Effect of Volume Fraction of Fibre on the Mechanical Properties of Nanoclay Reinforced E-Glass-Epoxy Composites

K. Krushnamurty, D. Rasmitha, I. Srikanth, K. Ramji, Ch. Subrahmanyam

Abstract—E-glass-epoxy laminated composites having different fiber volume fractions (40, 50, 60 and 70) were fabricated with and without the addition of nanoclay. Flexural strength and tensile strength of the composite laminates were determined. It was observed that, with increasing the fiber volume fraction (V_f) of fiber from 40 to 60, the ability of nanoclay to enhance the tensile and flexural strength of E-glass-epoxy composites decreases significantly. At $70V_f$, the tensile and flexural strength of the nanoclay reinforced E-glass-epoxy were found to be lowest when compared to the E-glass-epoxy composite made without the addition of nanoclay. Based on the obtained data and microstructure of the tested samples, plausible mechanism for the observed trends has been proposed. The enhanced mechanical properties for nanoclay reinforced E-glass-epoxy composites for 40-60 V_f, due to higher interface toughness coupled with strong interfilament bonding may have ensured the homogeneous load distribution across all the glass fibers. Results in the decrease in mechanical properties at $70V_f$ may be due to the inability of the matrix to bind the nanoclay and glass-fibers.

Keywords—E-glass-epoxy composite laminates, fiber volume fraction, e-glass fiber, mechanical properties, delamination.

I. INTRODUCTION

THERE is a great interest to use low-weight and high-performance composites to replace traditional metals due to high specific strength and significant weight reduction. Among various fiber reinforced composites, due to the cost effectiveness, glass fiber reinforced epoxy composites (E-glass–epoxy) are widely used in various applications, like armor tanks, automobile bodies and boat hulls [1], [2]. However, in each of these applications, a particular property like impact resistance, tensile strength or flexural plays a critical role. There is an urge to enhance these properties of E-glass-epoxy composites, to make them more effective for various applications. Over the years, many researchers are exploring the possibility of employing various additives like nanoclay, carbon nanofibere (CNFs), carbon nanotubes (CNTs) etc. to enhance the tensile,

flexural strength and impact resistance of the E-glass-epoxy composites [3]-[7]. Among all these, organically modified (octadecyl amine) montimorillonite nanoclay has better compatibility with both E-glass fabric and epoxy matrix. Especially, interface compatibility of the nanoclay with the epoxy matrix arises from the amino groups present on their surface [8]. It is also reported that, amino functional groups present on carbon nanomaterials (CNFs or CNTs) be able to react with epoxy functional groups present in the matrix and therefore form a good bond with the epoxy. matrix [9]. In common, enhancement in the tensile and flexural properties of E-glass-epoxy composites due to the addition of nanoclay is ascribed to the increased crosslink density, toughness of matrix and strong fibre–matrix interface [9], [10]. Though, fiber volume fraction (V_f) of the composite has a strong effect on the fibre–matrix interface area. Therefore, the degree of the tensile and flexural strength improvement due to the nanoclay addition in E-glass-epoxy composites depends on the V_f of the composite. For example, tensile strength improvements of fibre reinforced epoxy composites due to the addition of nanomaterials is reported up to as high as 28%, and also as low as 7% [11], [12]. So far, it has not been established that, how nanoclay addition influences the tensile and flexural strength of the GFRPs/E-glass-epoxy composite having different V_f.

The present study is aimed to understand how strengthening mechanisms of E-glass fiber in E-glass-epoxy composites changes as the fiber volume fraction of the E-glass-epoxy composite changes. In present study we vary the V_f from 40-70%, due to the composite used in the majority of the realistic applications commonly lies in this range only.

