Advanced Composite Materials

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tacm20

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Version of record first published: 02 Apr 2012.

Adv. Composite Mater., Vol. 9, No. 2, pp. 77–88 (2000)

Impact damage area and interlaminar toughness of modified FRP laminates

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Received 18 January 1999; accepted 24 August 1999

Abstract—Glass fibre reinforced epoxy laminates are improved to enhance their impact tolerance capabilities by modifying the epoxy resin with the addition of CTBN 1300X8 and triphenylphosphine under a controlled atmosphere of inert nitrogen gas. When impacted by a steel projectile, the damage area of a modified laminate is significantly smaller (35–55%) than that of an unmodified laminate for the same impact energy level. The interlaminar energy release rate in Mode I (G_{Ic}) is increased by 41% as a result of the modification while the interlaminar energy release rate in Mode II (G_{IIc}) is enhanced by 47%. No significant adverse effects were found on the modulus or ultimate tensile strength. The modification of epoxy causes precipitation and separation of discrete rubber particles during curing of the epoxy which also enhances the interlaminar toughness and impact tolerance.

Keywords: Impact damage; polymer composite laminates; interlaminar toughness; CTBN; toughened laminate; impact tolerance; foreign body impact; damage area.

1. INTRODUCTION

The ability to withstand impact by foreign objects is a necessary requirement for structural material. Composite structures have many useful properties, such as light weight, high strength, low thermal expansion and resistance to environment and corrosion. But composite laminates show a poor response to impact loading, even that of quite mild intensity, such as a bird hitting the material, dropping of tools during fabrication or maintenance, hailstorms, and flying debris encountered during the take-off stage of aircraft.

The impact behaviour of a polymer composite laminate is quite different from that of metals. Foreign body impact on a metal sheet just forms a dent locally near the centre of the impact. The metal usually does not crack; it only workhardens.

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With composite laminates, however, there is widespread delamination extending all around the impact point to considerable distances, with intense damage near the centre of impact in FRP laminates. This impact-induced delamination should be restricted or controlled.

Serious efforts to estimate the damage arising from the impact of a foreign body on fibre composite laminates started in the 1970s. Husman *et al*. [1] evaluated the residual compressive strength of impacted graphite/epoxy and glass/epoxy laminates. Dorey *et al*. [2] conducted similar experiments. Caprino [3] observed that the main parameter for evaluating the extent of panel damage is the incident energy of the impacting projectile and not the velocity. Many similar tests were carried out at different places, mainly to generate data. They are listed by Sierakowski and Chaturvedi [4] for glass, graphite, Kevlar, boron and their hybrids with polymer resin or metal matrix.

Several attempts have been made to improve the impact toughness of composite laminates by various methods, such as the addition of elastomers and thermoplastics to the matrix material, fibre hybridization, interleaving of a tough material, and stitching. Siebert and Rowe [5] attempted to increase the fracture toughness of neat epoxy by modification of epoxy resin with rubber (CTBN). In this work, though the toughness of modified neat resin was increased, there was very little improvement in fracture toughness of the composite laminate. Lee *et al*. [6] used the intermittent mylar layer in between the graphite prepreg laminate to enhance fracture toughness of laminates. In their work, fracture toughness was increased but tensile strength decreased. Manzione and Gillham [7] used rubber modified epoxy and found that impact properties depend upon the phase separation of rubber and strain rate loading. Duesk *et al*. [8] used carboxyl terminated and hydroxyl terminated liquid polybutadienes (PBD) in place of conventional CTBN liquids to toughen the bulk epoxy resin. Ishai *et al*. [9] used tough adhesive (HTS-7) at high points like flaws, discontinuities, etc. to fracture toughness. Chen and Jang [10] inserted thin layers of epoxy modified with reactive rubber (CTBN) or polyurethane between two plies. The G_{Ic} values of composite laminates improved by 25–50%; G_{IIc} increased by about 50% overall, though in one set of experiments they observed 2 to 3-fold improvement.

In this work, laminates of glass fibre reinforced epoxy are modified by mixing CTBN into epoxy resin in a controlled inert atmosphere of nitrogen gas. Improvement in damage tolerance is studied by impacting laminates with a steel projectile. Also, improvements in interlaminar toughnesses, both in Mode I and Mode II, are determined. The tensile properties are investigated to study whether modification of epoxy has any significant adverse effect on these properties.

