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Hygrothermal effects on the shear properties of carbon fiber/epoxy composites

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Abstract The environmental factors, such as humidity and temperature, can limit the applications of composites by deteriorating the mechanical properties over a period of time. Environmental factors play an important role during the manufacture step and during composite's life cycle. The degradation of composites due to environmental effects is mainly caused by chemical and/or physical damages in the polymer matrix, loss of adhesion at the fiber/matrix interface, and/or reduction of fiber strength and stiffness. Composite's degradation can be measure by shear tests because shear failure is a matrix dominated property. In this work, the influence of moisture in shear properties of carbon fiber/epoxy composites (laminates $[0/0]_s$ and $[0/90]_s$) have been investigated. The interlaminar shear strength (ILSS) was measured by using the short beam shear test, and Iosipescu shear strength and modulus (G_{12}) have been determinated by using the Iosipescu test. Results for laminates $[0/0]_s$ and $[0/90]_s$, after hygrothermal conditioning, exhibited a reduction of 21% and 18% on the interlaminar shear strenght, respectively, when compared to the unconditioned samples. Shear modulus follows the same trend. A reduction of 14.1 and 17.6% was found for $[0/0]_s$ and $[0/90]_s$, respectively, when compared to the unconditioned samples. Microstructural observations of the fracture surfaces by optical and scanning electron microscopies showed typical damage mechanisms for laminates $[0/0]_s$ and $[0/90]_s$.

Introduction

Fiber reinforced epoxy composites are used in a wide variety of applications in the aerospace field. These materials have high specific moduli, high specific strength and their properties can be tailored to application requirements. In order to screening optimum materials behavior, the effects of external environments on the mechanical properties during usage must be clearly understood [1–6].

The environmental action, such as high moisture concentration, high temperatures, corrosive fluids or ultraviolet radiation (UV), can affect the performance of advanced composites during service. These factors can limit the applications of composites by reducing the mechanical properties over a period of time. Properties deterioration is attributed to the chemical and/or physical damages caused in the polymer matrix, loss of adhesion at the fiber/matrix interface, and/or reduction of fiber strength and stiffness [7–10].

The bond between the reinforcing fiber and the matrix plays a very important role in the transfer of stresses in a composite during loading. In general, the influence of moisture in composite materials is most notably present in the matrix. Polymers, such as epoxies, are prone to absorb moisture when exposed to humid environments. This takes place through a diffusion process, in which water molecules are transported from areas of high concentration to areas of low moisture concentration [7–10].

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Composite's shear properties are mainly dominated by matrix and fiber/matrix interface. The determination of shear properties of advanced composites takes challenges as semi-static mechanical tests. One of the main difficulties in the development of shear test for these materials is to induce a pure shear stress state in the gauge section of a constant magnitude. This is a special concern for composites with high anisotropy and structural heterogeneity. The ideal shear test must be simple enough to perform, require small and easily fabricated specimens, enable measuring of very reproducible values for both shear modulus and shear strength at simple data procedure [11–13].

Different devices for the study of the shearing properties of composites are proposed in the literature [14–18]. By the end of the sixties, N. Iosipescu proposed a shear test, which allows a nearly pure-shear stress state at the shear plane [19]. This test was originally designed for measuring shear strengths and moduli of isotropic metals [19, 20]. Subsequently it was extended for testing composite materials by Walrath and Adams [21]. Due to a relatively simple geometry and loading configuration, the method has gradually evolved as the most popular test for shear characterization of materials [21–26]. The test essentially consists of a double V-notched beam specimen, in which two counter reacting forces are applied, such that a state of pure shear is generated along the specimen midsection.

For composite materials, Iosipescu test can also be performed in laminates having different fiber reinforcement orientation. For instance, it is reported in the literature that composites having fiber reinforcement placed to 0°, a visible crack typically develops at the notch root, causing a small drop in load prior to ultimate failure [21–26]. For laminates having reinforcement placed to 0/90°, the shear failure load may be lower than the maximum load attainable during the test. In such materials the fibers may reorient the shear failure, subsequently allowing the fibers to carry a major portion of the load. This reorientation is more likely to occur in composites with tough matrix materials that are very nonlinear in shear, or in laminates containing off-axis fibers. In such cases, the shear failure load can often be determined by correlating visual observation of failure in the test section with a load drop, or by a significant change in the slope of the load-displacement curve [27].