II. EXPERIMENTAL

A. Raw Materials

Epoxy resin (commercial name LY556, Huntsman Advanced Materials (India) Pvt. Limited, Mumbai) made from Bisphenol A and epichlorohydrin along with diamine based curing agent (DETDA, commercial name HY 5200, Huntsman Advanced Materials (India) Pvt. Limited, Mumbai) were used as the matrix phase. E-glass fabric with plain-woven (360 gsm, UTS of 40 GPa) was used as the main reinforcement. Nanoclay used in this study is octadecylamine modified montimorillonite (Nanomer I.30E) procured from M/s Nanocor Inc (USA).

K. Krushnamurty is with the Department of Chemistry, Indian Institute of Technology, Hyderabad 502 205, India (e-mail: krushna@iith.ac.in).
D. Rasmitha and I. K. Ramji are with the Centre for nanotechnology, A.U. College of Engineering, Visakhapatnam 530 003, India.
I. Srikanth is with the Advanced Systems Laboratory, DRDO, Hyderabad 500 058, India.
Ch. Subrahmanyam is with the Department of Chemistry, Indian Institute of Technology, Hyderabad 502 205, India (e-mail: csubbu@iith.ac.in).

World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:10, No:1, 2016

B. Method

1. Preparation of Nanoclay-Epoxy Mixture

Primarily predecided weight of nanoclay (3wt%) was directly added into the epoxy resin. Dispersion of the nanoclay was ensured by using a high speed ball milling at 1000 rpm (Insmart systems) for 60 min. The epoxy mixture was added to the hardener (24 parts of hardener to 100 parts of epoxy resin by weight) and the resulting mixture was ballmilled for 15 min.

2. Fabrication of Nanoclay-E-Glass-Epoxy Composites

For the fabrication of the nanoclay-E-glass-epoxy composites, nanoclay-epoxy resin was brush painted on E-glass fabric. These fabric layers were stacked up by hand layup method and compacted by sandwiching between two flat steel plates to get required fiber-matrix ratio. Curing was carried out for 1h at 110ºC followed by curing at 160ºC for 2h. Post curing was carried out for 3h at 180ºC, which is nearer to the peak cure temperature of the compositions under study (Fig. 1). V_f of the prepared composites was controlled by varying the number of fabric layers in the specified thickness. Different composite laminates that were fabricated with varying V_f are as shown in Table I. Composites fabricated without nanoclay addition are denoted as BGE (Blank–glass-epoxy) while the composites fabricated with the addition of nanoclay are denoted as NGE (nanoclay reinforced glass-epoxy). The numbers prefixing the BGE/NGE indicate the fibre volume fraction. For example, 60 BGE indicates blank glass-epoxy composite having $60v_f$, 60NGE indicates nanoclay reinforced glass-epoxy composite having 60Vf.
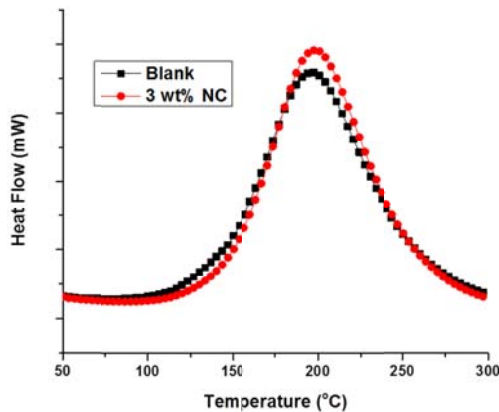

Fig. 1 Heat evolution during the curing

V_f of the composites was determined as per ASTM D3171, by using acid digestion test.

$$\text{Volume fraction } (V_f) = \frac{Wf \times \delta c}{Wc \times \delta f} X100 \qquad (1)$$

where, Wc = weight of composite, Wf = weight of fiber, δc = density of composite, δf = density of fiber (1.78).

Tensile strength and flexural strength of the fabricated composites were determined as per ASTM D638 and ASTM

D790 (three-point bending test), on the universal testing machine (United 50KN, USA). Minimum six numbers of samples were tested from each composite for each of the measured property and the results obtained are shown in table 2. Characteristic sample dimensions for tensile strength and flexural strength are 200 mm x 10 mm x 3 mm and 60 mm x 10 mm x 3 mm. Fracture modes and microstructure of the failed tested samples were analyzed with scanning electron microscopy (Supra 40, ZEISS).