2. LAMINATE PREPARATION

The matrix of the unmodified laminates consisted of the following constituents procured from Ciba Giegy Ltd., Mumbai, India:

1. LLY 556	(Diglycidyl ether of bisphenol A, DGEBA)	100 parts by weight
2. HT 973	(BF3-MEA)	1 part by weight
3. HT 976	(DDS)	35 parts by weight
4. Coupling agent (silane)		0.5 parts by weight

Glass fibres were used as a reinforcing material. For the purpose, unidirectional fabric was procured from Fothergill Engineered Fabric Ltd. England. The area density of the fabric was 500 gm/m^2 and the fabric was made from 1200 tex roving with 40 roving per cm. The fibres of the unidirectional fabric were kept in place with a reinforcing grid (10 mm × 10 mm) which was also made of glass fibres.

To fabricate modified laminates, the above epoxy mixture was modified by adding the following materials:

1. Carboxyl-terminated butadiene acrylonitrile rubbers (CTBN) 1300X8	15 parts by weight
2. Triphenylphosphine	0.5 parts by weight

To prepare the resin solution for modified epoxy, Epoxy (LY556) and CTBN 1300X8 are mixed and heated to 90°C and triphenylphosphine (0.15 parts) is added to solution. The solution is heated to 100°C in a nitrogen atmosphere for 50 min. The temperature is further raised to 125°C and maintained for a further 20 min. Now the prereacted mixture is transferred to another pot and HT 976 is mixed at 130°C. The mixture is then cooled to 60°C and mixed with HT 973 and silane coupling reagent.

Laminates are prepared with both modified and unmodified resin and Glass fibres by hand lay up technique. The orientation of glass fibre reinforced epoxy laminate is $[0/45/-45/90/90/-45/45/0]_s$. This orientation is the same for both modified and unmodified laminate. Then curing is accomplished between the two hot platens of a hydraulic press at 145°C for 3 h under the pressure of 0.7 MPa.

3. DETERMINATION OF DAMAGE AREA TOLERANCE THROUGH IMPACT TEST

The impact test has been carried out to determine and compare the impact damage of both unmodified and modified laminates. The tests were made by impacting a steel projectile on 150 mm × 150 mm laminates. For experiments of higher impact velocity, panel of 180 mm × 180 mm were used. The laminates were impacted by a cylindrical steel projectile with a hemispherical nose. The projectile weighed 11.8 gm and was 20 mm long. The laminate was rigidly fixed on its edges with the help of clamp rings, as shown in the schematic diagram for the experimental set-up for impact testing (Fig. 1). The projectile was accelerated horizontally inside a barrel of 11 mm bore and 3.6 m length with the help of compressed nitrogen gas. The velocity of the projectile is calculated just before the impact with the help of

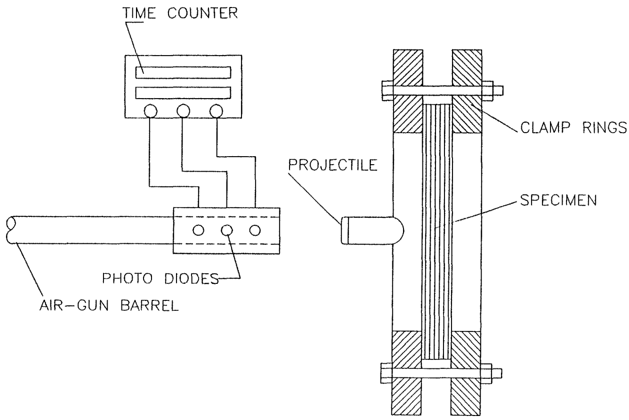

Figure 1. Schematic diagram of experimental set-up for impacting FRP laminates.

