Manufacturing and Characterization of Epoxy Matrix Hybrid Nanocomposite

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Abstract. Advanced recent materials such as continuous reinforced polymer matrix composites provide important enhancements to a variety of structures (especially resistance to breakage), compared to their bulk, monolithic counterparts. In this research work, flexural strength and tensile or mode I fracture resistance of the hybrid polymer composite, and a detailed investigation on the processing (using Hand Lay Up method) of reinforced, unidirectional hybrid composite has been extensively discussed. Microstructural studies have also been carried out to know the material fracture behaviour and its effect on the orientation of notch. The obtained results are critically examined and discussed elaborately to find: (i) orientation of fiber and its effect on the anisotropy in flexural strength and fracture behaviour, (ii) effect of notch root radii on the conditional fracture toughness and concurrence to linear elastic controlled fracture, and (iii) the nature of J integral resistance curves. The effectiveness of the conventionally used hand layup technique is amply demonstrated by the achievement of significantly higher strength (flexural) values and also, a very importantly combination of strength and fracture resistance; qualifying the composite thus manufactured to declare this hybrid composite as a novel composite material with high specific strength when compared with other polymer matrix composite (PMCs) for a wide group of aeronautical and aerospace structural applications.

1. Introduction
In recent years, material scientists and engineers have focused a huge research activity on the development of newer composite materials with enhanced fracture resistance [1-2]. Among several effective means of enhancing the fracture resistance, material design based on continuous fiber reinforcements resulted as the most successful and effective [3-4]. Several structural materials and components have since been devised, developed and fabricated successfully using this approach of fiber reinforcements. Among many competing polymer matrix composites, carbon fiber (Cf) and amino multi walled carbon nanotubes (AMWCNTs) reinforced epoxy matrix composite results various enhanced properties essentially required for several structural and technological applications [5]. The properties of epoxy matrix are very much enhanced with the introduction of Cf and CNTs as reinforcement so that the composite materials can be accepted for certain critical structural applications because the unique mechanical and thermal properties of CNTs which is having the ability to have a greater surface area to volume (aspect) ratio along with remarkable stiffness and strength [6-7]. One possible ways of obtaining enhanced mechanical properties of the rein matrix is by
adopting unidirectional network of continuous carbon fibers as reinforcement within the epoxy matrix. The second reinforcement (AMWCNTs) is introduced within the epoxy matrix. Such a design methodology was seen to enhance the overall strength and fracture energy of the composite greater as compared to bulk matrix. Flexural strength and fracture toughness are the commonly used mechanical properties in evaluating the advanced composites. Several studies have been carried out in the last twenty years on the toughening of continuous fiber reinforced composites [8-9]. However till date very limited studies have been reported on manufacturing by hand layup technique, flexural strength and fracture behaviour studies reported so far for the C_f and AMWCNTs reinforced hybrid polymer composites.

2. Synthesis of Material
Carbon fibers (unidirectional winding) are reinforced in a polymer thermosetting matrix of DGEBA based epoxy and CNTs. Epoxy is a cross-linked thermosetting polymer commonly used as a polymer matrix material for recent advanced polymer composites given its properties like superior stiffness, dimensional stability, chemical resistance and specific strength [10-11]. The present hybrid polymer nanocomposite is fabricated by the technique of traditional hand lay-up method. The entire synthesis process comprises of three important sequential steps as described below.

I: Epoxy mixture consists DGEBA based epoxy resin, 0.5 wt.% of CNTs and proportional amount of DETDA hardener. Planetary ball mill with Al₂O₃ balls is used for the preparation of epoxy resin mixture which is operated for mixing purpose about 180 minutes at 250 rpm. Thin films of epoxy resin mixture are examined by using microscope to ensure the distribution of AMWCNTs in the epoxy mixture. The higher surface area to volume ratio of AMWCNTs results agglomeration in the mixture. Recent research studies shown that all the mechanical properties were enhanced by adding multi walled CNTs to epoxy resin. The optimum percentage of nanotubes is 0.5 wt.%. The existence of voids created throughout blending the epoxy/AMWCNTs suspension with the DETDA hardener, likewise diminishes the properties especially mechanical properties. In reality, by including more AMWCNTs the thickness of the blend builds which makes degassing of combination more troublesome [12].

