture toughness values (G_{IC} & G_{IP}) is depicted in Fig. 3(a) and Fig. 3(b).

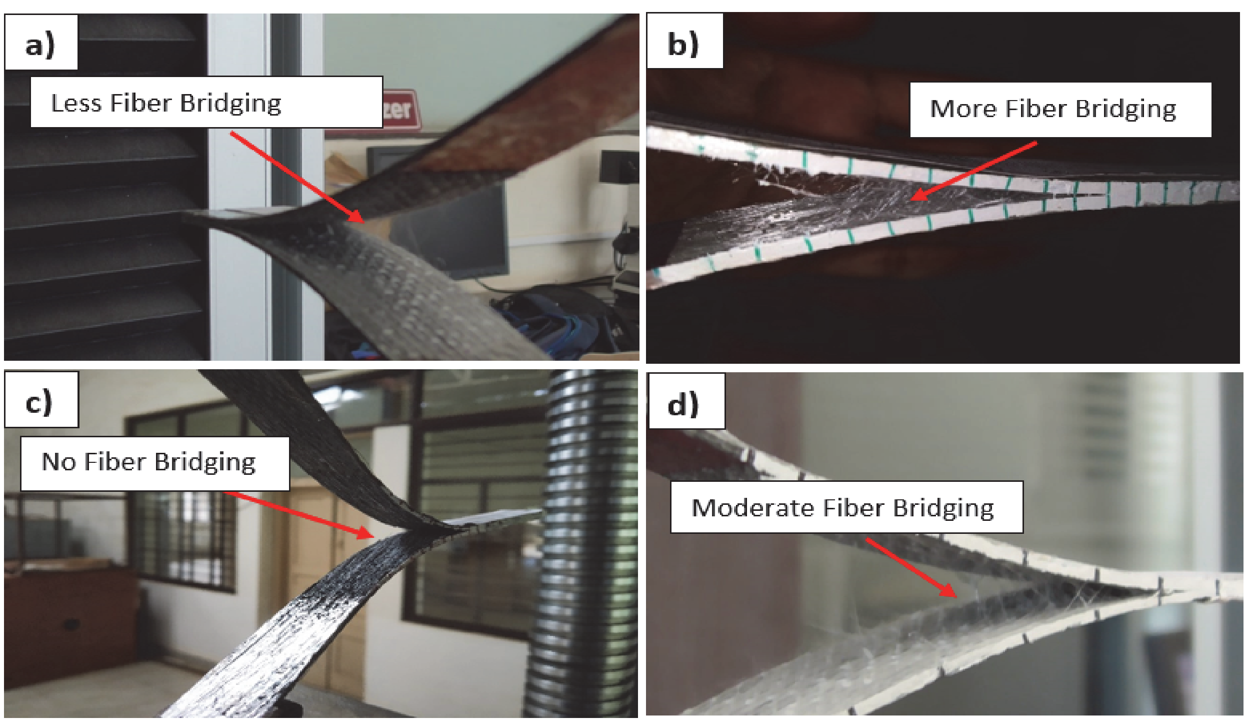

Figure 2: Fiber Bridging in Composites of Epoxy Matrix Modified with MWCNT- Glass Fiber (a) Plain Glass Epoxy (b)MWCNT-GE 2 (c) MWCNT-GE 3 (d) MWCNT-GE 1

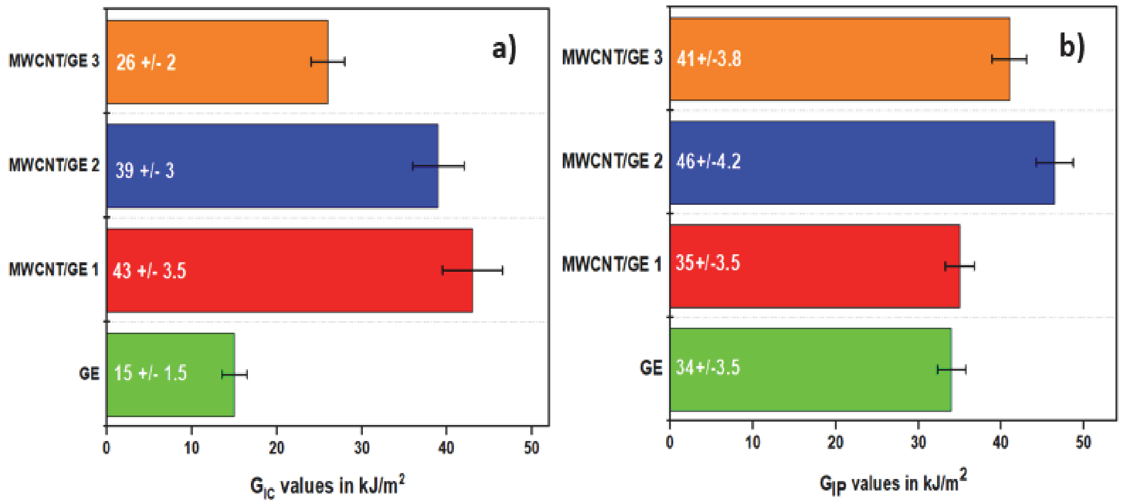

Figure 3: Mode I Interlaminar Fracture Toughness of Matrix Modified with MWCNT and Pristine Glass Fiber (a) G_{IC} values (b) G_{IP} values.

The composite with 0.5% MWCNT- Glass epoxy laminate displayed higher toughness at the initiation whereas, the composite with 1% MWCNT-Glass epoxy exhibited higher toughness values during crack propagation. This means that MWCNT matrix-modified laminates have better resistance against crack propagation in comparison with plain Glass epoxy composites.

Fig. 4a represents the load versus displacement curves of composite laminates which comprises a modified epoxy matrix with Aluminium oxide (Al_2O_3) and UD Glass fiber. The various concentrations of Al_2O_3 which are added to the epoxy matrix are 0.5wt%,1wt%, and 2wt%. The aluminum oxide (Al_2O_3) is mixed with an epoxy matrix using a mechanical stirrer.

