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Assessment of Microstructural Integrity of Glass/Epoxy Composites at Liquid Nitrogen Temperature

Surendra Kumar M, Nirmal Chawla, Asima Priyadarsini, Itishree Mishra and B. C. Ray*
Department of Metallurgical & Materials Engineering
National Institute of Technology, Rourkela-769008, India

*Author for correspondence:
Email: drberay@gmail.com (B C Ray)

ABSTRACT

Polymer composite materials have received increased attention for applications in cryogenic environment because of their light weight and unique properties. The research presented in this paper is an effort to understand the interlam inar fracture behavior of glass/epoxy composite laminates at cryogenic conditions and at different loading rates. This study uses 3-point flexural test to qualitatively assess such behavior for 50 weight percentage of E-glass fibers reinforced epoxy composites during cryogenic and ambient conditions. The specimens were tested at a range of 2 mm/min to 500 mm/min crosshead speed to evaluate the sensitivity of mechanical response during loading at these conditions. The mechanical performances of the laminated specimens at cryogenic conditions were compared with room temperature property by using SEM photographs. Phenomenological behavior of these materials may be attributed by polymer relaxation, low-temperature hardening, matrix cracking, and misfit strain due to differential thermal coefficient of the constituent phases and also by enhanced mechanical keying factor by compressive residual stresses at cryogenic temperatures.
INTRODUCTION

Polymer composite materials have received increased attention for applications in cryogenic environment because of their light weight and unique properties [1]. In space vehicles, size may not matter, but weight does. Lifting a payload straight up to escape Earth's gravity requires a phenomenal expenditure of energy, and every ounce saved in either launch vehicle or payload weight translates to lower project costs. Next generation space missions emphasize cutting the cost of launching payloads into orbit by designing and development of Reusable Launch Vehicles (RLV) [2]. Lightweight and strong composite materials are already used in many applications, such as launch vehicle bodies and payload components. But tankage for cryogenic propellant is the major contributor to the mass of the launcher. Recently one area identified as potential source for significant weight reduction is the replacement of traditional metallic cryogenic fuel tanks with advanced polymeric matrix composite (PMC) tanks. Carbon-fiber/epoxy-resin composites have been evaluated for cryogenic tankage in RLV (reusable launch vehicle) [3]. Woven-fabric glass/epoxy laminates are used as thermal insulation, electrical insulation, structural support and permeability barrier in superconducting magnets which are essential components of most current and planned fusion devices [4]. Interface is the heart of the composite. The local response of fiber matrix interface within the composite plays an important role in determining the gross mechanical performance [5]. It provides a means of stress transfer from fiber to fiber through the matrix. The concept of two-dimensional
interface between fiber and matrix has given way to the evolution of the 3-D region mere properly termed as “interphase”. This region includes the 2-D region of contact between fiber and matrix but also incorporates a region of some finite thickness extending on the both sides of the interface. This interphase will posses unique properties different from the bulk matrix [6]. It can include impurities, unreacted polymer components, non-polymerized additives etc. The thickness and properties of this interphase have crucial impact on the composite properties [7]. The interphase not only allows load transfer between fibers through matrix but also provides a matching of chemical and thermal compatibility between the constituents. A thin rigid interphase leads to a low fracture resistance while a thick soft interface results in a better fracture resistance but a lower composite stiffness [8]. The thickness of the interphase depends on the polymer/filler system. It is generally recognized that the bond strength variation at the interface greatly affects the integrity of composite materials. The bond strength depends on quality of interfacial adhesion [9]. Residual thermal stresses develop in these materials when they are exposed to cryogenic temperatures. These stresses are the result of a difference in the CTEs (coefficients of thermal expansion) between the reinforcement and the matrix. The non-zero state of residual thermal stresses at cryogenic temperatures is the underlying cause of microcracking in composites and the microcracks could have an important influence on their performance. A very large thermal expansion mismatch can result in debonding at the fiber/matrix interface and/or a possible matrix cracking due to thermal stress [10]. Epoxy resin and E-glass fiber are reported to be loading rate sensitive. The ductility of a matrix resin may become a limiting factor at high strain rate for composite strength. Epoxy resin is more ductile than it’s composite at low strain rate [11]. The main objective of this paper to
access the mechanical behavior of cryogenically conditioned glass epoxy composite at different loading rates.

**EXPERIMENTAL**

Araldite LY-556, an unmodified epoxy resin based on Bisphenol-A and hardener (Ciba-Geig, India) HY-951, aliphatic primary amine were used with woven roving E-glass fibers treated with silane based sizing system (Saint-Gobain Vetrotex) to fabricate the laminated composites. The fiber weight percentage 50% was targeted in the laminate fabrication. They were cured for 48 hours at room temperature. The laminates were cut into short beam shear (SBS) test specimens by diamond cutter. The SBS 3-point bend tests were conducted to determine the interlaminar shear strength (ILSS) of composites. The cured specimens were exposed to liquid nitrogen environment (77K) for one hour. After the exposure one batch of samples were taken out and kept at room temperature for one hour. Another batch of samples was tested in 3-point bend test immediately after exposure to cryogenic temperature. The former samples after exposure to room temperature and the untreated as-cured composite specimens were tested in 3-point bend test at room temperature. All the mechanical flexural tests were performed at 2, 50, 100, 200 and 500 mm/min crosshead speeds. The interlaminar shear strength (ILSS) was measured as follows,

\[ \text{ILSS} = 0.75p/bt \]
Where,

‘p’ is the breaking load, ‘b’ the width, and ‘t’ the thickness of the specimen.