II: The hand layup strategy is used for the fabrication of the present nanocomposite. A heat treated steel plate is (200×150mm) used as base plate for carbon fibers winding. The hardened steel plate is secured with a cover of TEFLON in an attempt for the simple expulsion of the segment after completion of densification of fibers and furthermore to forestall the carbon fibers densification along
the heat treated steel plate. A covering of this blend is applied to TEFON layer on the steel plate. Unidirectional winding of carbon fibers is done on the layer of TEFON.

i) The carbon fibers are unidirectionally wound over the steel plate with the overlapping carbon fiber layers and ensured no air holes between the layers. ii) After each and every carbon layer covering, an intensive layer of epoxy mixture resin is introduced on to it by using a regular paint brush. The technique is continued till the optimum 3.4 mm thickness (important to accomplish plane-strain condition). Extra resin is applied any place needed to cover the carbon fiber winding. Manually hand pressure is utilized to ensure the epoxy resin mixture completely wets all wounded layers, and voids (if any) are taken out. The extra epoxy resin mixture is removed from the carbon fibers and the by applying tension on heat treated steel plate and fixed with screws. The entire setup is kept for twelve hours to permit the extra resin to empty out totally.

III: The fabrication of composite is finished up by the heat treatment or curing cycle given in three stages which is given in fig 1. The resulted composite material got had 3.4 mm thickness of, and 1.56g/cm³ density, with 79% fiber volume fraction. The alignment of carbon filaments was examined by utilizing 20 kV SEM apparatus. The unidirectional carbon strands were seen to be uninterrupted all through the resin mixture, with higher fiber volume fraction. The approximate diameter of the carbon fiber was 6µm. Filaments were truly discernable and were not discovered diffused into each other despite three different heat treatment cycles. No aggravation or breakage among filaments was seen until and up to final manufacturing stage.

![SEM micrographs of hybrid composite](image)

3. Flexural Strength Properties

Test samples of rectangular area of cross section (with estimated width of 10mm and thickness of 3.4mm and span/range length of 54mm were utilized to decide the flexural properties of this hybrid nanocomposite. Flexural tests were evaluated using Instron UTM test framework utilizing load cell of ±50 kN and at 2mm/min cross head speed. All the tests were carried out under 3-point loading. The change in the test specimen displacement and the load values are powerfully recorded so as to get flexural stress-strain information, which was utilized to decide the flexural behavior of the present hybrid composite. This technique followed as determined by the D790 ASTM standard. The material has phenomenal flexural strength value in the longitudinal fibers orientation with an estimated average of 1084MPa. This is due to the existence of the carbon light weighted filaments and furthermore the particulate support the AMWCNTs. The load versus deflection is clearly given in figure 3 [13]. In transverse orientation of fibers, it is demonstrating that the value of flexural strength is considerably less value when compared with the longitudinal fibers orientation with an average estimation of 38MPa. The participation of fibers is largely neglected because fibers are oriented parallel to the crack
propagation. The modes of cracking are to a great extent sufficiently straight to legitimize this announcement. The matrix or resin mixture bears whole load and most extreme load estimates showed here is the applied load absorbed by the resin mixture alone. Maybe, the flexural values would much more low without CNT in the epoxy combination, which impede the crack extension.

Figure 3. Load-displacement data for Flexural test in longitudinal fibers orientation

4. Fracture Toughness Properties

The test method includes 3-point pin loading of pre cracked test specimens. Load-deflection information of the composite is recorded from UTM. Single edge notch bend (SENB) test samples of thickness 3.4mm, width 10mm and 40mm span/range length were utilized. The fracture behaviour was assessed in two different notch directions, in particular (i) longitudinal direction, where the pre crack is opposite to the direction of fibers and (ii) transverse direction, in which pre crack is corresponding to the direction of filaments or fibers. Notches of varied length (length to width proportion: 0.3 – 0.6) were presented utilizing diamond wafer blades, specially fixed on Isomet standard cutting machine. Likewise, examples with varied notch root radii, ρ (120μm to 828μm) were also tried so as to decide the basic notch root radius of the composite. The values of ρ were controlled by profile projector (Model: Delta TM 35 X-Y). Among these, examples with crack lengths as per D5045-99 ASTM standard (0.45 - 0.55 occasions the example width) were just considered for the assurance of $K_{IC}$ estimation. The fracture energy decided from load vs displacement information were utilized to decide the $J_{IC}$ (elastic-plastic fracture toughness), and $J_c$ (total fracture energy release rate) [14]. The load-deflection plot is used to calculate the various values of fracture behaviour, and the obtained results are reported and discussed in following sub sections.

4.1. $K_{IC}$ (Plane-Strain Fracture Toughness)

The $K_{IC}$ of $C_I$ and AMWCNTs dual reinforced, hybrid composite was assessed in the longitudinal direction and transverse directions as indicated by the ASTM standard D5045-99. The $C_I$ and AMWCNTs dual reinforced, hybrid composite displays higher $K_{IC}$ in the longitudinal direction when contrasted with the transverse direction of fibers. An average estimation of 20.63 MPa√m acquired in longitudinal direction is almost twenty magnitudes greater than the obtained value in the transverse direction ($K_{IC}= 0.862$ MPa√m) [14].
4.2. Effect of $\rho$ on $K_{IQ}$

The $K_{IQ}$ (conditional or apparent fracture toughness) value depends on the orientation of fibers. It tends to be seen that the failure mechanism is fundamentally same in both the directions. This critical notch root radius ($\rho_o$) is approximately 270 $\mu$m when the fibers are oriented longitudinally and 390 $\mu$m in the event of transverse [15-17].