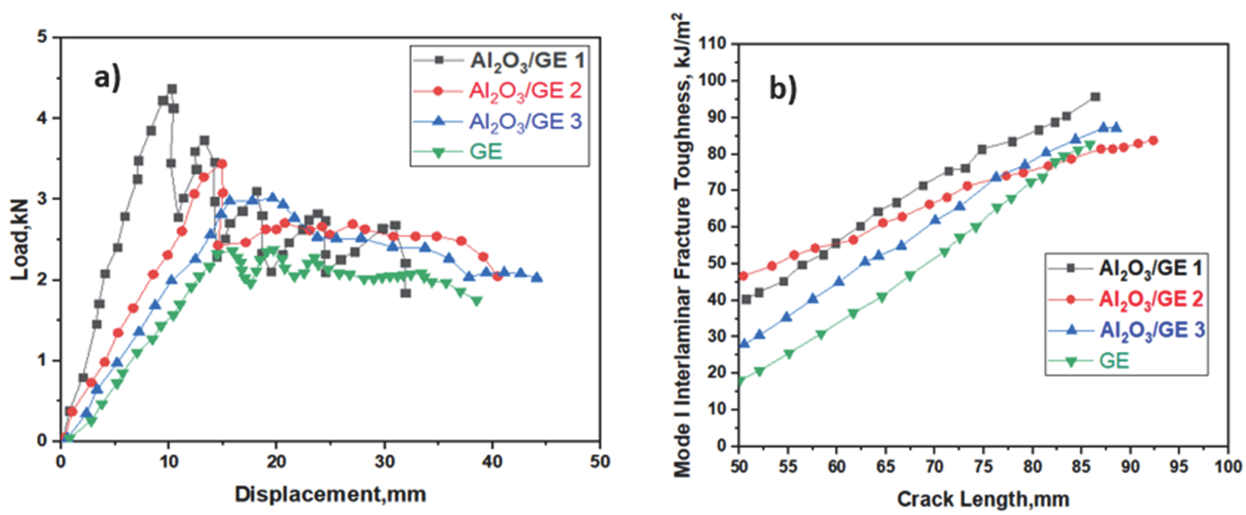

Figure 4: Mode I test of Al_2O_3 modified matrix-Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode I Interlaminar Fracture Toughness Values versus Crack Length.

The epoxy matrix modified with 0.5wt% Al_2O_3 - Glass fiber laminate exhibited higher load-carrying capacity as compared to other concentrations of Al_2O_3 - Glass fiber composites but pristine Glass Epoxy composites exhibited lower load-carrying capacity. The addition of 0.5wt% Al_2O_3 was found to slightly increase the stiffness of the Glass Epoxy composites. Fig. 4b shows the delamination resistance curves (R-curves) of all the concentrations of epoxy matrix modified with Al_2O_3-Glass epoxy composites and pristine Glass epoxy composites. The R-curves plotted indicated that composites of all the concentrations of epoxy modified with Al_2O_3- Glass epoxy composites considered for the study outperformed the pristine glass epoxy composites.

From Fig. 4b, it is observed that composites consisting of 1wt% Al_2O_3 in Glass epoxy composites exhibited higher toughness during crack initiation but the composites consisting of 0.5wt% Al_2O_3 in Glass epoxy composites showed higher toughness during crack propagation. The increase in toughness during crack propagation of composites consisting of 0.5wt% Al_2O_3 in Glass epoxy composites may be attributed to a good amount of fiber bridging which can be seen in Fig. 5(a) to Fig. 5(d).

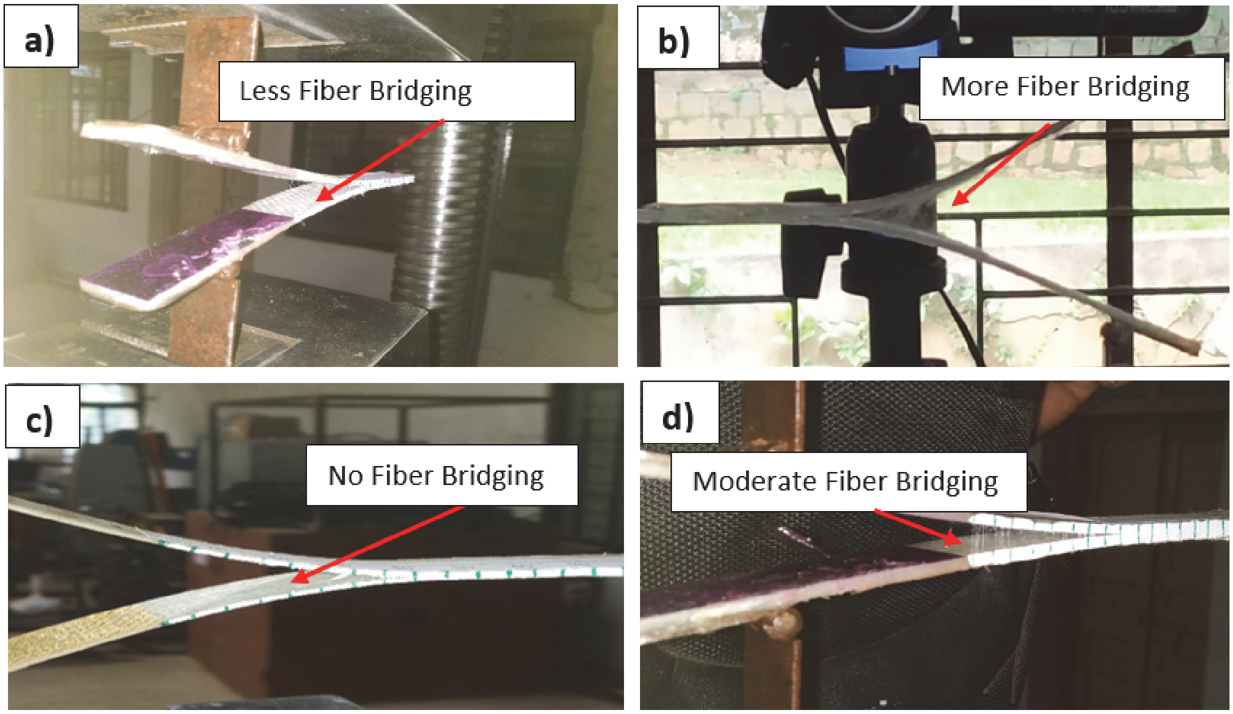

Figure 5: Fiber Bridging in Composites of Matrix Modified with Al_2O_3-Glass Fiber (a) Plain Glass Epoxy (b) Al_2O_3-GE 1 (c) Al_2O_3-GE 3 (d) Al_2O_3-GE 2.

Fig. 6 (a) and Fig. 6 (b) show the plot of initiation (G_{IC}) and propagation (G_{IP}) fracture toughness values respectively. The composites consisting of 1wt% Al_2O_3 in Glass epoxy composites exhibited higher toughness during crack initiation and the composites consisting of 0.5wt% Al_2O_3 in Glass epoxy composites exhibited higher toughness during crack propagation. Overall, it is observed that epoxy-modified composites with different concentrations of Al_2O_3 with Glass fiber reinforcement exhibited better fracture toughness as compared to plain Glass epoxy composites.