TABLE I
DETAILS OF THE FABRICATED E-GLASS-EPOXY COMPOSITES

Sample description	Sample code	Wt% nanoclay	V_f (±1)
Blank Glass-epoxy	40 BGE	0	40
nanoclay Glass-epoxy	40 NGE	3	40
Blank Glass-epoxy	50 BGE	0	50
Nanoclay-Glass-epoxy	50 NGE	3	50
Blank Glass-epoxy	60 BGE	0	60
nanoclay Glass-epoxy	60 NGE	3	60
Blank Glass-epoxy	70 BGE	0	70
nanoclay Glass-epoxy	70 NGE	3	70

III. RESULTS AND DISCUSSION

A. Tensile Strength

Noticeable enhancement in tensile strength due to the nanoclay addition was observed for E-glass-epoxy composites (Table II). Reasons for this can be understood from the fracture modes of the E-glass-epoxy composites (BGE and NGE samples) failed under tensile loads. BGE samples showing interfilament debonding and NGE samples showing good interfilament bonding between the fiber and matrix. Though, the degree of the interfilament bonding and debonding varied considerably from BGE to NGE at different V_f.
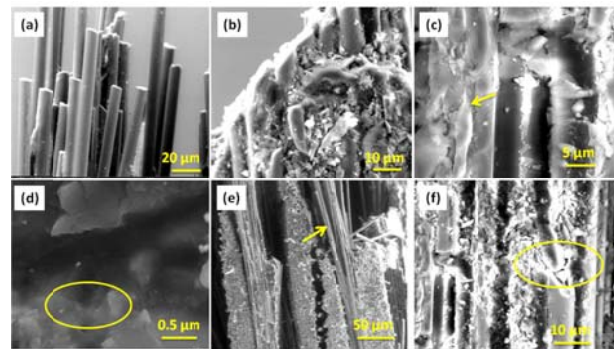

Fig. 2 SEM images of E-glass epoxy composites after failure under tensile load (a) 40 BGE showing more fiber matrix debonding with smooth surfaces. (b) 40 NGE showing strong interfilament bonding between the fiber and matrix (c) 60 NGE showing better resin toughening and strong interfacial bonding between E-glass fiber and matrix (d) magnified image of Fig. 2 (c) showing good dispersion of nanoclay (3 wt% nanoclay on glass-fabric) (e) 70 NGE showing significant interply debonding (f) magnified image of Fig. 2 (e) showing micro cracks at the interface and more fiber damage (encircled zone)

World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:10, No:1, 2016

Enhanced matrix toughness due to the nanoclay is evidenced from the strong inter-filament bonding and rough fracture surfaces of the nanoclay-glass-epoxy after the tensile failure as compared to blank (Fig. 2 (b))

TABLE II
FLEXURAL AND TENSILE PROPERTIES OF THE E-GLASS-EPOXY COMPOSITES AT DIFFERENT VOLUME FRACTIONS

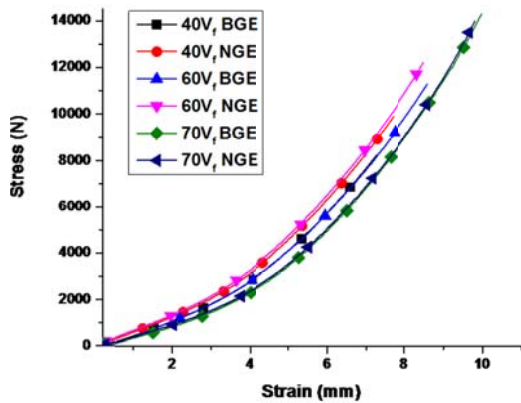| V_f	Flexural (MPa)		% imp	Tensile (Mpa)		% imp
	BGE	NGE		BGE	NGE	
40	368	441	20	274	329	21
50	451	513	14	343	388	13
60	536	578	8	417	445	7
70	603	587	-3	478	469	-2

