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Latin American Journal of Solids and Structures 5 (2008) 157–170

Study of the hygrothermal effects on the compression strength of carbon tape/epoxy composites

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Abstract

The wide range of composite material applications results in an almost inevitable contact with liquids and vapors, either organic or aqueous, which can affect both the immediate and the long-term performance of the material. In this study, the influence of the hygrothermal effects on the compression strength of carbon unidirectional tape/epoxy 8552 composites ([0/0]s) has been investigated. The moisture absorption was monitored placing the samples for a long time (until saturation) in a humidity-controlled chamber (80 ° C and 95% relative humidity (RH)) and other samples in a salt spray chamber (36 ° C and 95% RH). The compression tests were carried out in two temperatures: the room temperature (22 ° C) and the high temperature (82 ° C). It was observed that the samples tested at room temperature and submitted to the salt spray chamber presented a decrease of 8% on the compression strength when compared with the non-conditioned specimens. On the other hand, the samples tested at high temperature and submitted to the humidity-controlled chamber presented a decrease of 25.7% on the compression strength, when compared with non-conditioned samples. However, the specimens submitted to the salt spray chamber presented a decrease of 18.5% on the compression strength when compared with non-conditioned specimens at high temperature. These results showed that the combined effect of the humidity with the high temperature test, decrease the compression strength of the composite materials.

Keywords: polymeric composites, hygrothermal effects, compression strength, salt spray.

1 Introduction

The polymeric composites, when placed in service, are exposed to a variety of ambient conditions and different types of mechanical loading. The main atmospheric agent causing s ambient attacks are the temperature, the relative humidity of air, the effect of ultraviolet radiation, the chemical exposition, the saline water and the contact with hydraulic fuels, gases and fluids. Thermosetting polymeric composites must perform under both environmental conditional combined with deformations and mechanical stresses [4].

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The environmental effect caused by temperature and relative humidity of air can be reversible, when the period of exposition is of short duration. However, when the exposition occurs in long cycles, where the combination of humidity with temperature change is present, the produced effect can be irreversible due to the water affinity with specific functional groups of polymeric matrices of polar nature. In this case , generally destructive alterations in the reinforcement/polymeric matrix interface occur due to the degradation of the physical and chemistry interactions between the resin and the fiber. In consequence, the displacement of fibers can occur, causing the delamination of the composite with the reduction of the mechanical and thermophysical properties of the composite material [1–4].

The temperature conditioning in polymer matrix composites may result in significant reductions in matrix dominated mechanical properties such as interlaminar shear, compressive and flexural strength.

Therefore, temperature affect the same mechanical properties affected by humidity conditions. Moreover, determinate temperature facilitates moisture absorption and might also provide paths by which air may reach the carbon fibers, resulting in either oxidation of fibers or degradation of the fiber-matrix interface Considerable efforts have been made to determine the effects of moisture on the mechanical properties of organic matrix composites [1–3,5–14,16,18]; also, there is a significant literature of the hygrothermal effect on the mechanical strength of composite laminates [15]. However, the problem of determining the effect of salt, temperature and humidity has not yet been addressed. The present work aims to contribute with the study of the mechanical behavior of thermosetting polymeric composites submitted to the humidity-controlled chamber and to the salt spray chamber.

2 Materials and methods

2.1 Materials

The material used in this study is prepreg of carbon/epoxy produced by Hexcel Composites and supplied by EMBRAER (Empresa Brasileira de Aeronáutica S.A.) in the form of continuous unidirectional tape. This material is commonly used in the manufacturing of aircraft primary structure. The used prepreg material has the 8552 modified epoxy resin system. The reinforcement of the prepregs used in this work was the IM7 carbon fiber of high resistance, with density of 1,75 g/cm^3, maximum deformation of 1.4 % and previously treated for a better chemical compatibility with the epoxy resin system.

The cure cycle used for the composite consolidation was suggested by the prepreg supplier. This procedure involved 4 steps. Firstly, the heating from room temperature up to 116°C at 2.5^{o}C/min, after an isothermal period in this temperature for 60 min; heating to 181°C at 2.5^{o}C/min and finally an isothermal period of at least 120 min.

The samples had been divided in three families with six specimens for each: the first one was kept in the room condition of the laboratory, called as dry or non-conditioning, the second

family was conditioned in a humidity-controlled chamber and the third was conditioned in a salt spray chamber until the humidity saturation. Later compression strength tests have been performed in two conditions: at room temperature (22 ± 3^oC) and high temperature (82 ± 1^oC). The tests carried out at high temperature aim to comply with the requirements recommended by the standard MIL-HDBK-17 – 02 [5], considered the most critical condition for the polymeric composite test.