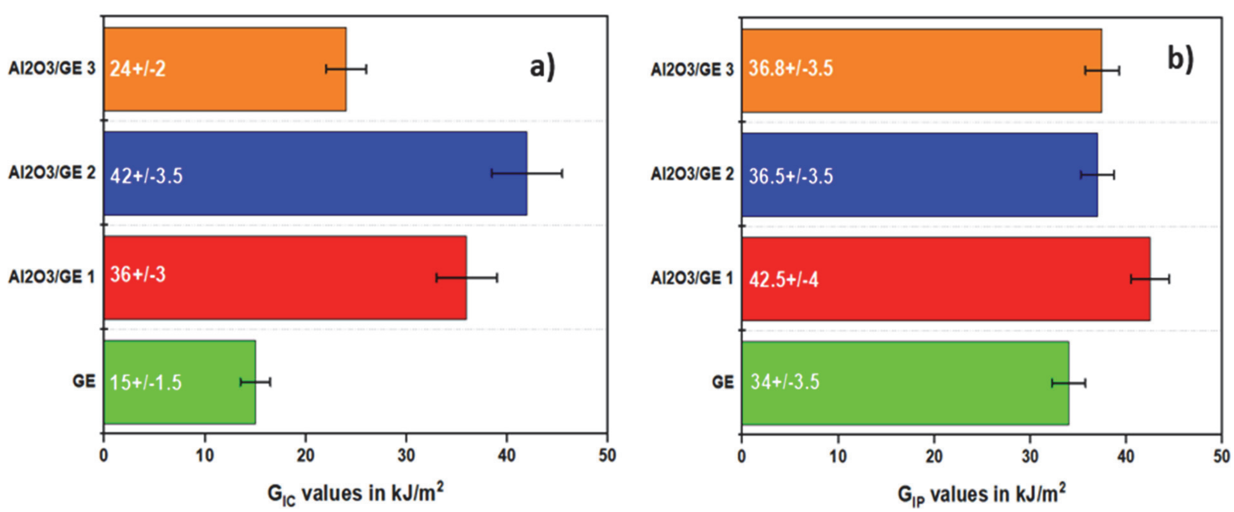

Figure 6: Mode I Interlaminar Fracture Toughness of Epoxy Matrix Modified with Al_2O_3 and Pristine Glass Fiber (a) G_{IC} values (b) G_{IP} values.

Fig. 7a represents the load versus displacement curves of composites which comprises a modified epoxy matrix with sodium carbonate (Na_2CO_3) fillers and Glass fibers. The different concentrations of Sodium Carbonate fillers added to epoxy are 0.5wt%,1wt%, and 2wt%. The filler Na_2CO_3 is thoroughly mixed with epoxy matrix using the mechanical stirrer method.

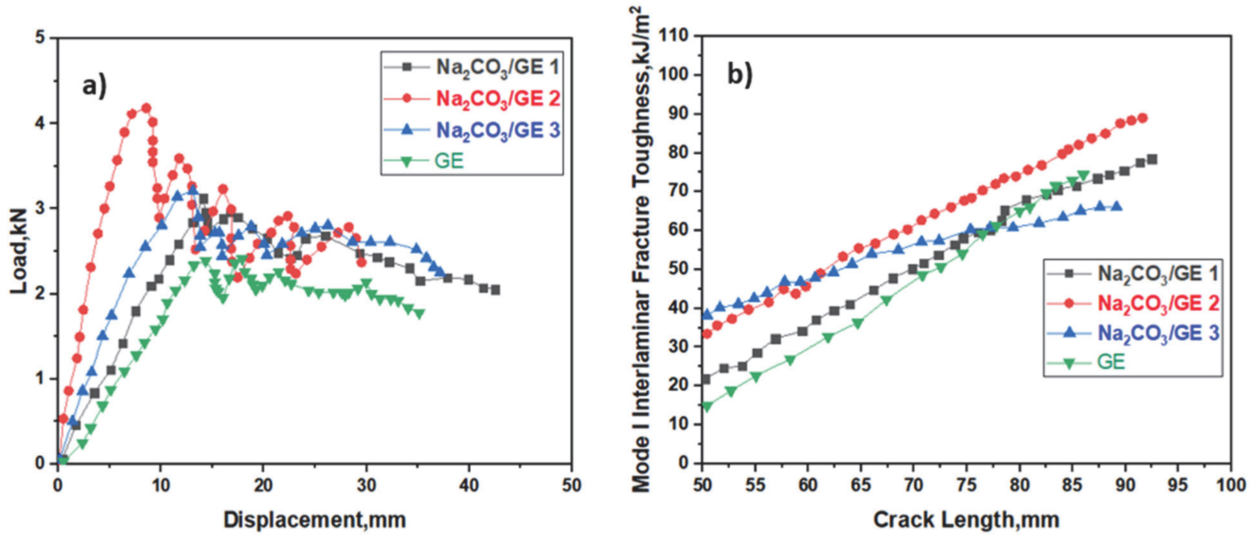

Figure 7: Mode I test of Na_2CO_3 modified matrix-Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode I Interlaminar Fracture Toughness Values versus Crack Length.

The epoxy matrix modified with 1wt% Na_2CO_3 and Glass fiber exhibited higher load-carrying capacity in comparison with other concentrations of Na_2CO_3 and Glass fiber composites. The pristine Glass Epoxy composites exhibited lower load-carrying capacity. The overall stiffness of the Glass Epoxy composites is found to increase with the addition of Al_2O_3. Out of all the concentrations of Na_2CO_3 used, 1wt%Na_2CO_3 modified epoxy matrix Glass fiber composite showed relatively higher load-bearing capacity.

Fig. 7b shows the delamination resistance curves (R-curves) of all the concentrations of epoxy matrix modified with Na_2CO_3-Glass epoxy composites and pristine glass epoxy composites. According to the R-curves displayed, the pristine glass epoxy composites were outperformed by composites of all concentrations of epoxy treated with Na_2CO_3.

