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Hygrothermal Effects on the Tensile Strength of Carbon/Epoxy Laminates with Molded Edges

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The interlaminar stresses are confined to a region near the free edge. Therefore, the laminate stacking sequence and the free edge finishing are some of the factors that affect the strength of the laminate and limit its life. The use of molded edges eliminates the need for trimming and machining the laminates edges thus improving productivity. However, this fabrication technique may have a detrimental effect on the laminate strength for certain stacking sequences. This effect in the presence of moisture has not been characterized.

This work presents the results of a comparative study of the resistance to delamination of laminates with machined edges and molded edges. Additionally, two environmental conditions were considered: dry laminates and laminates saturated with moisture. The tensile strength of the laminates were measured and micrographs were used to analyze the microstructure of the laminates near the free edges. It is concluded that the mechanical properties of advanced composites depend on the environmental conditions and the fabrication techniques used to produce the laminates. Therefore, it is necessary to account for these factors when experimentally determining the design allowables.

Keywords: *composites, hygrothermal effects, free edges, tensile strength*

1. Introduction

Due to their low specific mass combined with very high strength and stiffness the advanced composite materials have been widely used in substitution of traditional structural materials in a number of applications. Composites present not only very high static strength but also high resistance to fatigue and corrosion. The great variety of types of fibers and resins that are commercially available enables the designer to tailor a wide range of physical and mechanical properties that are suitable for a specific application.

The great acceptance of these materials for the aeronautical and space applications is due to the fact that composite materials provide the high specific strength and stiffness required for structural components, presenting lower weight than the metallic components. However, one important problem associated to the composite materials processing is their tendency to suffer delaminations initiated at free edges. This kind of damage can be introduced in laminated composites during the manufacturing process or part operation. The free edge delamination effect reduces the strength, the stiffness and the fatigue life of the laminate, and may lead to a catastrophic failure of the whole structure. Free edge delaminations appear as a consequence of high interlaminar stresses, concentrated in the vicinity of free edges, combined with low interlaminar fracture toughness of laminated composites, which are dependent on the toughness of the polymeric matrix.

Almeida and Cândido[1] investigated a fabrication process based on laminates with molded edges that increases the productivity and reduces the manufacturing cost without penalizing the structural weight of the part. The free edge

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finishing is a required operation in the conventional fabrication process of composite laminated parts. The conventional manufacturing process for these parts consists in the lamination of several pre-impregnated layers on the polished internal surface of a mold; the layers are cut with a small excess of material with respect to the geometry of the finished part. After cure, the laminate must be cut and trimmed to the dimensions and tolerances specified in the design. This operation may induce undesirable free edge delaminations.

On the other hand, the fabrication process for laminates with molded edges consists in stacking pre-impregnated layers, which have been cut to the exact required size, matching the mold dimensions, eliminating the finishing operation mentioned above and, as a consequence, increasing productivity, reducing cost and minimizing free edge delaminations. Moreover, molded edges may also appear in composite plates with co-cured stringers. In this situation, the region at the intersection of the end of the stringer with the plate is also prone to delamination and present similar characteristics to the molded edges studied in this work.

However, it has been experimentally verified[1] that if the laminate construction is not adequate, the use of molded edges may lead to a significant reduction in the laminate tensile strength. This occurs because, during the curing process, resin and fibers move towards the molded edge under the action of external pressure. Depending of the stacking sequence pure resin pockets and/or voids may be formed near the free edge affecting the interlaminar stress magnitude and the local toughness of the laminate. Experimental results[1] demonstrated that the arrangement of resin and longitudinal fibers near the free edge strongly influences the laminate strength.

The laminate damage tolerance may be improved if the onset of free edge delamination is prevented and/or its growth retarded. The main factors related to the material properties and laminate manufacture that influence the damage tolerance, thus affecting the free edge delamination are: the polymer matrix toughness, the fiber/matrix interface strength, the fibers orientation, lamina stacking sequence and the laminate thickness. The proposed methods to increase the delamination strength are based either on the reduction of interlaminar stresses or in techniques that increase the out of plane laminate strength. These techniques will be acceptable in practical applications only if they do not penalize the weight and the cost of the structure, and preserve the laminate stiffness and strength under any possible load applied to the part.