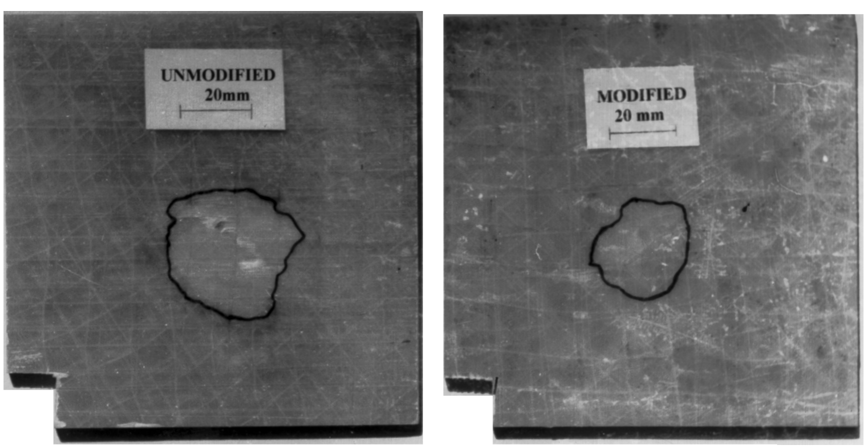

Figure 2. Impact induced damage in unmodified and modified laminates.

photodiodes. The projectile velocity was not so high that it would penetrate through the laminate.

The impact damage tolerance of a laminate is deduced using the damage area measurement technique. After the impact, the specimen is inspected by placing at against bright light and the damage area is marked. Figure 2 shows the damage area induced in unmodified and modified laminates when impacted by the projectile with almost the same velocity (55 m/s for unmodified and 54 m/s for modified). Clearly, the damage area in the modified laminate is considerably less.

The damage area of a laminate, measured with the help of a planimeter, is employed as a parameter for comparing the impact damage tolerances of unmodified and modified laminates. Figure 3 shows the damage area for unmodified and modified laminates for different impact velocity and impact energy respectively. The damage area of the modified laminate is significantly smaller (35–55%) than that of the unmodified laminate for the same impact energy level.

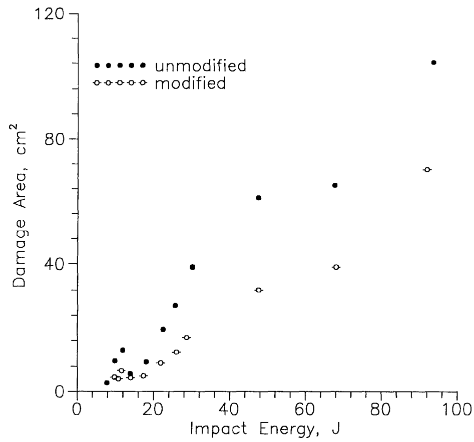

Figure 3. Damage area vs. impact energy for modified and unmodified laminates.

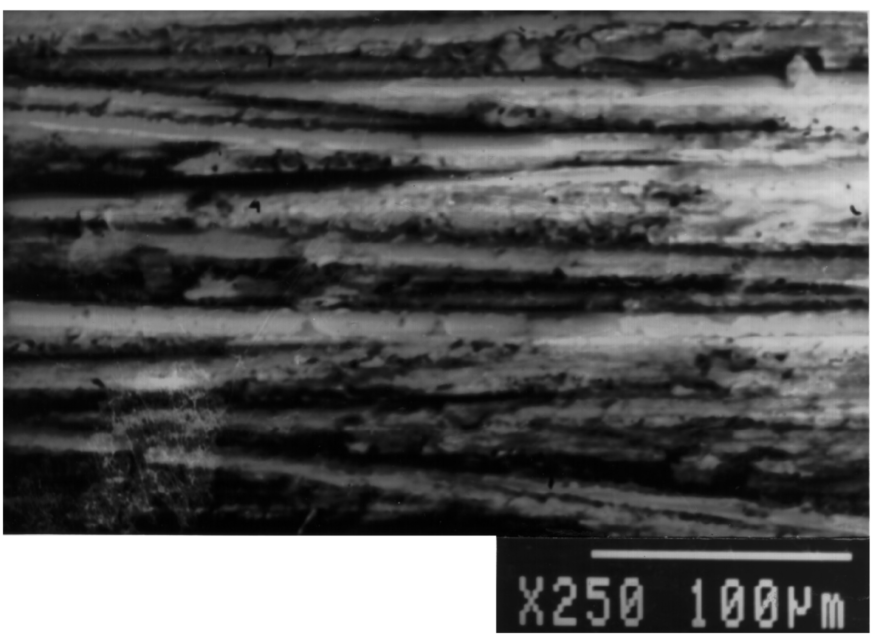

Figure 4. SEM micrograph of fractured surface of unmodified specimen at $\times 250$ magnification.