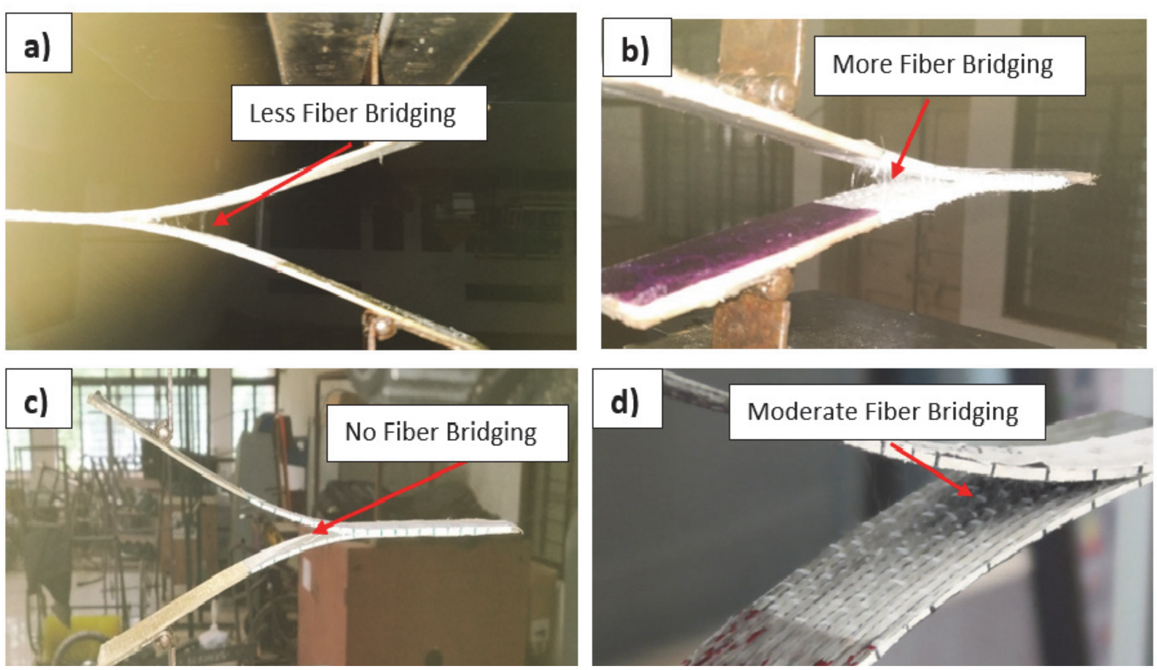

Figure 8: Fiber Bridging in Composites of Epoxy Matrix Modified with Na_2CO_3-Glass Fiber (a) Plain Glass Epoxy (b) Na_2CO_3-GE 2 (c) Na_2CO_3-GE 1 (d) Na_2CO_3-GE 3.

The mode I Interlaminar fracture toughness values against the crack length of the composites made of Na_2CO_3 modified matrix and Glass fiber and plain Glass epoxy composites are shown in Fig. 7b indicates that composites consisting of 2wt% Na_2CO_3 modified matrix-Glass fiber composite exhibited higher toughness during crack initiation and composites made of 1wt% Na_2CO_3 modified matrix-Glass fiber composite displayed higher toughness during crack propagation. The rise in the values of fracture toughness during crack propagation may be attributed to a considerable amount of fiber bridging [6] which can be seen in Fig. 8(a) to Fig. 8(d).

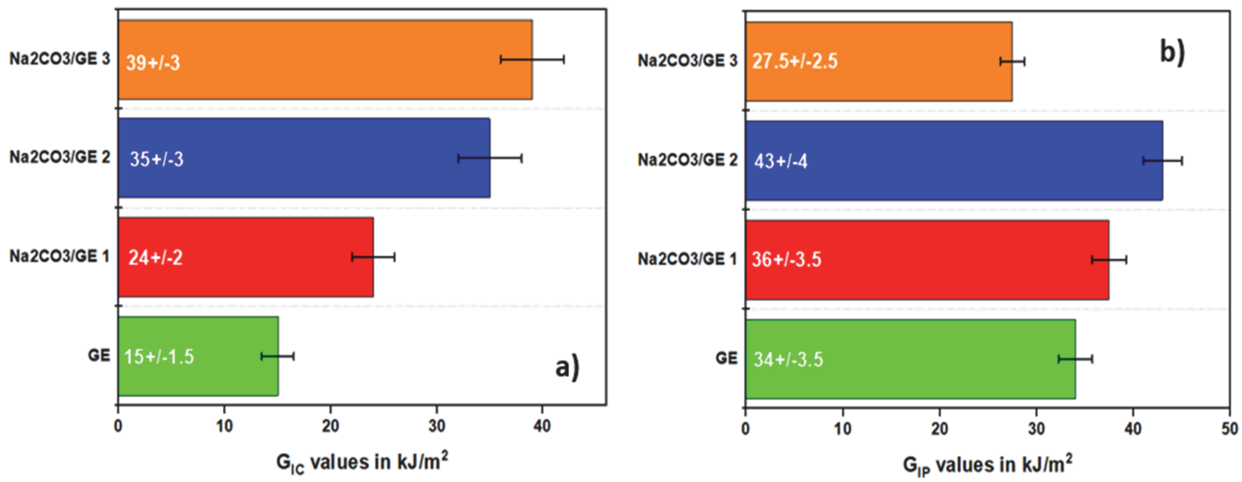

Figure 9: Mode I Interlaminar Fracture Toughness of Epoxy Matrix Modified with Na_2CO_3 and Pristine Glass Fiber (a) G_{IC} values (b) G_{IP} values.

Fig. 9 (a) and Fig. 9 (b) show the fracture toughness values during crack initiation (G_{IC}) and crack propagation (G_{IP}) respectively. The composites consisting of 2wt% Na_2CO_3 in Glass fiber epoxy composites exhibited higher toughness during crack initiation and the composites consisting of 1wt% Na_2CO_3 in Glass fiber epoxy composites exhibited higher toughness during crack propagation. Overall, it is observed that composites consisting of epoxy matrix modified with 2wt% Na_2CO_3 in Glass fiber epoxy composites exhibited higher toughness during crack initiation but during crack propagation, the same concentration of Na_2CO_3 exhibited very less fracture toughness. The Tab. 4 shows the details of number of specimens tested for mode I fracture toughness during crack initiation (G_{IC}) with standard deviation,average and co-efficient of variance.

SL.No	Laminate	No.of Specimens Tested	Average value of Mode I fracture toughness		Standard deviation of Mode I fracture toughness		Co-efficient of variance of Mode I fracture toughness	
			G_{IC}	G_{IP}	G_{IC}	G_{IP}	G_{IC}	G_{IC}
1	GE	6	15	34	1.5	3.5	10	10.29
2	MWCNT/GE 1	6	43	35	3.5	3.5	2.94	10
3	MWCNT/GE 2	6	39	46	3	4.2	3.62	9.13
4	MWCNT/GE 3	6	25	41	2	3.8	4.38	9.26
5	Al_2O_3/GE 1	6	36	42.5	3	4	8.33	9.41
6	Al_2O_3/GE 2	6	42	36.5	3.5	3.5	8.33	9.58
7	Al_2O_3/GE 3	6	24	36.8	2	3.5	8.33	9.51
8	Na_2CO_3/GE 1	6	24	36	2	3.5	8.33	9.72
9	Na_2CO_3/GE 2	6	35	43	3	4	8.57	9.30
10	Na_2CO_3/GE 3	6	39	27.5	3	2.5	7.69	9.09

Table 4: Number of samples tested, Average, Standard deviation, Co-efficient of variance for Mode I fracture toughness during crack initiation (G_{IC}) and crack propagation (G_{IP}).