The fracture toughness in the vicinity of a free edge may be improved by stitching, braiding or capping the edge[2]. It is well known that stitching can effectively arrest edge delamination but might have adverse effects on the strength and fatigue life of laminates[3]. Braiding provides effective fiber reinforcement in the out of plane direction but its complexity results in manufacturing constraints in the laminate design. The use of a U-shaped cap along the edge can also improve the laminate strength[4]. The strength and fatigue life of laminates with U-shaped cap increase but this technique is limited by manufacturing constraints and lead to a major increase in the cost of the structure because of the higher complexity of the fabrication process and required non-destructive inspection.

Other techniques have also been proposed, for example, adding elastomers to the resin, and/or decreasing of crosslink bonds thus increasing the polymer chain flexibility. These chemical modifications can increase the interlaminar fracture toughness of the composite. Other alternative is the addition of thermoplastics to the thermoset resin. Thermoplastic matrices (for example polietheretherketone - PEEK) present larger values of fracture toughness than thermoset resins.

Another concept proposed to minimize the interlaminar stresses at the free edges is by terminating critical plies at a certain distance from the free edge[5]. In this case, severe Poisson ratio mismatches between adjacent plies can be avoided[2]. Similarly, the use of adhesive layers at the critical interfaces, may reduce the interlaminar stresses, increasing the static strength and fatigue life of the laminate[6]. Another proposed technique[7] is to produce narrow notches at the free edge laminate resulting in a decrease of the interlaminar stresses near the free edge. However, the presence of the notches introduce high in plane stress concentrations. Therefore, this technique is effective for thin laminates only.

The environmental action, such as high moisture concentration, high temperatures, corrosive fluids and ultraviolet radiation (UV), can also affect the performance of advanced composites. These factors can limit the applications of composites by deteriorating the mechanical properties over a period of time. This degradation is due to the chemical and/or physical damages caused in the polymer matrix, loss of adhesion of fiber/resin interface, and/or reduction of fiber strength and stiffness.

Most studies involving environmental effects on advanced composites have addressed the combined effect of moisture content, time and temperature[8-10]. Many advanced composites parts can absorb water when exposed to moisture. The laminated thickness, temperature, and ambient moisture content will establish the moisture concentration equilibrium reached by the composite. The environmental effects on the composites have been the subject to numerous experimental and theoretical studies because of their significant effect on their mechanical properties and because of the easy and effective application of the Fick Diffusion Theory[8-10].

The main advantage of producing laminates with molded edges is the simplification of the finishing operation of the part eliminating cutting and grinding thus reducing manufacturing costs. However, this fabrication technique may have a detrimental effect on the laminate strength for certain stacking sequences. The objective of this work is to analyze the effect of the environmental conditioning tensile strength of carbon/epoxy cross-ply laminates with machined and molded edges.

2. Fabrication of the Laminates

Three different families of laminates were produced using unidirectional carbon/epoxy tape. They were manufactured with pre-impregnated high strength carbon fiber, previously treated to improve the chemical compatibility with the modified epoxy resin F-584, from Hexcel Composites. This catalyzed resin system is partially polymerized, presenting higher toughness and impact strength values than the conventional thermoset resins.

The main engineering properties of this material were obtained by the following static tests: (a) uniaxial tension; (b) uniaxial compression; (c) in plane shear; and (d) interlaminar shear. The test specimens were tested in dry and wet conditions using ASTM standards. Considering that this material has two principal directions it is necessary to measure the tensile and compression properties in both directions. In addition, all specimens were obtained with material from the same batch to minimize the data dispersion. The laminate mechanical properties are presented in Table 1.