Thus, the purpose of the present work is to evaluate the effect of the environmental conditioning in the shear behavior of carbon fiber/epoxy composites specimens comparing results from Iosipescu shear and short beam shear tests.

Experimental

Processing of the laminates

Two different families of laminates were produced using unidirectional carbon/epoxy prepreg tapes. They were manufactured with a high strength carbon fiber (250 GPa) and F-155 epoxy resin system prepreg, supplied by Hexcel Composites. The families of laminates were: $[0/0]_s$ and $[0/90]_s$. The fiber content in each laminate is approximately 60% (v/v). The laminates were simultaneously cured in autoclave at 121 °C, under a pressure of 0.69 MPa and vacuum of 0.083 MPa, according to the manufacturer's cure cycle. The composite laminates consisted of 14 laminated plies resulting in a nominal thickness of 4.1 mm for both orientation patterns.

Environmental conditioning

In order to assess the influence of the environmental conditioning on the shear strength, the carbon/epoxy specimens were exposed to a combination of temperature and humidity in an environmental conditioning chamber. The conditions selected to saturate the specimens before the mechanical tests were based on Procedure B of ASTM Standard D 5229 M-92 [28]. The moisture level attained by the laminate was periodically monitored as a function of time by measuring the mass of traveler samples until the moisture equilibrium state is reached. During conditioning, the temperature was set at 80 °C and the relative humidity in the chamber was set to 90%. The temperature must remain well below the resin glass transition temperature in order to avoid the onset of irreversible damage (swelling and cracks), which permanently changes the absorption characteristics of the material.

Interlaminar shear strength tests (ILSS)

The interlaminar shear test (short beam shear test) was performed according to ASTM D2344 [29]. Ten specimens were tested for each type of stacking sequence in order to assess the effect of environmental condition on the ILSS. The dimensions of the sample were $24.0 \times 6.35 \times 4.10$ mm (length × width × thickness). The tests were performed in an Instron mechanical testing machine using a test speed of 1.3 mm/min.

Iosipescu shear tests

The Iosipescu test method determines the shear properties of composite materials. The sample geometry coupon is shown in Fig. 1. The square area covered by the gages is referred to as the test region in this study. The radius of the notch tips is 1.3 mm. The thickness of specimen was 4.1 mm. Shear strain was measured by bonding strain gages at ±45°, placed at the mid-section between the two notch tips (Fig. 1).

The specimen properly fit into the fixture assuring that the notch is located along the line-of-action of loading (Fig. 1). The notches influence the shear strain along the loading direction, resulting in a more uniform stress distribution than would be without the notches. By applying two force couples that generate two counter-acting moments, a pure and uniform shear stress state is generated at section a–b (Fig. 2—notch roots). During the Iosipescu test, the applied compressive load is along the long axis of the specimen, which is defined as 1 direction (σ_{11}) and the fibers are aligned at an angle θ from the loading axis.

The resulting shear and moment diagrams are also shown in Fig. 2.

The shear modulus can be obtained by the shear stress and corresponding shear strain as long as the stress and strain are uniformly distributed in the test region where they are measured. Therefore, the uniformity of shear stress and corresponding strain in the test region need to be examined to assess the accuracy of the shear modulus determination. The apparent in-plane shear strain, γ, at the cross-section along two notch tips can be calculated from the measurement of normal (longitudinal) strain of the gauges, ϵ ± 45°, as follows:

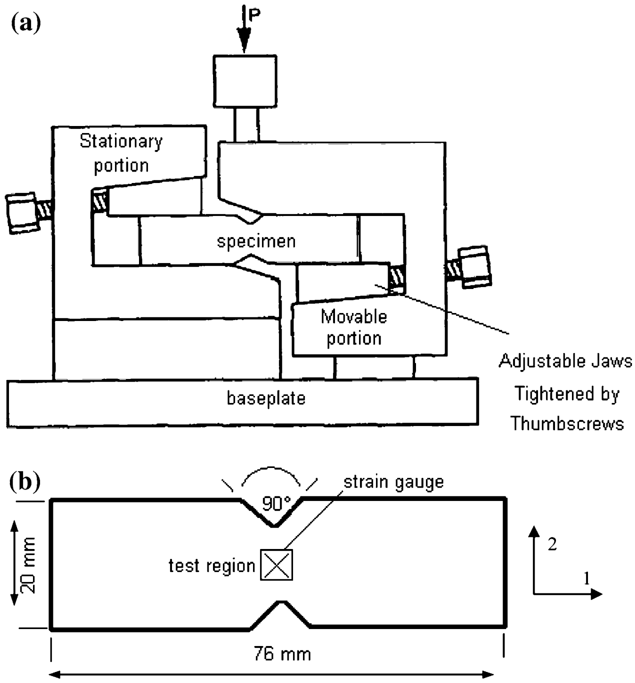


Fig. 1 Iosipescu test Scheme: (**a**) Iosipescu dispositive; (**b**) specimen configuration

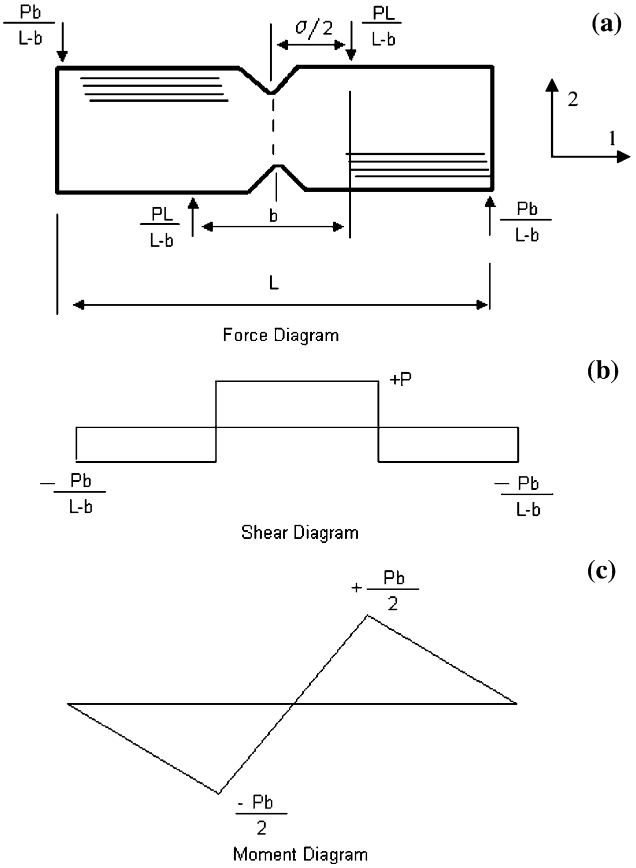


Fig. 2 Idealized force, shear and moment diagrams for Iosipescu test

$$\gamma = \varepsilon^{+45^\circ} - \varepsilon^{-45} \quad (1)$$

The Iosipescu tests were performed according to ASTM D 5379 [27] and performed in an Instron mechanical testing machine using a test speed of 0.5 mm/min.

Microstructural analysis

Scanning Electron Microscopy (SEM) and Optical Microscopy (OM) were employed to characterize the shear failure modes in the carbon/epoxy shear coupons. A Leo 435VPI Microscope was used for SEM analysis. Optical microscopy was performed using an Olympus BH equipment.

Results and discussion

Evaluation of laminate quality

Figure 3a–b show scanning electron microscopies of carbon fiber/epoxy composite cross-sections having fiber orientation at 0° (Fig. 3a) and 0/90° (Fig. 3b). A homogeneous distribution of matrix around fibers and the absence of porosity cracks are observed in the micrographs, which is an indication of a void free laminate.

Theoretical results

Composite micromechanics calculations were performed by using the Fabric Geometry Model (FGM) software in order to compare theoretical and experimental results. The FGM software allows predicting the stiffness of composite materials with spatially oriented reinforcements [30].

The parameters used to calculate the elastic constants of composite materials are presented in Table 1. The theoretical elastic values obtained using the FGM program for all specimens are listed in Table 2 [30].