2.2 Hygrothermal conditioning

The samples were submitted to two different hygrothermal conditionings: in a humidity-controlled chamber and in a salt pray chamber. The moisture absorption was controlled by weight changes of traveler specimens (representative specimens used to measure the mass gain) according to procedure B of ASTM D 5229/D5229 M-04 [3]. In this study the moisture absorption was monitored placing the samples for nine weeks (until saturation) in a humidity-controlled chamber at 80 ° C and 95% of relative humidity (RH). The mechanical test specimens were kept in the environmental chamber until saturation. The specimens were then removed from the environmental chamber and tested at room temperature.

The conditioning in salt spray chamber was carried out according to ASTM 117 – 03 [2] to verify the strength of the sample in saline condition for a period of nine weeks. The traveler samples and the test samples were suspended between 15° and 30° in a parallel vertical line, which guarantees no contact between them inside the chamber. The main direction of the mist flow in the chamber is horizontal. The saline solution was prepared by the dissolution of 5 sodium chloride parts (w/w %) in 95 distilled water parts, with pH in the range of 6.5 to 7.2. The samples were submitted to the temperature of 35 ° C.

2.3 Compressive strength

Compression tests were carried out according to the IITRI experiment (developed for the Illinois Institute of Technology Research Institute), which follows the ASTM D 3410/D 3410M-03 [1], in a universal machine of test MTS with load cell of 100kN. This device uses a relatively small sample with bearing and steel columns to guarantee the alignment of the loading. The grips in the form of wedges are lodged in machined sockets (Fig. 1).

3 Results

3.1 Moisture absorption

The humidity increment can be gravimetrically measured through the change in the weight of the polymeric composite. The content of humidity absorbed for the families of unidirectional composites of carbon fiber IM7 impregnated with epoxy resin 8552 is presented in Tab. 1, as resulted of the weight average gain weekly in the traveler samples.

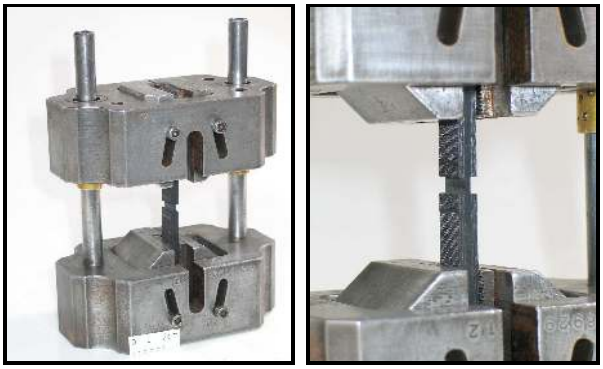

Figure 1: Device of compression test used.

It is observed, in Fig. 2, that in the initial period of exposition, it practically exists a linear relation between the absorption of humidity and the square root of the time. For short periods of time, the humidity content increases linearly until reaches a known state as pseudo-equilibrium, approximately between three and four weeks of exposition. This period of pseudo-equilibrium is practically the same for the majority of the thermosetting polymeric composites. Therefore this is characteristic of a behavior of humidity absorption according to the Fick's diffusion law. The water remains free in the composite and it tends, with the time, to penetrate in the resin through the concentration gradient. Above of this linear portion, the humidity absorption starts to confer a concave format in the curve indicating a positive shunting line of the state of Fick's pseudo-equilibrium. With the continuous exposition, the process of humidity absorption becomes slower, and many authors attribute to this period, the beginning of the process of relaxation of the polymeric chain [9–14, 16–18]. For the composites submitted to the humidity-controlled chamber and salt spray chamber it was observed a weight average gain between 0.3-0.4%.

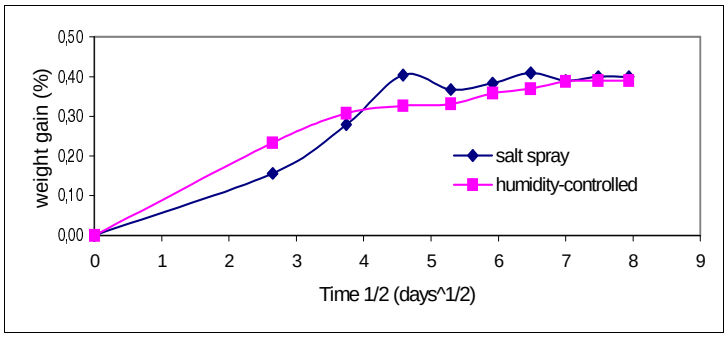

Figure 2: Weight average gain of the composite submitted to the humidity-controlled and the salt spray chambers.