Mode II Interlaminar Fracture Toughness

The load versus displacement curves from Mode II ENF tests for GFRP composites with epoxy matrix modified with MWCNT and pristine Glass epoxy are shown on a graph in Fig. 10a. It was seen that the composites consisting of epoxy matrix modified with 1wt% MWCNT in Glass fiber epoxy composites exhibited higher load-bearing capacity in comparison with other concentration composites and plain Glass epoxy composites. The slope of composites comprised of epoxy matrix modified with 2wt% MWCNT with Glass fiber is similar to that of composites consisting of epoxy matrix modified with 1wt% MWCNT with Glass fiber composites under mode II loading. This shows that the stiffness of all the composites is almost nearly similar. The addition of 1wt% MWCNT to the epoxy matrix proved to be effective in increasing the toughness of the composites.

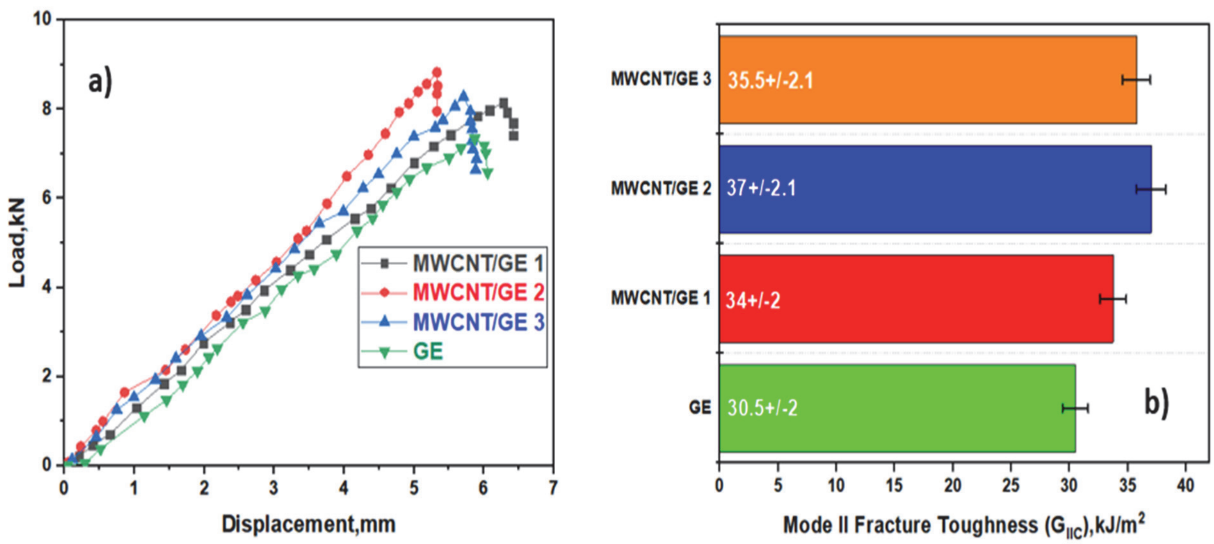

Figure 10: Mode II test of MWCNT modified matrix/Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode II Interlaminar Fracture Toughness (G_{IIC}).

Fig. 10b shows the mode II interlaminar fracture toughness for the MWCNT-modified composites under consideration. The composites comprised of epoxy matrix modified with various concentrations of MWCNT displayed higher fracture toughness as compared to plain Glass epoxy composites. However, the composites with epoxy matrix modified with 1wt% MWCNT in Glass fiber exhibited approximately 19% improvement in their fracture toughness as compared to plain Glass epoxy composites. Whereas, increasing the concentration of MWCNT did not increase the fracture toughness of composites. This was proven in our earlier studies [12] on L-Bend laminates. The reason may be ascribed to the increase in the brittle behavior of the composites. Further, reducing the concentration of MWCNT also did not increase the fracture toughness in the composites. Hence, it is found that the addition of 1wt% MWCNT in the matrix proved to be efficient in achieving the higher toughness values.

The load versus displacement curves from mode II ENF tests for composites with epoxy matrix modified with Al_2O_3 in Glass fiber and pristine Glass epoxy are shown on a graph in Fig. 11a. It is observed that the epoxy matrix modified with all the concentrations of Al_2O_3 considered for this study showed higher load-bearing capacity when compared with plain Glass epoxy composites. But the behavior of all the composites which are comprised of various percentages of Al_2O_3 was almost nearly similar. Therefore, the stiffness from all curves of the epoxy matrix modified with Al_2O_3 is nearly the same.

An interesting observation was seen the load-bearing capacities of composites comprised of epoxy matrix modified with 1wt% Al_2O_3-Glass fiber and 0.5wt% Al_2O_3-Glass fiber were almost identical.

Fig. 11b displays the mode II interlaminar fracture toughness for the composites under investigation. The FRP composites contained epoxy matrix modified with different concentrations of Al_2O_3 and exhibited higher fracture toughness in comparison with plain Glass epoxy composites. The mode II fracture toughness of composites made of epoxy matrix modified with Al_2O_3-Glass fiber was higher as compared to pristine glass epoxy composites. However, the composite consisting of 1wt%Al_2O_3 in an epoxy matrix exhibited higher fracture toughness when compared to other concentrations of Al_2O_3. The increase in the concentration of Al_2O_3 showed no improvement in fracture toughness.

Fig. 12a shows load versus displacement plots from mode II ENF tests for composites with epoxy matrix modified with Na_2CO_3 in Glass fiber and pristine Glass epoxy composites. The Glass epoxy laminates composites comprised of epoxy matrix modified with Na_2CO_3 exhibited comparable higher load-bearing capacities in comparison to plain Glass epoxy composites. The inclusion of Na_2CO_3 in Glass fiber epoxy composites showed improvement in load-carrying capacity. The slope of the curves of various concentrations of Na_2CO_3 was almost the same and hence stiffness also remains the same in all the composites. The composites with 1wt% Na_2CO_3 in Glass fiber epoxy composites displayed higher load-bearing capacity as compared to other concentrations like 0.5wt% and 2wt% addition of Na_2CO_3 in Glass fiber epoxy composites. Overall, the load-bearing capacity of plain Glass epoxy composites.