Table 1. Mechanical properties of the unidirecional carbon fiber reinforced F584 epoxy resin tape.

Mechanical property	Dry	Wet	ASTM Standard
Longitudinal modulus, E_1 (GPa)	130.1	133.9	D 3039
Transverse modulus, E_2 (GPa)	2.0	2.0	D 3039
Shear modulus, G_{12} (GPa)	5.8	5.8	D 4255
Poisson ratio, ν_{12} (GPa)	0.27	0.29	D 3039
Longitudinal tensile strength, X_T (MPa)	1721.2	1085.3	D 3039
Transverse tensile strength, Y_T (MPa)	42.9	26.9	D 3039
Longitudinal compressive strength, X_C (MPa)	702.6	558.1	D 3410
Transverse compressive strength, Y_C (MPa)	133.3	112.8	D 3410
In-plane shear strength, S_{12} (MPa)	88.0	88.0	D 4255
Interlaminar shear strength, S_{13} (MPa)	84.4	57.5	D 2344
Density, ρ (kg/mm^3 x 10^{-5})	0.155	0.155	D 3171

The three cross-ply laminated families studied in this work were selected such that they present the same value of the in plane mechanical properties, according to the Classical Lamination Theory[11]. The families of laminates obtained are: $[0/90/90]_S$, $[90/0/90]_S$ and $[90/90/0]_S$. The delaminations typically emanate from the free edge at the (0, 90) interfaces propagating towards the laminate center. This phenomenon occurs due to Poisson ratio mismatches between adjacent 0° and 90° layers.

In order to study the free edge effect on the laminate strength, one group of test specimens was prepared with machined edges and another with molded edges for each family of laminates. Three plates, one for each family, were manufactured to produce the specimens with machined edge. All plates were simultaneously cured at 181 °C and 0.69 MPa (6.9 bar) of pressure and vacuum of 0.083 MPa (0.83 bar), according to the cure cycle showed in Fig. 1.

The cured plates were ultrasonically inspected for defects such as porosities, resin starved areas, cracks and delaminations. The pulse-echo technique was used and no discontinuities were observed. The laminates were approved for the next operation, the surface preparation for bonding the tabs. The tabs were manufactured with glass/epoxy using cloth 7881 pre-impregnated with resin F-155. The tabs were bonded with structural adhesive EA 9628.045 PSF supplied by Hysol and cured at 120 °C.

After the tabs were bonded, five specimens were carefully cut from each plate using a high-speed diamond disk to avoid edge delaminations. The edges of each specimen were then polished with grinding machine producing an excellent finishing. The tensile strength tests followed the procedure described by Lagace[12]; the specimen has nominal dimensions of 350 mm in length and 50 mm in width, as shown in Fig. 2.

The specimens with molded edges were manufactured in an aluminum mold specially developed for this purpose.

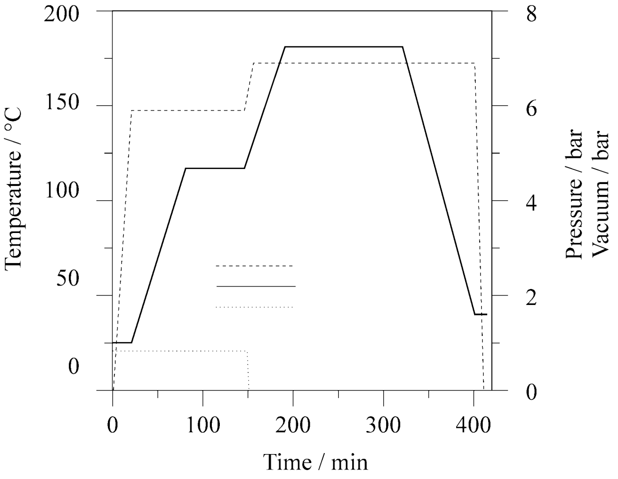

Figure 1. Cure cycle.