Table 1: Results of humidity absorption in the composites submitted to the humidity-controlled and the salt spray chambers.

Average mass gain (%)		
Time	Conditioning	
	Salt Spray Chamber	Environmental Chamber
1st week	0.16 ± 0,128	0.23 ± 0,004
2nd week	0.28 ± 0,009	0.31 ± 0,014
3rd week	0.40 ± 0,008	0.33 ± 0,011
4th week	0.37 ± 0,152	0.33 ± 0,039
5th week	0.38 ± 0,157	0.36 ± 0,014
6th week	0.41 ± 0,180	0.37 ± 0,010
7th week	0.39 ± 0,180	0.39 ± 0,012
8th week	0.40 ± 0,120	0.39 ± 0,013
9th week	0.40 ± 0,090	0.39 ± 0,009

3.2 Compressive strength

The way in which composite materials absorb moisture depends upon factors such as temperature, fiber volume fraction, reinforcement orientation, fiber nature (that is, permeability, polarity, density), area of exposed surfaces, diffusivity, and surface protection [16, 17]. Moisture penetration into the composite materials is conducted by one major mechanism, namely, diffusion. This mechanism involves the direct diffusion of water into the matrix and in a much less extent into the fibers. The other common mechanisms are capillarity and transport through microcracks and voids. The capillarity mechanism involves the flow of water molecules along the fiber/matrix interface, followed by diffusion from the interface into the bulk resin. Transport of moisture by microcracks and voids involves both flow and storage of water into the microcracks and other forms of microdamages [9].

The strength of the interface determines how much of the applied stress can be transferred to the load-bearing fibers. This strength is largely determined by the contact area between fibers and matrix and the level of adhesion at the contact points. However, it is not clear how the interface should be tailored to optimize composite properties and whether optimum adhesion is desirable or essential to achieve the best balance of mechanical properties [4, 5, 7, 13, 15].

Table 2 presents the results of the compressive strength tests, at room and elevated temperatures, of the samples submitted to the hygrothermal conditioning and the salt spray chamber. The result has been obtained using six samples for each family that were depicted in Figures 3 and 4.

Table 2: Compressive strength results.

Room temperature tests	Maximum (MPa)	**Minimum (MPa)**	**Average (MPa)**
Non-conditioned	1841.7	1770.0	**1812.2 ± 30.6**
Salt spray chamber	1850.4	1538.7	**1667.8 ± 107.3**
Elevated temperature tests	Maximum (MPa)	**Minimum (MPa)**	**Average (MPa)**
Non-conditioned	1587.5	1461.7	**1519.1 ± 52.6**
Hygrothermal chamber	1159.9	1072.6	**1129.0 ± 32.5**
Salt spray chamber	1304.1	1179.9	**1237.8 ± 36.9**

Despite of the data dispersion, it was observed in Tab. 2 and Fig. 3, that the composites tested at the room temperature (22°C), previously submitted to the salt spray chamber (1667.8 ± 107.3MPa), presented the reduction of 8.0% on the compressive strength, when compared with the non-conditioned specimens (1812.2 ± 30.6MPa).

According to the literature, the moisture absorbed in the carbon fiber/epoxy composites may reduce the laminate strength and stiffness due to the matrix plasticization and/or degradation of fiber/resin interface. Plasticization reduces residual stresses and increases viscoelasticity. Exposure to water may also cause some decrease in cross link density [3]. Such effects are usually recoverable on drying. On the other hand, when water within the system, combined with external or internal stress (due to loading or thermoelastic effects), cause internal damage (matrix cracking, interfacial debonding or chemical degradation of the fibers) the composite undergoes permanent weakening [6].

The interface and the effects of environmental conditions on its strength play a major role in determining the strength of composite laminates in the presence of moisture. Madhukar e Drzal [17] studied experimentally the effect of fiber/resin adhesion in the interlaminar shear of graphite fiber/epoxy composites. The fiber/matrix adhesion was changed using the same fiber submitted to different surface treatments. These modifications increase the interlaminar shear strength by factor superior to two, without changing the fiber's properties. Imaz et al [15] studied a carbon fiber reinforced epoxy resin composite material, analyzing the humidity absorption kinetics under different hygrothermal conditions together with the kinetics variation as a function of moisture and temperature variables. The authors concluded that the absorbed moisture as a function of time for all wet environmental treatments studied has a Fickian variation. The moisture sorption implies a clear drop of mechanical properties of the materials, such as flexural

and compressive stress.