Table 1 Parameters used in the FGM program

Parameter	Epoxy resin	Carbon fiber
Volume content (%)	40.0	60.0
E_1 (GPa)	5.00	220
E_2 (GPa)	5.00	20.0
G_{12} (GPa)	1.85	15.0
ν_{12}	0.30	0.20

Table 2 Theoretical engineering constants

Properties	0°	0/90°
G_{12} (GPa)	3.86	3.86
G_{13} (GPa)	2.92	3.39
G_{23} (GPa)	3.86	3.39

Environmental conditioning

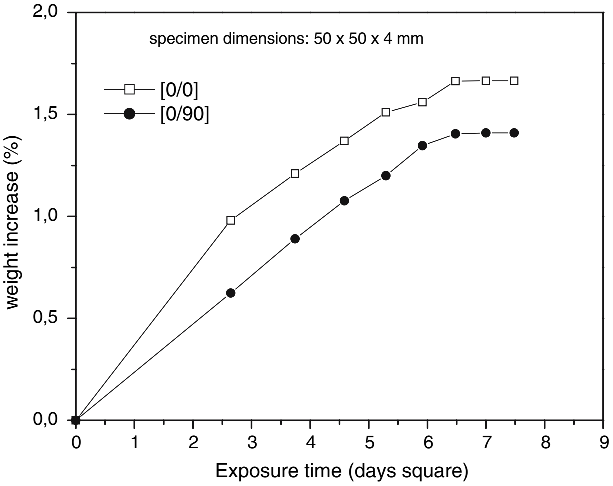


Fig. 4 Weight increase of carbon fiber/epoxy composites specimens exposed at 80°C and 90% RH

Figure 4 shows the weight increase as a function of exposed time for carbon fiber/epoxy composites specimens exposed at 80°C and 90% RH. Like any other polymers, epoxies can absorb moisture when exposed to humid environments. Moisture absorption takes place through of a diffusion process, in which water molecules are transported from areas with higher concentration to areas with lower moisture concentration [1]. As a result of different fiber orientation in composite materials, moisture can penetrate more or less inside the polymeric matrix. Moreover, the moisture absorption is not uniform throughout the material [1]. It was observed that cross-ply composites leads to a lower rate of moisture absorption (maximum of 1.4%) compared to unidirectional reinforced composites (maximum of 1.8%). Since the 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. However, the bidirectional reinforced composites presents resin rich areas at the edge of the composite when compared with unidirectional reinforced composite, leading to a higher moisture absorption.

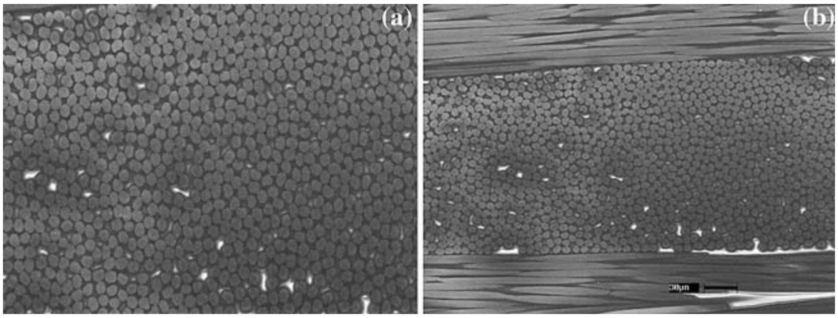


Fig. 3 Micrographs of carbon fiber/epoxy laminates used in this work: (**a**) laminate $[0/0°]_s$; (**b**) laminate $[0/90°]_s$

The kinetics of the diffusion process depends on the temperature and relative moisture absorption. The higher is the relative moisture absorption the greater is the absorption rate. The diffusion process can be describe by Fick's law [7–10].

Interlaminar shear strength

The short beam shear test measures the apparent interlaminar shear strength of a composite material. Therefore, the short beam shear method is not suitable for design purpose. Despite this restriction, data generated from this test method is still for design allowables purposes, primarily because of the lack of any alternative test methods for measuring interlaminar strength [21, 22].

Table 3 presents the interlaminar shear strength (ILSS) results for carbon/epoxy composites. As expected, the ILSS values decreases when exposed to hydrothermal conditioning. Wet conditioning induces strong matrix plasticization [7].