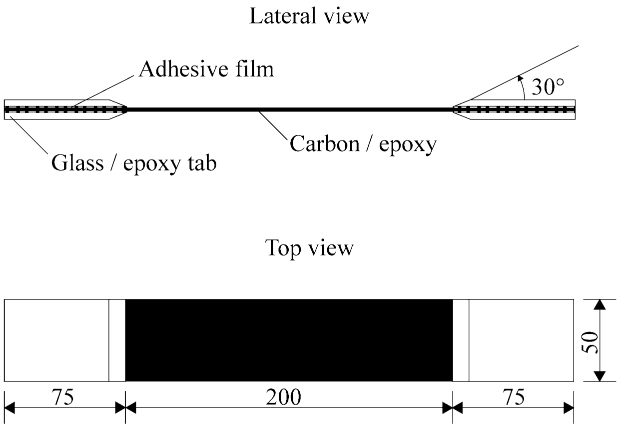

Figure 2. Geometry of the specimen.

Five specimens with the same nominal dimensions described above can be simultaneously cured in the mold. The pre-impregnated layers were cut to a width slightly smaller than the nominal value to account for material accommodation and thermal expansion during cure. The cure cycle used was the same used for the specimens with machined edge, Fig. 1.

After cure, all the irregularities at the edge surface were gently removed with a fine sand paper. A total of five specimens with molded edge were produced for each family of cross ply laminates. All specimens, with machined edges and molded edges, were once more subjected to ultrasonic inspection using a transmission technique. The aim was to detect the existence of delaminations that could be introduced during the machining or finishing of the edges. No defects were found and all specimens were prepared for the environmental conditioning.

A total of 30 tensile specimens were manufactured (five specimens of each type for three stacking sequences and two types of edge finishing).

3. Environmental Conditioning

In order to assess the influence of the environmental conditioning on the tensile strength, the specimens with machined edges and molded edges were exposed to a combination of high temperature and humidity in an environmental conditioning chamber. The condition selected to saturate the specimens before the mechanical tests is based on Procedure B of ASTM Standard D 5229 M-92. The variation in the moisture level in the laminate as a function of time is monitored by periodically measuring the mass of traveler samples totally exposed to a certain environmental condition until the moisture equilibrium state is reached. The test temperature was defined to be 80 °C, that corresponds to the maximum value recommended by the standard for epoxy resin matrices cured at 177 °C; the relative humidity in the chamber was set to 95%.

Four traveler samples without tabs, as recommended by the standard, of each family of laminates with machined edges and molded edges were cut to nominal dimensions of 50 x 50 mm. All tensile test specimens and traveler samples were previously dried in an oven and weighted. They were then stored in an environmental conditioning chamber Heraeus Vötsch model VUK 08/1000 calibrated to maintain the prescribed test temperature and relative humidity level within ±1 °C and ±3%, respectively.

A total of 24 moisture absorption specimens were manufactured (four specimens of each type for three stacking sequences and two types of edge finishing).

4. Tests and Results

Three types of tests were conducted in this work: (a) the mass gain of the material subjected to the environmental conditioning described in the previous section; (b) tensile test of all specimens; and (c) microscopic analyses of all laminates types.

The mass gain due to moisture absorption was measured in terms of a percentage of the dry mass of the material as specified in the standard ASTM D 5229 M-92. Once a week, all traveler samples were removed from the chamber and weighed in an analytical balance to register the mass gain while the tensile test specimens remained in the chamber. The samples were quickly removed from the chamber and immediately stored in a plastic bag to minimize interference in the moisture content. After the traveler samples were found to be saturated with moisture, the specimens were removed, stored in bags and immediately sent to the tension test. Table 2 presents the average mass gain of the three families of laminates with machined edges and molded edges along the five weeks of conditioning in the chamber. The standard deviation was smaller than 12% for all measurements.

The importance of the free edge effect on the strength of the cross ply laminates for both types of edge finishing studied in this work was assessed by the stacking sequence

Table 2. Average mass gain of traveler samples.