The fractured surfaces of impacted panels of unmodified resin matrix laminate are shown through scanning electron micrographs in Figs. 4 and 5. Matrix cracking and debonding between fiber and matrix is seen clearly in micrograph (Fig. 4) at $\times 250$ magnification. It shows brittle type behaviour of matrix resin failure. The chipping of matrix resin has also been observed at higher magnification ($\times 1000$). The micrograph ($\times 250$) of the fracture surface of an elastomer modified matrix resin laminate is given in Fig. 6 where matrix resin cracking and debonding (fibre/matrix) are not observed prominently. In fact, the micrograph of Fig. 7 of the fractured

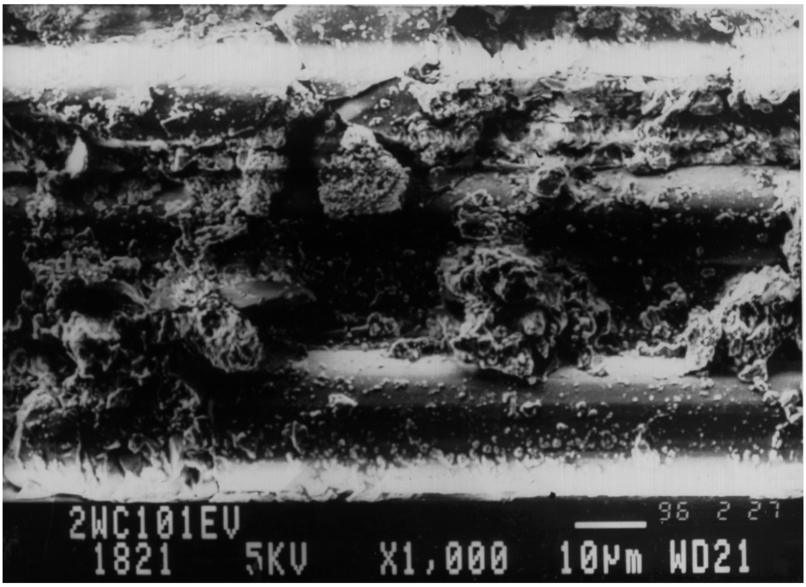

Figure 5. SEM micrograph of fractured surface of unmodified specimen at ×1000 magnification.

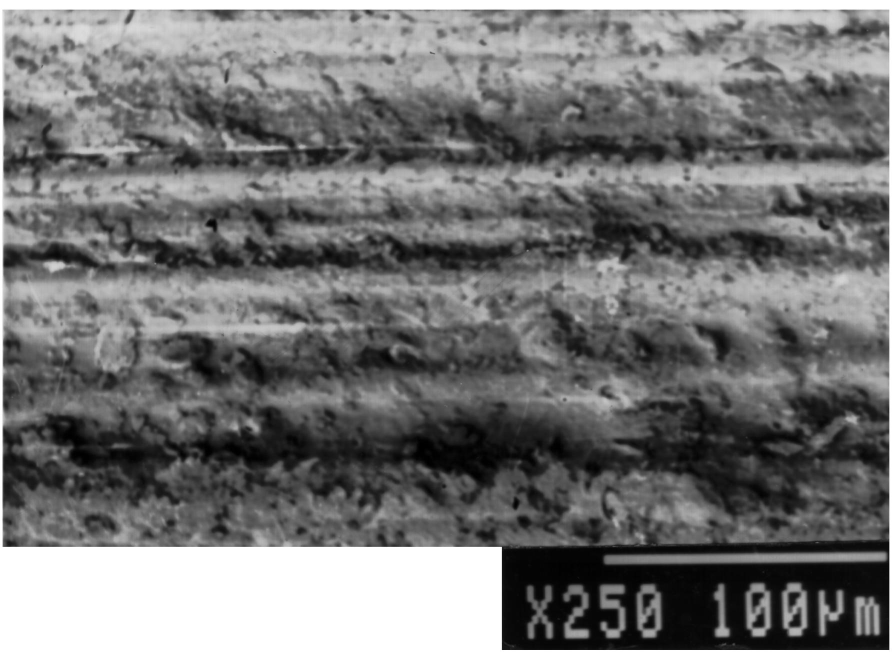

Figure 6. SEM micrograph of fractured surface of chemically modified specimen at ×250 magnification.

surface of a modified laminate is quite different from that of the unmodified laminate (Fig. 5); the fracture of modified laminate is of ductile nature.