Fig. 3 Stress-Strain curves of E-glass-epoxy composites failed under tensile load

From SEM images it is observed that the fracture modes of the BGE were soft with full interfilament debonding (Fig. 2 (a)), whereas NGE have shown good interfilament bonding (Fig. 2 (b)). SEM images also demonstrate that there is considerable toughening at the fibre–matrix interface as inferred from the rough surfaces that were observed on the fractured surfaces (Figs. 2 (b) and (c)). This could be due to the fact that, amino functional groups present on the surface of the nanoclay, can contribute in the cross linking reaction with the epoxy functional groups present in epoxy matrix resulting in improved interface crosslink density [9]. The improved crosslink density at the interface results in an improved interface toughness [10]. Therefore, due to the strong interfilament bond strengthening by nanoclay in NGE is shown in Fig. 2 (c) and schematic of poor interfilament bonding in BGE is shown in Fig. 2 (a). Enhanced interface toughness due to nanoclay, can involve more number of reinforcing filaments/fibres throughout tensile failure of the laminate [13]. Hence for NGE, strong interfilament bonding together with higher interface toughness ensured homogeneous load distribution across all the glass fibres [14]. Therefore, NGE samples have shown superior tensile strength for $40V_f$ - $60V_f$ samples.

The Tensile strength of 70 NGE was observed to be lower as compared to 70 BGE. Fracture surface of the 70 NGE was observed to be mostly with interlayer debonding as shown in Fig. 2 (e). This is due to the lack of ability of the matrix to form perfect bond with both nanoclay and glass-fibres. Also 70 NGE sample showing micro cracks at the interface and more fiber damage (encircled zone), leading to early failure of fibers and premature failure of the composite.

B. Flexural Strength

Nanoclay reinforced E-glass-epoxy (NGE) composite have shown higher flexural strength as compared to their corresponding blank E-glass-epoxy (BCE) composite having same V_f (Table II). Though, as the V_f of the fibre increases, there is a noticeable downward fashion in the percentage improvements of the flexural strength for NGE against their corresponding BGE sample (having same Vf). For example, flexural strength of 40NGE is 20% higher than 40BGE, whereas it is only 8% higher for 60CCE as compared to 60BCE. The reasons for such a fashion in the flexural strength imposed by V_f can be understood from the failure modes of the E-glass-epoxy composites. 40 BGE sample has shown more of a localized failure at the loading point near the top of the layers indicative of reduced load transmittance to the bottom layers, resulted in the major formation of interlaminar cracks due to interface shearing (Fig. 4 (a)). It is well reported that, the crack propagation through the matrix-rich interface zones encounters less resistance [15]. Hence, 40BGE failed at lower strength. On the other hand, in case of 40NGE sample has shown failure of the sample involving all the glass fabric layers present across the thickness (Fig. 5 (b)) indicating enhanced ability of the matrix to transmit the load uniformly across all the layers when it is reinforced with the nanoclay. This resulted in enhanced flexural strength for the 40 NGE composites. As the V_f of the composite improved to $60V_f$, the fracture modes have distorted considerably. In case of both 60BGE and 60NGE crack propagation mode throughout flexural failure was observed to be mainly translaminar (Figs. 4 (c) and (d)). Reduced interface shearing even for BGE at higher V_f can be attributed to the fold of the woven fabrics which is a deformation arising out of weaving. Hence, at the higher V_f of E-glass-epoxy, crack propagation is taking place with break of glass fabric layers even for BGE samples. Therefore, require of nanoclay to resist the inter laminar cracks would be narrow for higher V_f E-glass-epoxy composites. Therefore, as both, 60BGE and 60NGE failed in a similar mode, a considerable enhancement in the flexural strength due to the addition of nanoclay was not observed. Insignificant enhancement in the flexural strength for 60NGE as compared to 60BGE, even at $60V_f$ can be attributed to nanoclay present in NGE, which can still offer enhanced matrix stiffness. This results in improved resistance to the glass fibre bending and thus improved flexural strength.