According to the Table 3, the interlaminar shear strength values for carbon fiber/epoxy composites shows a decrease of 21% and 18% for $[0/0°]_s$ and $[0/90°]_s$ laminates, respectively. The hygrothermal effects on the mechanical properties of composites are described in detail in the literature [7–10] but there is not a general agreement over the magnitude of this effect. The difficulty is to evaluate the influence of both moisture absorption and temperature at the same time on the mechanical properties. It is reported in the literature that the interlaminar shear strength for unidirectional carbon fiber/epoxy composites are: 112 MPa (before hygrothermal conditioning) and 77 MPa (after hygrothermal conditioning) [9]. For this case, carbon fiber/epoxy composites shows a decrease of 31% due to matrix plasticization and degradation of carbon fiber/epoxy interface properties. The differences between ILSS values found in this work and results from literature can be attributed to differences in experimental hot/wet conditioning parameters.

Table 3 Interlaminar shear strength values for carbon fiber/epoxy composites

τ (MPa)	Before hygrothermal conditioning	After hygrothermal conditioning
$[0/0]_s$	84.5 ± 3.3	60.8 ± 2.0
$[0/90]_s$	66.4 ± 2.1	49.6 ± 1.9

Figure 5a–b shows the aspect of the failure mode in composites specimens after ILSS test. The tested specimens revealed interlaminar failures, either in simple or multiple delamination modes. It can be observed in Fig. 5a that laminate $[0/0°]_s$ shows a typical interlaminar fracture between the layers by multiple shear cracks running along parallel planes. On the other hand, the $[0/90°]_s$ laminate can exhibit interlaminar cracks at horizontal and vertical positions in relation to the thickness of the sample (Fig. 5b), showing that crack pattern is changed during testing. Similar failure modes are observed for the specimens submitted to environmental conditioning.

Iosipescu tests results

Figure 6 shows a plot of the shear stress as a function of the displacement for the specimens tested in this work. The measurements were performed before and after hygrothermal conditioning. It is observed that there is a decrease in the shear strength of the $[0/0]_s$ and [0/90] composite laminates, after hygrothermal conditioning, ~8 and ~5%$_s$, respectively, in relation to the non-conditioned samples. As pointed out before, moisture uptake induces resin plasticization and, consequently, reduces shear strength values of the laminates.

From results of Table 4 it is clear that the influence of moisture is more predominant in $[0/0]_s$ laminate, confirming the results of Fig. 4. According to the literature, moisture affects the distribution of interlaminar stresses as well as the mechanical behavior of the resin [7–10]. It is well recognized that epoxy resins can suffer substantial reduction in T_g and mechanical properties following the water absorption [7–10]. Some

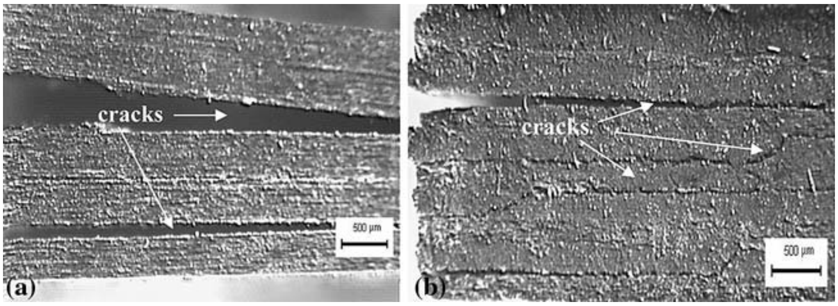
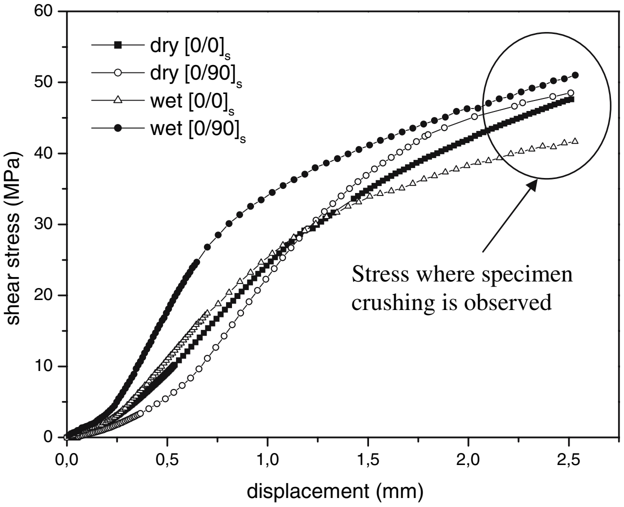


Fig. 5 Failure mode in carbon fiber/epoxy composite materials: (**a**) 0° and (**b**) 0/90°



Stress where specimen crushing is observed

Fig. 6 Shear stress as a function of displacement for Iosipescu (Orientation 12)

researchers have found that the deleterious effects of water absorption are reversible, but prolonged hydrothermal ageing may lead to irreversible damage of the resin due to susceptibility of the polymer to hydrolysis, oxidation, and changes in molecular weight [7–10]. This was also confirmed by the results obtained in this work.