An Instron1195 tensile testing machine was used to perform SBS tests in accordance with ASTM D 2344-84 standard. Multiple samples were tested at each point of experiment and the average value was reported.

**RESULTS AND DISCUSSION**

The effects of different crosshead speeds on ILSS value at cryogenic temperature (♦), ambient temperature after exposure to cryogenic temperature (■) and at ambient temperature of untreated samples (▲) of glass-epoxy laminates are shown in figure 1. Figure 1 shows the variation of ILSS value of glass-epoxy composite with different loading rates at ambient temperature condition (▲). It is evident from the graph that the nature of the curve is different at above and below 50mm/min crosshead speed. The ILSS value is low for lower speed and increase with the loading rate upto 50mm/min. This lower value at lower crosshead speed may be due to higher failure strain at low strain rates so with increase in speed the ILSS increases. Here more time is available at lower crosshead loading speeds for the failure to take place, which results in more deterioration causing the reduction in the value of ILSS. With higher crosshead speeds (above 50mm/min) the curve is just the opposite. Here very less time is available for the failure to take place, it is more like an impact, so may be the matrix is unable to transfer the load properly which leads to matrix cracking (figure 2). The higher crosshead speed during test restricts and minimizes the relaxation processes at the crack tip. These stress-induced cracks may grow without blunting leading to reduced ILSS at higher loading rates [12]. Figure 1 shows the variation
of ILSS value of glass-epoxy composite with different loading rates at cryogenic condition (♦). The ILSS values are higher than the untreated samples for almost all loading rates. Here also a slight increase in the shear strength was marked with increasing in loading rate upto 50mm/min after that the shear strength decreases. The higher value of ILSS may be due to cryogenic hardening of matrix phase which leads to the improvement in shear strength at lower loading rates. Residual stresses build due to differential contraction during sudden cooling from room temperature to cryogenic temperature (77K). The cryogenic conditioning causes differential contraction and increases the resistance to debonding by mechanical keying factor [13]. When cooling from the cure temperature, differences in thermal contraction between the matrix and fiber will generate shear stresses in the resin. When a stress is applied, shear stresses greater than the shear strength of the resin is readily generated and failure of the resin phase will result. When the composite is stressed further by cooling and loading in the cold state, it is likely that there will be resin/fiber debonding. As shown in the figure 3, due differential contraction of epoxy at cryogenic temperature the epoxy will try to come out from the bulk composite. The characteristic of the interfacial adhesion is strongly influenced by the presence of residual stresses. Some of the stresses developed are relaxed by viscoelastic flow in polymer matrix [14]. It was also observed that above 50mm/min loading speed the ILSS value drops due to matrix cracking. Here the ductility of a resin matrix could become a limiting factor at high loading rate for the composite strength. The specimens tested at a cryogenic temperature are characterized by a greater order of micro-cracking and delamination (figure 4). A plastic deformation zone ahead of crack tip region may possible be formed by matrix deformation and micro cracking [15]. A weaker of interfacial bond may result in a low flexural strength of the
laminate. The deteriorated integrity can cause low strength at high loading. Figure 1 shows the variation of ILSS value of glass-epoxy composite with different loading rates at room temperature after exposing to cryogenic temperature (■). The matrix hardening which was in the cryogenic condition for which we got higher values of ILSS at lower speed may be relaxed at room temperature lowering its shear strength but higher than non-treated samples. At lower crosshead speeds the stress redistribution occurs which limit the initiation of new cracks. The high and complex nature of residual stresses at cryogenic temperature and also at room temperature after exposure to cryogenic temperature may possibly result in larger debonded interfaces (figure 5). Higher crosshead speed during testing restricts and/or minimizes the relaxation processes at the crack tip [12]. Thermal stress-induced cracks may possibly grow without blunting at a steady rate. That could reduce the interlaminar strength at higher loading rate.

**CONCLUSIONS**

It can be concluded that the ILSS values for cryogenic conditioned samples are higher than the rest two cases for almost all the crosshead speeds. This may be attributed to mechanical keying factor at the interface due to contraction of epoxy at cryogenic temperature. Also it was found that the glass-epoxy composites are loading rate sensitive. At lower crosshead speeds the shear strength value increases and decreases at higher speeds for all the three cases. It may possibly be attributed to the less prevalent relaxation process at the crack tip. The crack blunting may happen to be less common occurrence at higher rate of loading. Relaxation phenomena may be attributed to the samples exposed to room temperature after cryogenically conditioning for the mechanical behavior at different loading rates.
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FIGURE CAPTIONS

Figure 1  Variation of ILSS of glass-epoxy composites with crosshead speed at cryogenic temperature (♦), ambient temperature after cryogenic conditioning (■) and ambient temperature (▲).

Figure 2  Scanning micrograph shows matrix cracking at high loading rate (500mm/min) of as cured specimen at ambient temperature.

Figure 3  Scanning micrograph shows fiber/matrix debonding due to differential contraction of cryogenically conditioned specimen.

Figure 4  Scanning micrograph shows massive microcracking and delamination of cryogenically conditioned specimen.

Figure 5  Scanning micrograph shows debonded interfaces of specimens at room temperature after cryogenic conditioning.
Figure 1
Figure 2
Figure 3
Figure 4
Figure 5