Addition of nanoclay to the E-glass-epoxy having $70V_f$ was found to decrease the flexural strength (Table II) by 3% as compared to 70BGE. Failure of the 70NGE concerned a mixed mode of failure with fracture of the

World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:10, No:1, 2016

glass fabric coupled with the interlaminar failure (Fig. 4 (e)). Unlike in 40NGE, where nanoclay could hold interlayer cracks, in case of 70NGE, they could not hold the interlayer crack propagation. This could be attributed to poor wetting of the glass fibres in 70NGE. When, nanoclay are present in the composite, they compete with the glass fibres in consuming the resin for wetting their surfaces. Therefore, at such higher V_f, the epoxy resin may not be enough to complete ideal wetting of glass-fibres [16].
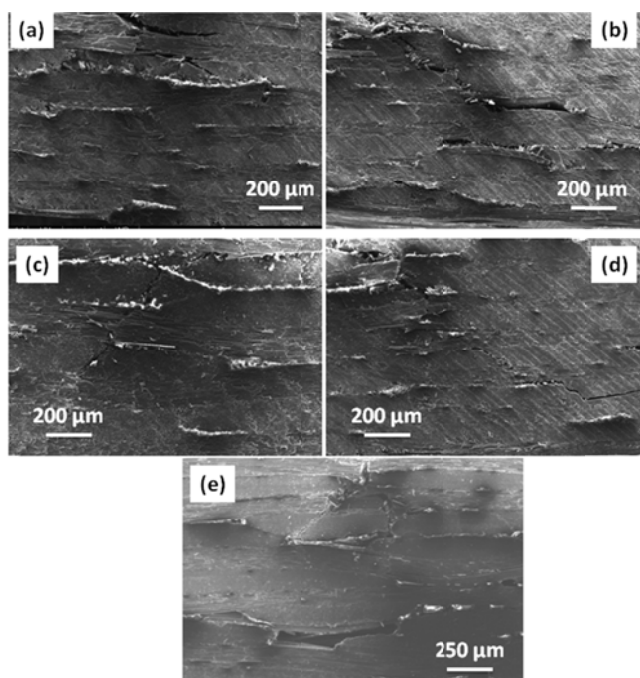

Fig. 4 E-glass-epoxy composites failed under flexural loads. (**a**) 40BGE showing failure predominantly by interlaminar crack propagation, (**b**) 40NGE showing predominant failure by rupture of glass fabrics with no interlaminar crack propagation, (**c**) 60BGE and (**d**) 60NGE showing failure by rupture of glass fabric layers with minimum interlaminar crack propagation, (**e**) showing mixed mode of failure of 70NGE sample with significant interlaminar crack propagation coupled with rupture of glass fabrics

IV. CONCLUSION

1. Nanoclay can impart considerable improvement in the tensile and flexural strength of the E-glass-epoxy composites having lower fibre volume fractions (40%). Though, as the fibre volume fraction of the composite increased (~60%), strengthening mechanisms due to nanoclay addition are down their importance and hence giving only insignificant enhancement in the tensile and flexural properties.
2. Addition of nanoclay to E-glass-epoxy composite having especially high fibre volume fraction (70%, in current study) results in decrease in flexural and tensile strength due to lack of ability of the available matrix to form good bond with both nanoclay and glass-fibres.
3. Addition of nanoclay would be helpful for E-glass-epoxy composites having fibre volume fractions in the range of 40–60%, whereas beyond 60%V_f, it may degrade the flexural and tensile strength.