The strain gauge response as a function of shear stress, for the $[0/0]_s$ and $[0/90]_s$ composites, are presented in Fig. 7a–b. These plots confirm the non-linear shearing behavior of these materials.

Table 4 summarizes results for the shear modulus (G_{12}) and shear strength (τ_{12}), from laminates $[0/0]_s$ and $[0/90]_s$ obtained by Iosipescu coupon test. In both cases, it was observed that laminate $[0/90]_s$ have G_{12} and τ values higher than laminates $[0/0]_s$ due to the reinforcement orientation. The values of G_{12} for the $[0/90]_s$ composite (3.86 GPa) is 14% higher than for $[0/0]_s$ composite. The shear strength of $[0/90]_s$ composite (~50 MPa) is 10% higher than for $[0/0]_s$ composite. It is observed that the shear modulus obtained by Iosipescu test for carbon fiber/epoxy composites is closer to the epoxy resin values (1.85 GPa) than for carbon fiber (15 GPa) [6]. So, shear behavior is a matrix-dominated properties in composites.

Table 4 Stress and modulus values obtained by Iosipescu test

Parameters	$[0/0]_s$	$[0/90]_s$
Before hygrothermal conditioning		
τ (MPa)	45.2 ± 1.7	49.8 ± 1.3
G_{12} (GPa)	2.8 ± 0.1	3.7 ± 1.2
*After hygrothermal conditioning**		
τ (MPa)	41.3 ± 1.7	47.2 ± 1.3
Theoretical G_{12} value (GPa)	3.86	3.86

* It was not possible measure G_{12} in this condition due to problems with strain gauge in this condition