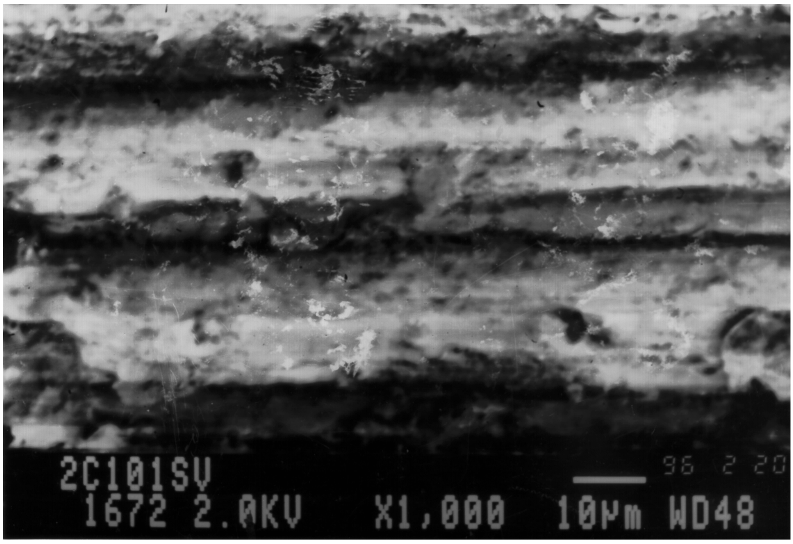

Figure 7. SEM micrograph of fractured surface of chemically modified specimen at ×1000 magnification.

It is felt that there are two possible causes of damage area reduction under impact loading of chemically modified laminate over unmodified laminate:

(i) As the interlaminar crack propagates, stretching and tearing of elastomer particles occurs. Also, rubber particles bridge the gap between the fractured surfaces and reduces the crack opening displacement making the matrix tough. These rubber particles are stretched to large strain before tearing takes place. Thus, compared to unmodified epoxy specimens, modified specimens absorbed large energy.

(ii) The matrix of modified epoxy is less brittle and deforms in a ductile manner.

4. INTERLAMINAR FRACTURE TOUGHNESS TEST

Impact of a foreign body causes extensive interlaminar failure in Mode I and Mode II. It is therefore worthwhile to investigate how interlaminar toughness is affected by modification in the epoxy. It has been found convenient to employ energy release rate as a parameter to monitor interlaminar toughness.

4.1. Interlaminar fracture toughness in Mode I (G_{Ic})

The fracture toughness in tensile mode (Mode I) is determined by employing a double cantilever beam (DCB) specimen. The specimen is loaded in an Instron test machine under displacement control mode such that the crack extends by 5–10 mm. The specimen is then unloaded completely and loaded again to determine the compliance of the extended crack. Thus, compliance is obtained for several crack lengths through loading–unloading cycles. Typical load–displacement plots obtained during testing is given in Fig. 8. From the figure it is obvious that the enclosed area between load–displacement curve for chemically modified laminate is more than that for unmodified laminate. From the data, the critical energy rate G_{Ic} is calculated with the help of the compliance method. The method is widely used and details of the procedure and data analysis are the same as those reported in published investigations [11].

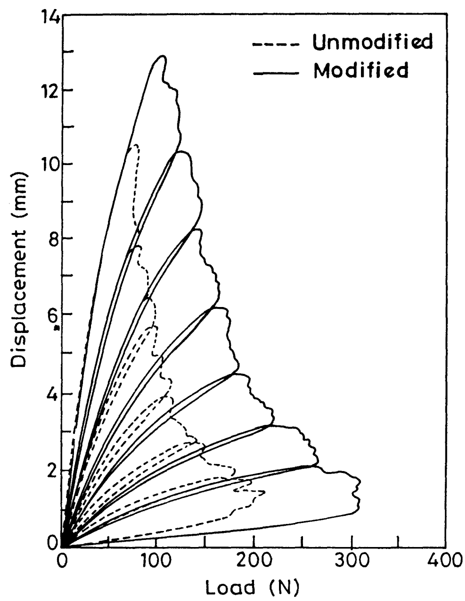

Figure 8. Typical load vs. displacement curve for test specimen of modified and unmodified laminates for determining G_{Ic}.