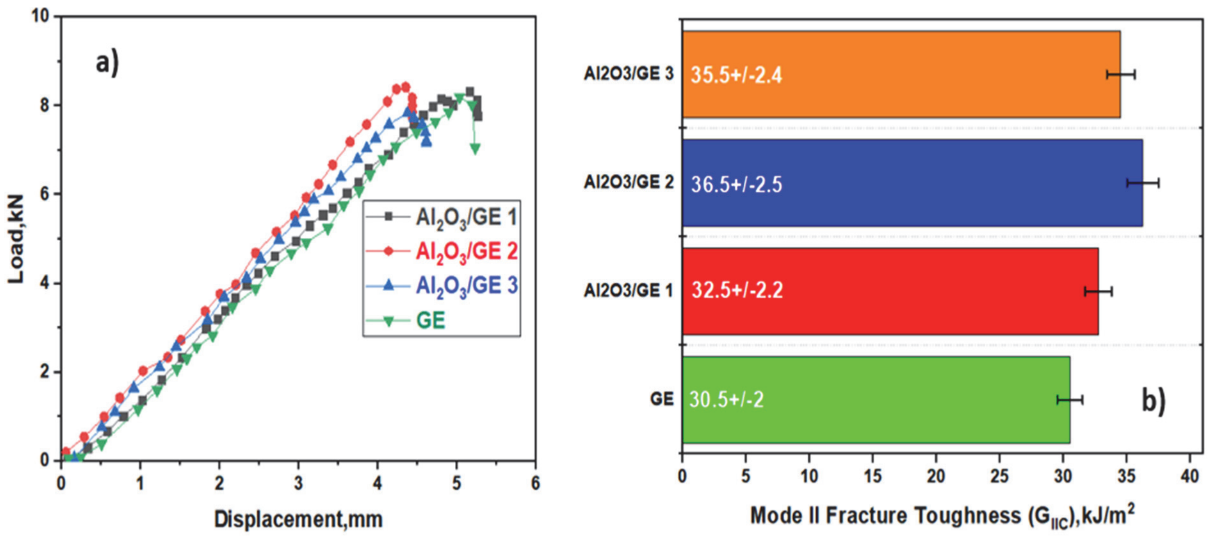

Figure 11: Mode II test of Al_2O_3 modified matrix/Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode II Interlaminar Fracture Toughness (G_{IIC}).

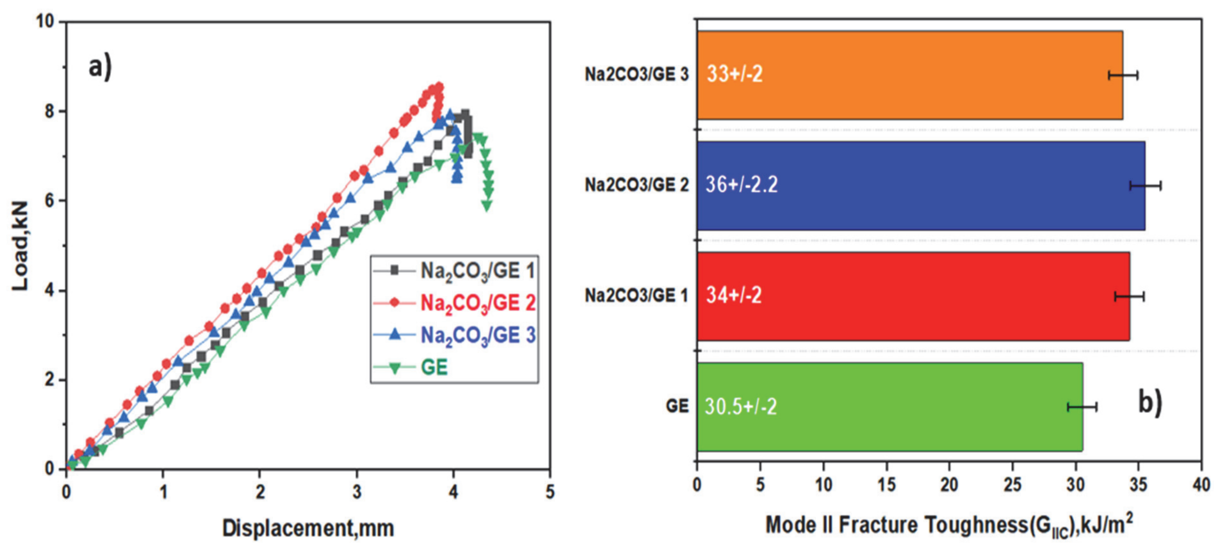

Figure 12: Mode II test of Na_2CO_3 modified matrix-Glass Fiber and Plain Glass Epoxy Composites a) Load versus Displacement b) Mode II Interlaminar Fracture Toughness (G_{IIC}).

Fig. 12b displays the mode II interlaminar fracture toughness values for the Na_2CO_3-Glass fiber composites under investigation. The composites comprised of epoxy matrix modified with different concentrations of Na_2CO_3 exhibited marginally higher fracture toughness in comparison with plain Glass epoxy composites. The composites comprised of 1wt% Na_2CO_3 in Glass fiber epoxy composites exhibited higher fracture toughness under mode II loading as compared to other concentrations such as 0.5wt% and 2wt% Na_2CO_3 in Glass fiber epoxy composites. The plain Glass epoxy composites showed lower fracture toughness under mode II loading. The Tab. 5 shows the details of number of specimens tested for mode II fracture toughness with standard deviation, average and co-efficient of variance.

SL.No	Laminate	No.of Specimens Tested	Average value of Mode II fracture toughness	Standard deviation of Mode II fracture toughness	Co-efficient of variance of Mode II fracture toughness
1	GE	6	30.5	2	6.55
2	MWCNT/GE 1	6	34	2	5.88
3	MWCNT/GE 2	6	37	2.1	5.67
4	MWCNT/GE 3	6	35.5	2.1	5.91
5	Al_2O_3/GE 1	6	32.5	2.2	6.76
6	Al_2O_3/GE 2	6	36.5	2.5	6.84
7	Al_2O_3/GE 3	6	35.5	2.4	6.76
8	Na_2CO_3/GE 1	6	34	2	5.88
9	Na_2CO_3/GE 2	6	36	2.2	6.11
10	Na_2CO_3/GE 3	6	33	2	6.06

Table 5: Number of samples tested, Average, Standard deviation, Co-efficient of variance for Mode II fracture toughness (G_{IIC}).