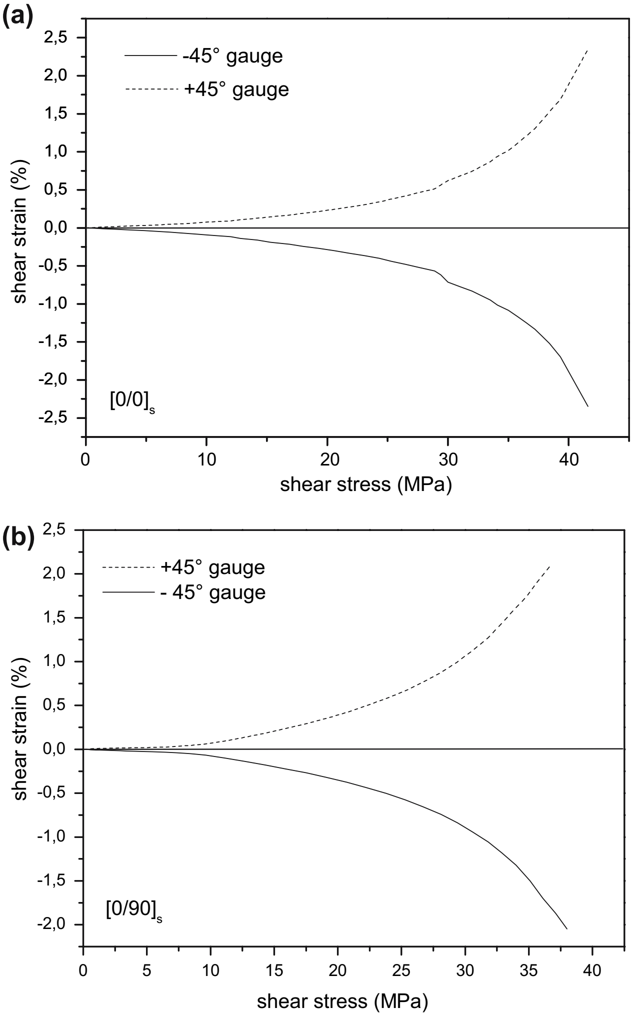
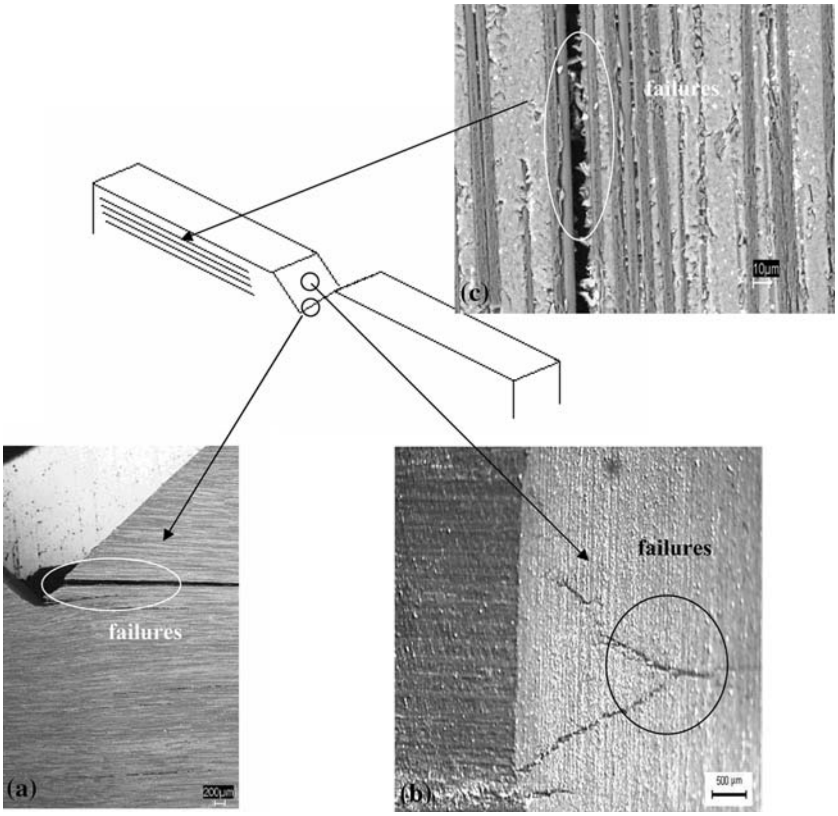


Fig. 7 Strain gauge typical response—Orientation 12: (**a**) $[0/0]_s$ and (**b**) $[0/90]_s$

In order to give support to Iosipescu experimental results, shear modulus was calculated by the FGM software for laminates $[0/0]_s$ and $[0/90]_s$. It is observed that experimental shear modulus values exhibited a decrease of 27% and 4% for laminates $[0/0]_s$ and $[0/90]_s$, respectively, when compared to calculated values. Micromechanics calcutations, which are the basis of FGM software, do not takes into account fiber/matrix interface effects and void presence. For $[0/90]_s$ laminate these factors were more pronounced than for $[0/0]_s$ laminate. In the case of $[0/0]_s$ laminate the counter-reacting loading forces during testing can facilitate shear at fiber/matrix interface lowering the experimental shear modulus value.

Laminates $[0/0]_s$ and $[0/90]_s$ exhibit different failure modes. Figure 8a shows that for $[0/0]_s$ composites the failure occurred mainly at the notch tip region, showing

Fig. 8 Iosipescu failure behavior in [0/0] carbon fiber/epoxy composite materials (**a**) failure mode, (**b**) failure behavior in the notch and (**c**) interior failure in carbon fiber/epoxy composite materials



perpendicular damages in relation to the reinforcing fibers. Figure 8b shows that for $[0/0]_s$ laminates crack branch failure may occur. In this case, interlaminar and intralaminar failure cracks at the notch tip region can be found. The failure mode is characterized by the formation of branched cracks having a common starting point. The split-formation process is associated with the notch geometry and the local stress concentrations in these areas. Figure 8c shows a failure mode associated with delamination due to shear deformation. Delamination cracks, perpendicular to loading direction, are located mainly at the Iosipescu coupon midsection (a–b region).

Figure 9a shows that delamination mainly occurred between the 0° and 90° laminae for $[0/90]_s$ composites. Figure 9b shows interlaminar and intralaminar cracks at the notch tip region. These cracks are a result of fiber splitting inside the lamina due to interlaminar shear deformation. As a result, the most common failure mode is the formation of several cracks (~500 μm) running through the 90° laminae.