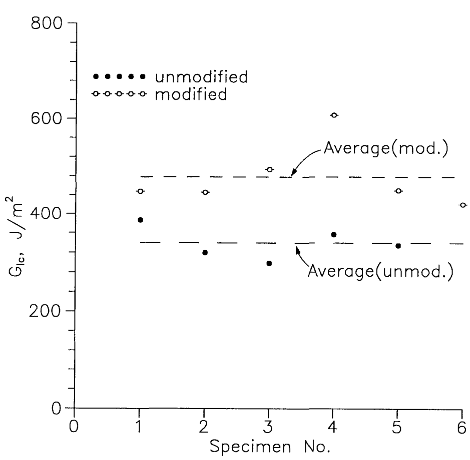

Figure 9. G_{Ic} of various specimen of modified and unmodified laminates.

Figure 9 shows that the G_{Ic} of modified laminate is about 41% more than that for unmodified laminate. This increase is consistent with the finding of Chen and Jang [10] where they reported a 50% increase in G_{Ic}. However, they conducted experiments by inserting layers of epoxy/CTBN between the plies as against mixing CTBN in the bulk epoxy material used in this study. It is felt that the production method would be much simpler if CTBN is mixed into the bulk epoxy resin.

From a microscopic point of view, the increase in G_{Ic} of a composite laminate that arises from chemical modification can be associated with a large crack tip deformation zone of the modified laminate. The large crack tip deformation permits blunting of the crack tip and this blunting decreases the local stress concentration at the tip of the crack. As a result, higher loads are required for crack initiation in a rubber modified matrix laminate. On energy considerations, the matrix cracking in a modified laminate is more ductile and thus more energy is absorbed during crack propagation.

4.2. Interlaminar toughness in Mode II (G_{IIc})

The end notched flexural (ENF) specimen has been used to determine the G_{IIc}. The specimen was loaded on a three point bending fixture in the Instron testing machine and testing was performed under displacement controlled mode. The typical load–displacement curves obtained during testing are shown in Fig. 10. The critical energy release rate in Mode II is determined from elastic beam theory with the help of data points (Carlsson *et al.* [12]).

The G_{IIc} for unmodified and modified laminates is shown in Fig. 11. From these data points it can be deduced that the G_{IIc} of rubber modified laminate is 47% more than the corresponding G_{IIc} of an unmodified laminate. It can be concluded that Mode II delamination resistance of a modified laminate also contributes significantly towards the improvement of impact induced damage of the

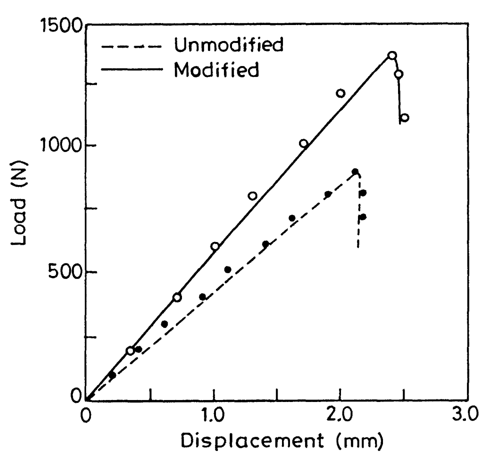

Figure 10. Typical load vs. displacement curves for test specimen of modified and unmodified laminates for determining G_{IIc}.

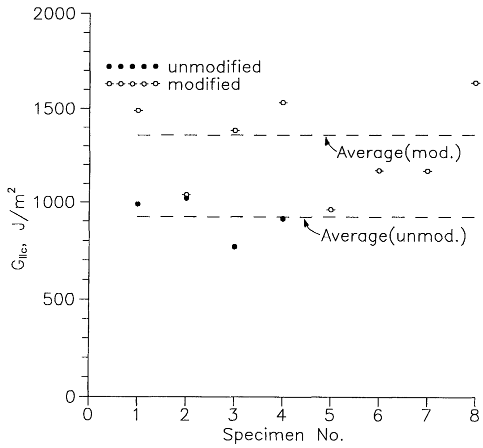

Figure 11. G_{IIc} of various specimen of modified and unmodified laminates.

modified laminate over unmodified laminate. This increase is consistent with about 50% increase of G_{IIc} in the work of Chen and Jang [10] where they interleaved epoxy/CTBN layers between prepregs.