FRACTOGRAPHY

The fractographical examination of cracked fractured surfaces of Glass fiber composites consisting of epoxy matrix modified with various fillers like MWCNTs, Al_2O_3, and Na_2CO_3 was done by SEM instrument to identify the two-phase morphology of the composites. The fiber pullout, debonding and plastic deformation of the matrix around the fiber were the prime factors considered for composite failure. The Fig. 13 depicts the fractured surfaces of composites made of epoxy matrix modified with MWCNT on the Glass fiber surface of lamina. The fracture toughness values of composites with epoxy matrix modified by MWCNT on Glass fiber exhibited higher values during crack propagation. Composites made by the addition of 1wt% MWCNT to the epoxy matrix on Glass fiber reinforcements were found to be optimum in attaining higher fracture toughness. Composites with matrix modified by the addition of 1wt% MWCNT exhibited 26% improvement in mode I interlaminar fracture toughness during crack propagation as compared to plain Glass epoxy composites. Further, the addition of MWCNTs in the epoxy matrix showed no further improvements in fracture toughness values. For mode II loading conditions the composites with modified matrix by the addition of 1wt%MWCNT showed an 18% increase in fracture toughness as compared to plain Glass epoxy composites. The fiber/matrix bonding between Glass fiber and modified epoxy matrix with MWCNTs was good. Hence, the quantity of fiber breakage was low as observed in Fig.13(b).

Fig. 14 shows the fractured surfaces of Al_2O_3- Glass fiber composites. The addition of Al_2O_3 in the epoxy matrix resulted in the enhanced, Mode I of fracture toughness values when compared to neat Glass epoxy composites. Adding 0.5wt% Al_2O_3 in the epoxy matrix resulted in increased mode I interlaminar fracture toughness by 20% during crack propagation. The delamination of Glass fiber from the matrix during crack initiation was observed in Fig. 14(a). Similarly, there was a fiber breakage which is observed during crack propagation as seen in Fig. 14(b). In addition to this, under mode II loading the composites with a modified matrix with 1wt% Al_2O_3 exhibited 16% improvement in mode II fracture toughness. In Fig. 14(c) cusps were observed in the composite fractured surfaces under mode II loading.

Fig. 15 represents the fractured surfaces of epoxy matrix Glass fiber composites modified with Na_2CO_3 fillers. The composites consisting of Na_2CO_3 in Glass epoxy composites marginally increased fracture toughness in comparison with plain Glass epoxy composites. The composites with matrix modified with the addition of 1wt% of Na_2CO_3 demonstrated a 21% increase in mode I interlaminar fracture toughness during crack propagation as compared to pristine Glass epoxy composites. The delamination of fibers is the significant failure mechanism during crack initiation which is seen in Fig. 15(a). The fiber breakage during crack propagation was also observed in Fig. 15(b). Under mode II loading, the 1wt% Na_2CO_3 modified matrix composites exhibited a 15% increase in mode II fracture toughness. Further fiber kinking was seen in Fig. 15(c) during mode II loading and adhesion between the fiber and matrix appeared to be good which can be justified by lower fiber breakage as seen in Fig. 15(c).

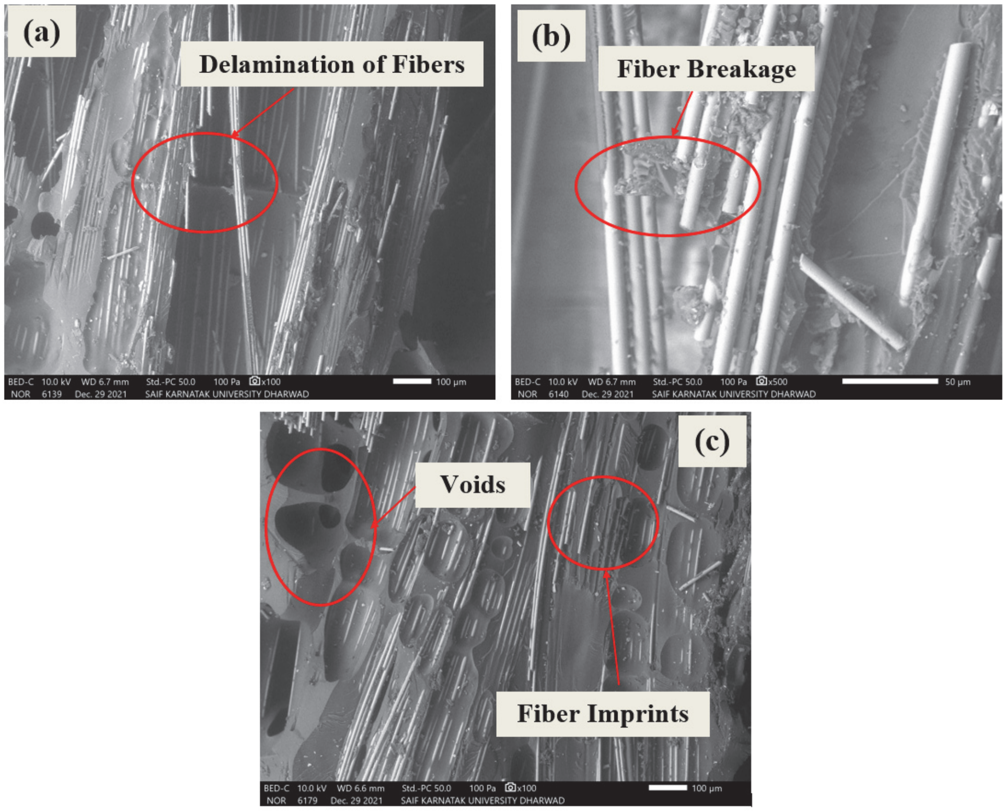

Figure 13: SEM Images of Fractured Epoxy Matrix Modified with MWCNT-Glass Fiber Composites in Mode I Loading (a) Crack Initiation (b) Crack Propagation and (c) Fractured Surface under Mode II Loading.