4.3. $J_{IC}$ (Elastic Plastic Fracture Toughness)

The evaluation procedure proposed by Landes and Begley [18] and the $J_{IC}$ calculation carried out as per the ASTM standard E-813. The $J_{IC}$ is determined as 46.6 kJ/mm$^2$ in longitudinal direction of the hybrid composite, which is roughly hundred magnitudes greater than the value of $J_{IC}$ in the transverse direction (0.34 kJ/mm$^2$). The result shows that the variation in $J_{IC}$ in two different test directions enhances radically when the non linear trend in load-deflection plots (sign of fracture including relative movement between fibers of carbon and epoxy mixture) is considered [14].

5. Conclusions

The hybrid composite was effectively fabricated by hand layup method. The hybrid or dual reinforced composite has possess higher flexural strength, $K_{IC}$ and $J_{IC}$ in the longitudinal direction, when contrasted with the transverse direction. The change of $J_{IC}$ with the normalized displacement ($\delta/\delta_c$) is significantly rising in both the orientation of fibers i.e., longitudinal and transverse directions.

References

[1] Kepple K L, Sanborn G P, Lacasse P A, K.M. Gruenberg, and Ready W J 2008 Carbon 46 2026
[2] Sang-Bok L, OyoungCh, Wonoh L, Jin-Woo Y, Byung-Sun K, Joon-Hyung B, Myung-Keun Y, Hao Fong, Erik T. Thostenson and Tsu-Wei C 2011 Compos. Part A Appl. Sci. Manuf. 42 337
[3] Evans A G 1990 J. Am. Ceram. Soc. 73 187
[4] Eswara Prasad N, Sweety Kumari, Kamat S.V., Vijayakumar M and Malakondaiah G 2004 Eng. Fract. Mech. 71 2589
[5] Godara A, Mezzo L, Luizi F, Warrier A, S V Lomov, Van Vuure A W, Gorbatikh L, P. Moldenaers P and Verpoest I 2009 Carbon. 47 2914
[6] Zdenko Spitalsky, Dimitrios Tasis, Konstantinos Papagelis and Costas Galiotis 2010 Progress in Polym. Sci. 35 357
[7] Florian H. Gojny, Malte H.G. Wichmann, Bodo Fiedler and Karl Schulte 2005 Compos Sci Technol. 65 2300
[8] Bernd Wetzel, Patrick Rosso, Frank Haupert and Klaus Friedrich 2006 Eng. Fract. Mech. 73 2375
[9] Daniel R. Bortz, César Merino and Ignacio Martin-Gullon 2011 Compos Sci Technol. 71 31
[10] Hernandez-Perez A, AvilesF, May-Pat A, Valadez-Gonzalez A, Herrera-Franco P J and Bartolo-Perez P 2008 Compos Sci Technol. 68 1422
[11] Daniel C. Davis, Justin W. Wilkerson, Jiang Zhu and Viktor G. Hadjiev 2011 Compos Sci Technol. 71 1089
[12] Ayatollahi M R, ShadlouS and Shokrieh M M 2011 Mater. Des. 32 2115
[13] Chandra Shekar K, Sai Priya M, Naveen Kumar M, Krishna Kanth V, Subramanian P K, Anil Kumar, Anjaneeya Prasad B and Eswara Prasad N 2014 Bull. Mater. Sci. 37 597
[14] Chandra Shekar K, Krishna Kanth V, Sai Priya M, Naveen Kumar M, Subramanian P K, Anil Kumar, Anjaneya Prasad B and Eswara Prasad N 2015 Int. J. Mater. Prod. Technol. 51 1
[15] Chandra Shekar K, Naveen Kumar M, Sai Priya M, Krishna Kanth N, Subramanian P K, Anil Kumar, Anjaneya Prasad B and Eswara Prasad N 2014 Trans. Indian Inst. Met. 67 33
[16] Chandra Shekar K, Anjaneya Prasad B and Eswara Prasad N 2016 Trans. Indian Inst. Met. 69 1069
[17] Chandra Shekar K, Anjaneya Prasad B, and Eswara Prasad N 2016 Sadhana 41 1443
[18] Landes J D and Begley J A 1974 Test results from J-integral studies: An attempt to establish a J_int test procedure. In: Fracture analysis, ASTM STP No. 560 170