Laminate	Edge finishing	Average mass gain (%)				
		1st week	2nd week	3rd week	4th week	5th week
$[90/90/0]_S$	Machined	0.65	0.67	0.69	0.70	0.71
	Molded	0.62	0.65	0.68	0.68	0.72
$[90/0/90]_S$	Machined	0.63	0.66	0.68	0.67	0.71
	Molded	0.77	0.80	0.83	0.82	0.85
$[0/90/90]_S$	Machined	0.63	0.66	0.68	0.67	0.71
	Molded	0.65	0.68	0.73	0.72	0.78

effect on the tensile strength of the laminates for a given environmental conditioning. The tensile tests were conducted in an Instron test machine model 1332, at room temperature with speed of 1.0 mm/min.

The ultimate (total fracture) tensile strength of each family of the saturated laminates for each type of edge finishing is presented in Fig. 3. For comparison purposes, Fig 3. also includes the results obtained for dry specimens obtained by Almeida and Cândido[1] using the same procedure. The highest and lowest measurement for each laminate type is shown to characterize the data dispersion.

Typical micrographs of the cross section of the laminates analyzed in the region near the free edge are shown in Figs. 4-7.

5. Discussion

The moisture absorption results presented in Table 2 show that all specimens had mass gains of the same order. This should be expected because the stacking sequence and the edge finishing should have small influence on the moisture absorption. In fact, the laminate with molded edge presented, in general, a slightly higher mass gain. This is probably due to the fact that the molded edges may be resin rich, as shown in Figs. 5-7. Since the molded edge region is very narrow (of the order of 0.3 mm) the higher moisture concentration in this area should have a small effect in the total mass gain of the sample.

The tensile test results, presented in Fig. 3, show a small stacking sequence effect for laminates with machined edges for both dry and saturated conditions. The data dispersion for these laminates tend to be small because of the excellent edge finishing provided by the grinding machine, much superior to the quality obtained when the usual production tools are used. On the other hand, the tensile strength of laminates with molded edge is very sensitive to the stacking sequence. Laminate $[0/90/90]_S$ presents a substantially lower strength; this can only be explained by free edge effects as the laminates are equivalent in terms of in-plane mechanical properties according to the Classical Lamination Theory[11]. The distribution of fibers and matrix in the free edge region of this laminate explains its lower strength.

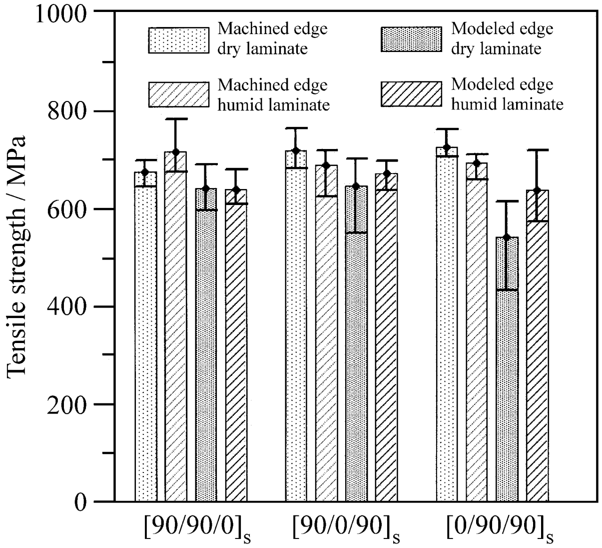

Figure 3. Tensile tests results.