Conclusion

In this work the shear behavior of $[0/0]_s$ and $[0/90°]_s$ lamintes, submitted to hygrothermal conditioning, were investigated. Interlaminar Shear and Iosipescu shear tests were used.

The interlaminar shear strength values obtained by short beam test for carbon fiber/epoxy composites exhibited a decrease of 21% and 18% for $[0/0°]_s$ and $[0/90°]_s$ laminates in relation to the unconditioned materials. The decrease in interlaminar shear is due to the degradation of the epoxy matrix. The tested specimens revealed interlaminar failures, either in simple or multiple shear modes.

The shear response obtained during Iosipescu tests confirms the non-linear shearing behavior of the composite specimens. The Iosipescu shear strength (τ_{12}) and shear modulus (G_{12}) for the [0/90] laminate are 49.8 and 3.7, respectively. The Iosipescu test shows that in both cases, it was observed that laminates $[0/90]_s$ has presented G_{12} and τ values higher than laminates $[0/0]_s$ (10 and 24%, respectively) due probably to residual tension induced by added of notch tips. When compared to numerical calculated values, the experimental shear modulus (G_{12}) for [0/0]s is 27% lower. On the other hand, for [0/90]s laminates the theoretical calculated shear modulus is 4% higher than experimental results obtained for this laminate. This difference can be attributed to experimental errors or void presence, which is not considered in the theoretical analysis.

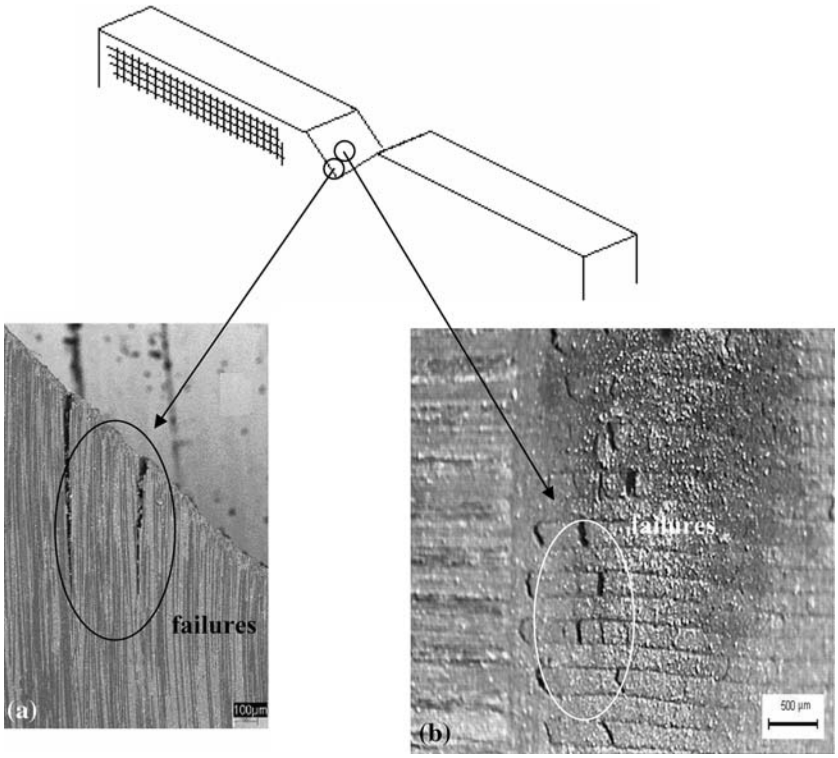


Fig. 9 Iosipescu failure behavior in [0/90] carbon fiber/epoxy composite materials (**a**) failure mode and (**b**) failure behavior in the notch

Therefore, the shear modulus value (2.8 GPa) obtained by laminate $[0/0]_s$ indicates that notch tips can be responsible for lowering the experimental result. The shear strength for specimens $[0/0]_s$ and $[0/90]_s$, before being submitted to hygrothermal conditioning, exhibited a decrease in the shear strength of 8.6 and 5.2%, respectively, in relation to the unconditioned specimens. The influence of moisture is most notably present in the epoxy resin.

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