REFERENCES

[1] Manfredi LB, De Santis H, Vázquez A. Influence of the addition of montmorillonite to the matrix of unidirectional glass fibre/epoxy composites on their mechanical and water absorption properties. Composites Part A: Applied Science and Manufacturing 2008; 39(11): 1726-1731.
[2] Esfahani JM, Esfandeh M, Sabet AR. High-velocity impact behavior of glass fiber-reinforced polyester filled with nanoclay. Journal of Applied Polymer Science 2012; 125(S1): E583-E591.
[3] Krushnamurty K, Reddy P M K, Srikanth I, Prasad P S, Ghosal P, Subrahmanyam C. The Reinforcement Ability of Plasma-Etched Carbon Nanofibers on Mechanical Properties of C-Epoxy Composites. Plasma Processes and Polymers 2014; 11(6): 588-595.
[4] Xu Y, Hoa S V. Mechanical properties of carbon fiber reinforced epoxy/clay nanocomposites. Composites Science and Technology 2008; 68(3): 854-861.
[5] Qi B, Zhang Q X, Bannister M, Mai YW. Investigation of the mechanical properties of DGEBA-based epoxy resin with nanoclay additives. Composite structures 2006; 75(1): 514-519.
[6] Zhou Y, Pervin F, Jeelani S, Mallick P K. Improvement in mechanical properties of carbon fabric–epoxy composite using carbon nanofibers. Journal of materials processing technology 2008; 198(1): 445-453.
[7] Richardson M C, Kim J, Ho D, Snyder C R, D'souza N A, Holmes G A. Organofunctionalized montmorillonite/epoxy nanocomposites: the effect of interlayer cation distribution on mechanical properties. Polymer Composites 2011, 32(1), 67-78.
[8] Azeez A A, Rhee K Y, Park S J, Hui D. Epoxy clay nanocomposites–processing, properties and applications: a review. Composites Part B: Engineering 2013; 45(1): 308-320.
[9] Srikanth I, Kumar S, Kumar A, Ghosal P, Subrahmanyam C. Effect of amino functionalized MWCNT on the crosslink density, fracture toughness of epoxy and mechanical properties of carbon–epoxy composites. Composites Part A: Applied Science and Manufacturing 2012; 43(11): 2083-2086.
[10] K. Krushnamurty, I. Srikanth, B. Rangababu, S. K. Majee, R. Bauri, Ch. Subrahmanyam Effect of nanoclay on the toughness of epoxy and mechanical, impact properties of E-glass-epoxy composites. Adv. Mater. Lett. DOI: 10.5185/amlett.2015.5817
[11] Zhou Y, Pervin F, Jeelani S, Mallick PK. Improvement in mechanical properties of carbon fabric–epoxy composite using carbon nanofibers. Journal of materials processing technology. 2008 Mar 3;198(1):445-53.
[12] Iwahori Y, Ishiwata S, Sumizawa T, Ishikawa T. Mechanical properties improvements in two-phase and three-phase composites using carbon nano-fiber dispersed resin. Composites Part A: Applied Science and Manufacturing. 2005 Oct 31;36(10):1430-9.
[13] Tsantzalis S, Karapappas P, Vavouliotis A, Tsotra P, Kostopoulos V, Tanimoto T, Friedrich K. On the improvement of toughness of CFRPs with resin doped with CNF and PZT particles. Composites Part A: Applied Science and Manufacturing. 2007 Apr 30;38(4):1159-62.
[14] Karapappas P, Vavouliotis A, Tsotra P, Kostopoulos V, Palpetis A. Enhanced fracture properties of carbon reinforced composites by the addition of multi-wall carbon nanotubes. Journal of Composite Materials. 2009 Jan 29.
[15] Wichmann MH, Sumfleth J, Gojny FH, Quaresimin M, Fiedler B, Schulte K. Glass-fibre-reinforced composites with enhanced mechanical and electrical properties–benefits and limitations of a nanoparticle modified matrix. Engineering Fracture Mechanics. 2006 Nov 30;73(16):2346-59.
[16] Srikanth I, Kumar S, Singh V, Rangababu B, Ghosal P, Subrahmanyam C. Effect of carbon nanofibre addition on the mechanical properties of different V f carbon-epoxy composites. Bulletin of Materials Science. 2015 Apr 1;38(2):309-17.