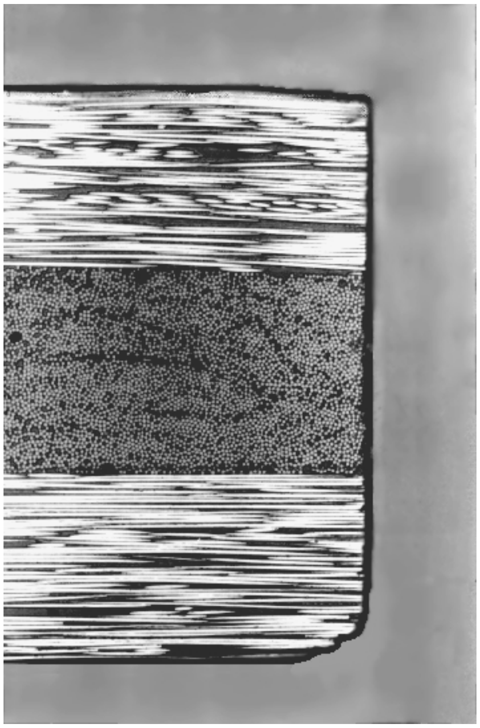

Figure 4. Micrograph of the cross section of a $[90/90/0]_S$ laminate with machined edge (100 X).

Micrographs of the cross section of the free edge region of each laminate type were analyzed after the environmental conditioning to observe the arrangement of fibers and matrix in this area, and the quality of the laminate manufacturing. Figure 4 shows a typical cross section view of the free edge region of a $[90/90/0]_S$ laminate with machined edges, demonstrating the excellent edge finishing obtained with this technique.

Figures 5-7 show the cross section of laminates $[90/90/0]_S$, $[90/0/90]_S$ and $[0/90/90]_S$ with molded edges. With this type of finishing, the longitudinal fibers along the edge (0°) move laterally towards the edge under the external pressure applied during the cure cycle in the autoclave. As a consequence, a small resin rich area reinforced only by longitudinal fibers is formed at the molded edges. This region spans typically over 0.3 mm from the edge. Pockets of pure resin or relatively large voids may be formed at the molded edge depending on the stacking sequence. It can also be observed that for laminates $[90/90/0]_S$ e $[90/0/90]_S$, shown in Figs. 5 and 6, the longitudinal fibers move from the internal layers to the external layers forming a reasonably homogeneous region of resin reinforced by 0° fibers.

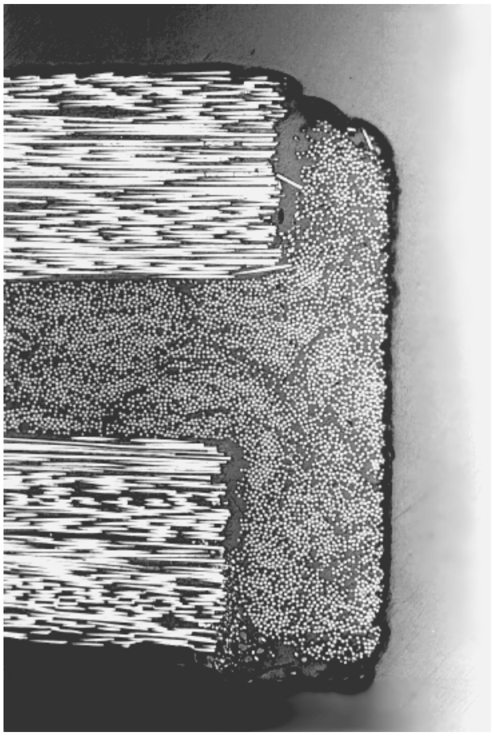

Figure 5. Micrograph of the cross section of a $[90/90/0]_S$ laminate with molded edge (100 X).

On the other hand, Fig. 7 shows that, in the case of the $[0/90/90]_S$ laminate, the longitudinal fibers tend to move from the external to the internal layers. However, this movement is practically prevented by the flow of the excess of resin from the internal layers forming a pocket of pure resin or a relatively large void near the edge, as shown in the figure. Therefore, for laminates with molded edge the arrangement of fibers and resin in the free edge region are dependent on the stacking sequence and affect the laminate strength.