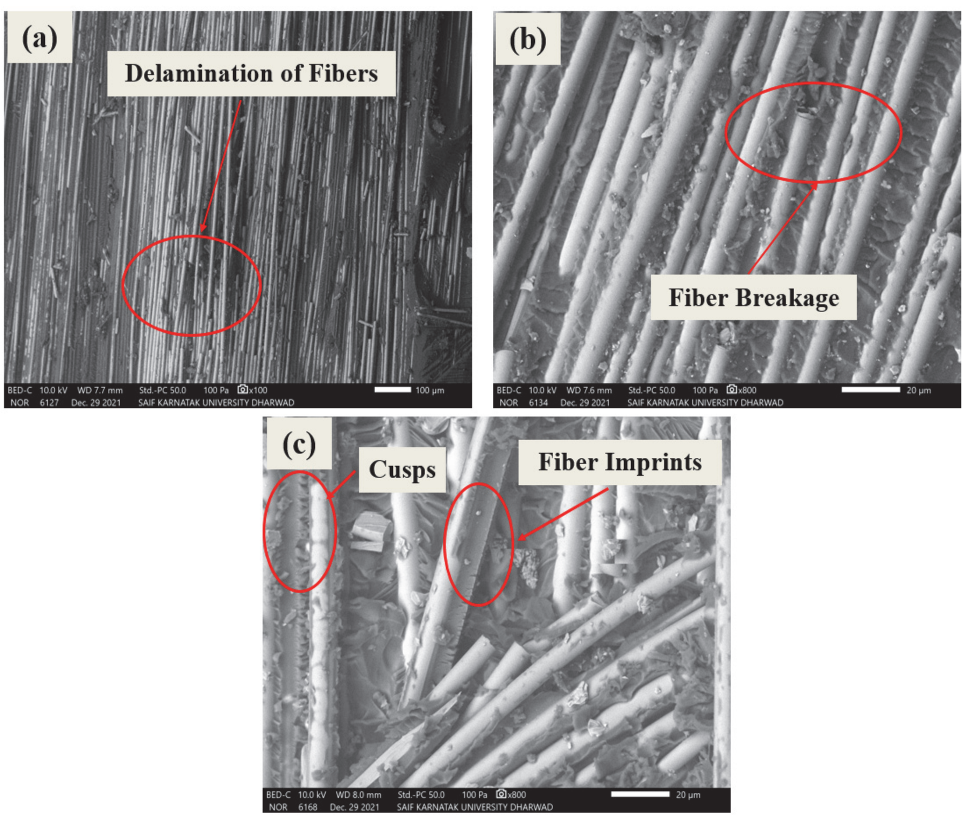

Figure 14: SEM Images of Fractured Epoxy Matrix Modified with Al_2O_3- Glass Fiber Composites in Mode I Loading (a) Crack Initiation (b) Crack Propagation (c) Fractured Surface under Mode II Loading.

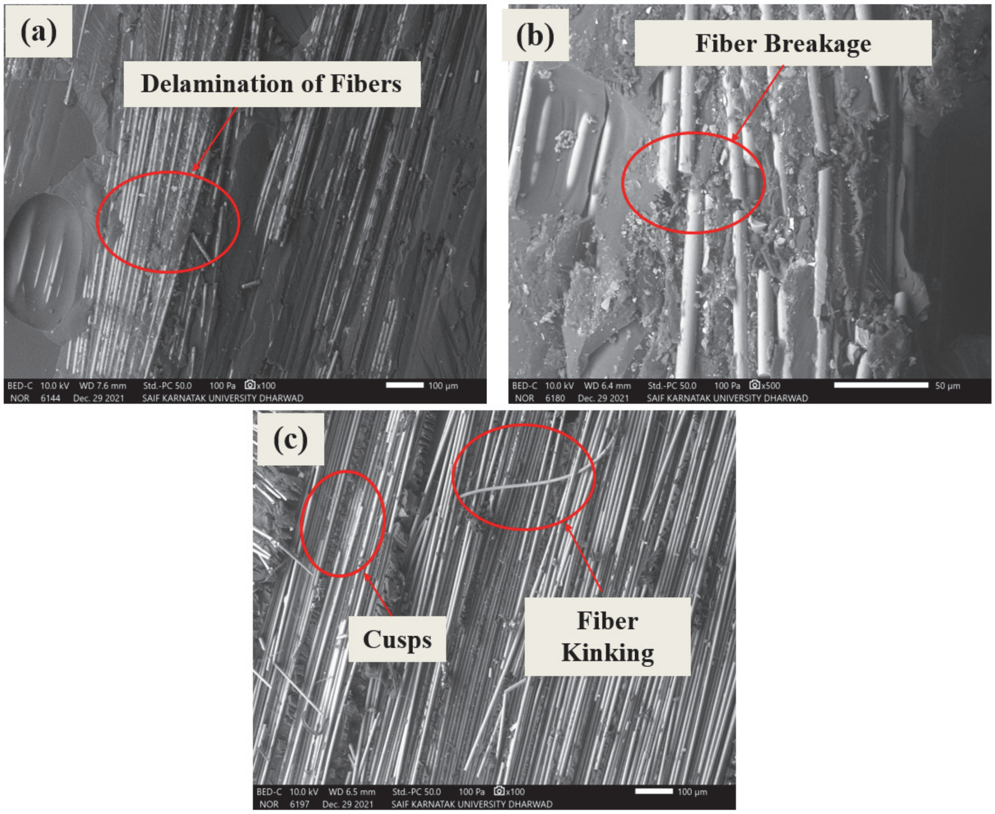

Figure 15: SEM Images of Fractured Epoxy Matrix Modified with Na_2CO_3-Glass Fiber Composites in Mode I Loading (a) Crack Initiation (b) Crack Propagation and (c) Fractured Surface under Mode II Loading.

CONCLUSIONS

The matrix modification was done effectively by adding Nano fillers like Multi-Walled Carbon Nano Tubes (MWCNTs) and micro fillers like Aluminium Oxide (Al_2O_3) and Sodium Carbonate (Na_2CO_3) to the epoxy matrix. The modified epoxy matrices were used to make the composites comprised of UD Glass fiber reinforcements. The addition of these fillers in the epoxy matrix was found to be effective in increasing the out-of-plane load-bearing capacity of the composites as compared to plain Glass epoxy composites. Also, the fracture toughness enhanced in the range of 20-26% and 14-17.5% under mode I and mode II loading respectively after adding the fillers. The 1wt% MWCNT filler seems to be the optimum concentration to get enhanced fracture toughness values. Further increasing MWCNTs showed no significant improvement in fracture toughness values. For 0.5wt% Aluminium oxide (Al_2O_3) micro filler concentration in epoxy matrix gave promising in attaining the higher load carrying capacity of the composites.

The fracture toughness values under mode I and mode II loading were increased significantly by 20% and 7% respectively in composites composed of epoxy matrix modified with 0.5wt% Al_2O_3. The filler Na_2CO_3 added in epoxy glass fiber composites was also favorable in increasing the composites' out-of-plane load-bearing capacities. The optimal concentration of 1wt% Na_2CO_3 in glass fiber epoxy composite is found to have a higher load-bearing capability as compared to other concentrations such as 0.5wt% and 2wt% addition of Na_2CO_3 in glass fiber epoxy composites.

The composites consisting of 1wt% Na_2CO_3 in Glass epoxy composites showed higher fracture toughness during crack propagation under mode I loading whereas, composites consisting of 2wt% Na_2CO_3 in Glass epoxy composites exhibited higher toughness during crack initiation under mode I loading. The composites with 1wt% Na_2CO_3 exhibited higher load-bearing capacity and higher fracture toughness values under mode II loading.

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