5. TENSILE TEST

There have been several investigations reported for enhancement of fracture toughness (Manzione and Gillham [7]; Ishai *et al.*, [9]; Williams and Kousionnals [13]). But most of the developments were at the cost of the strength of the composite materials. Therefore, the concern of this work was to see whether tensile strength and modulus deteriorate or improve while trying to increase impact toughness.

To find the tensile properties of laminates, the tests were carried out in the Instron tensile testing machine under displacement controlled mode. The ultimate tensile strength and modulus of unmodified and modified laminates are given in Fig. 12 and Fig. 13 respectively. As compared to unmodified laminates, the chemically modified laminates show marginal (8.2%) deterioration in modulus. The reduction in modulus of rubber modified laminate is due to a plasticizing effect of rubber particles in the rubber epoxy solution. The ultimate strength of modified laminate shows marginal (10.8%) improvement over unmodified laminates.

6. CONCLUSIONS

Glass fibre reinforced epoxy laminates have been modified to enhance their impact tolerance capabilities. The epoxy resin was modified by adding CTBN 1300X8 and triphenylphosphine under a controlled atmosphere of inert nitrogen gas. Modified and unmodified panels were investigated to seek a possible reduction in the size of the damage area when a laminate is impacted by a steel projectile. The steel

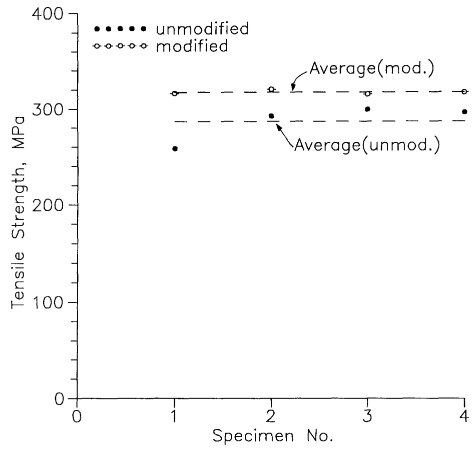

Figure 12. Ultimate tensile strength (σ_u) of various specimen of modified and unmodified laminates.

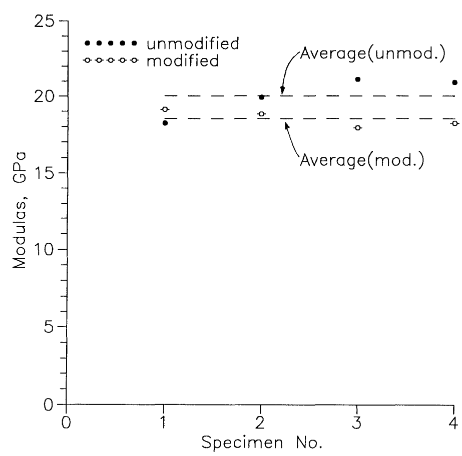

Figure 13. Tensile modulus of various specimen of modified and unmodified laminates.

projectile was accelerated in an air gun and its impact energy was accurately determined. As a result of the modification, a significant improvement in damage area reduction was observed.

The effect of modification on the interlaminar toughness was also determined. Interlaminar G_{Ic} increased by 40%, while the interlaminar G_{IIc} was enhanced by 47%. No significant adverse effect was found on modulus and ultimate tensile strength of the laminate. The increase in interlaminar toughness is consistent with the result of Chen and Jang [10] in which they interleaved epoxy/CTBN layers between plies of laminate. However, it is felt that modification of bulk epoxy with CTBN would simplify the production method. The CTBN precipitates and separates as discrete rubber particles during curing of the epoxy which also makes the epoxy tougher. It is concluded that modification in epoxy shows great potential for improving the impact induced toughness of fiber reinforced epoxy laminates.

Acknowledgements

The authors acknowledge the financial support given by the Aeronautical Research and Development Board (Structures Panel) and the supply of chemicals by Defence Materials and Stores Research and Development Establishment Kanpur. Also, authors acknowledge the assistance of Mr. R. P. Gupta in conducting the experiments.

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