Moisture affects the distribution of interlaminar stresses as well as the mechanical behavior of the resin. Figure 3 shows that the result of these combined effects may either increase or decrease the laminate strength, depending on the relative importance of them for the particular case at hand. For example, the strength of the $[0/90/90]_S$ laminate with molded edges is substantially increased by moisture. In general, it is noted that when moisture has a favorable effect on the strength of a laminate with molded edge it is detrimental to the strength of the corresponding laminate with machined edge and vice-versa. The analysis of the moisture effect would require accurate moisture absorption models including its effects on the interlaminar stresses distributions and on the mechanical properties of the resin.

6. Conclusions

The correct specification of the cure cycle was important to produce high quality specimens for both types of edge finishing considered. The adequate pressure and temperature profiles used resulted in uniform thickness laminates free of defects, as verified in both ultrasonic inspections. Moreover, the manufacture processes used presented good repeatability. The microscopic analyses did not show the existence of damage caused by the environmental conditioning.

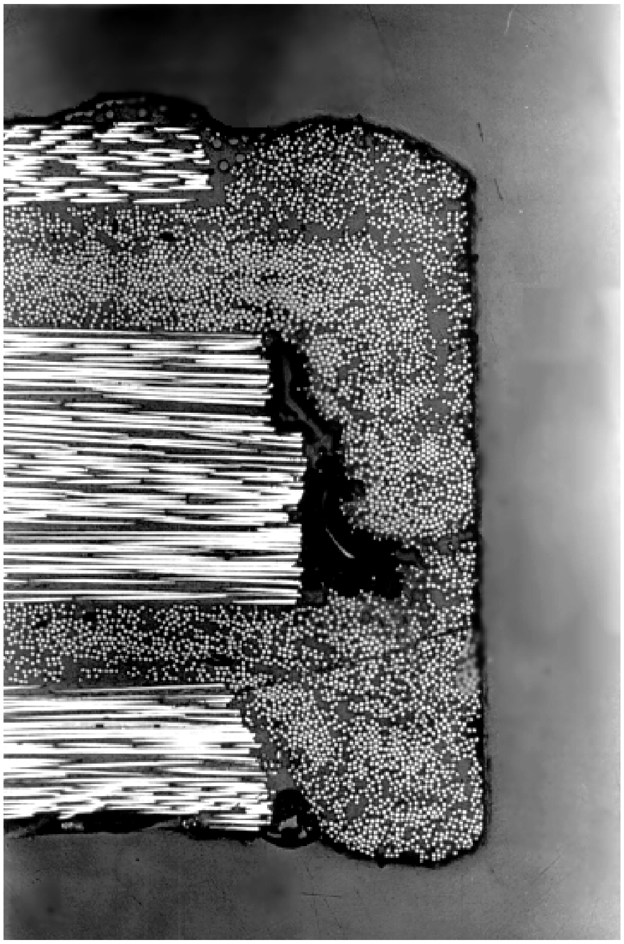

Figure 6. Micrograph of the cross section of a $[90/0/90]_S$ laminate with molded edge (100 X).

The tensile test results show that both the manufacturing process and the environmental conditions may significantly affect the laminate strength. The results show a small stacking sequence effect for laminates with machined edges for both dry and saturated conditions. On the other hand, the tensile strength of laminates with molded edge is very sensitive to the stacking sequence. It is found that, depending on the stacking sequence, voids and/or pockets of pure resin may be formed at the molded edge causing a significant reduction in the tensile strength.

The effect of moisture content on the tensile strength is complex because it affects both the distribution of interlaminar stresses as well as the mechanical behavior of the resin. The experimental results indicate that these combined effects may either increase or decrease the

Figure 7. Micrograph of the cross section of a $[0/90/90]_S$ laminate with molded edge (100 X).

laminate strength, depending on the relative importance of them for the particular stacking sequence and type of edge finishing.

The use of molded edge reduces the manufacturing cost of composite parts. However, it may have a detrimental effect on the laminate strength. Therefore, the effects of edge finishing and environmental conditions must be taken into account in the experimental characterization of the laminates when establishing design allowables for composite structures.

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