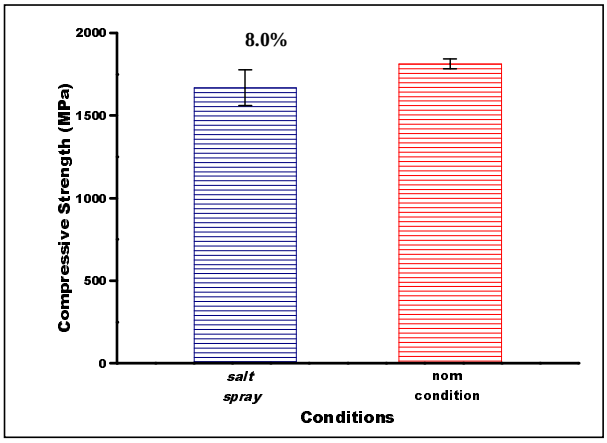

Figure 3: Compressive strength values at room temperature.

The sample compression strength results (Tab. 2 and Fig. 4), tested in elevated temperature (82°C), show that the composites submitted to the humidity-controlled chamber (1129.0 $\pm$ 32.5MPa) presented a decrease of 25.7% in the strength, when compared with the specimens non-conditioned (1519.1 $\pm$ 52.6MPa). The literature confirms this behavior [15]. On the other hand, the composites submitted to the salt spray chamber (1237.8 $\pm$ 36.9MPa) presented a reduction of 18.5% on the compressive strength in comparison with the non-conditioned composites.

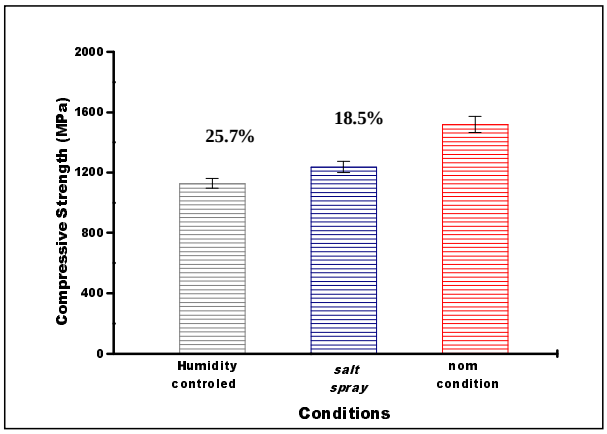

Figure 4: Compressive strength values at elevated temperature.

The compression strength results of the composites tested at room and elevated temperatures (Tab. 2 and Fig. 5) show that non-conditioned composites tested at elevated temperature (1519.1 $\pm$ 52.6MPa) presented a decrease of 16.2% when compared to the specimens tested at

room temperature (1812.2 ± 30.6MPa). Once again, this fact indicates that the temperature has influence on the composite mechanical properties, as shown by Tang and Springer [15]]. Comparing the results of the samples submitted to the salt spray chamber and tested at room temperature (1667.8 ± 107.3MPa) with the values of the specimens tested in elevated temperature (1237.8 ± 36.9MPa), it was observed that the composites tested at high temperature show a reduction of 25.8% on the strength.

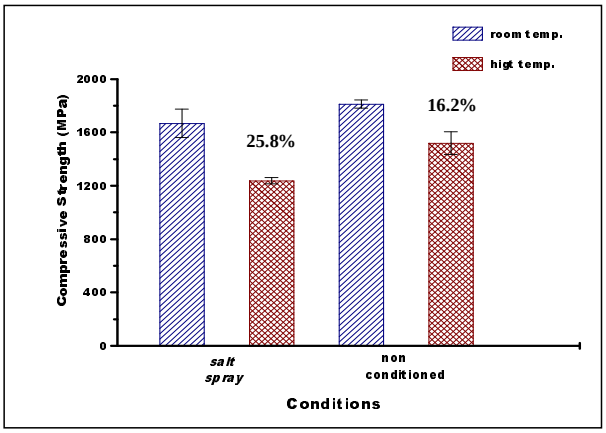

Figure 5: Compressive strength values at room and elevated temperatures.

3.3 Failure mechanisms

Due to the anisotropy of the construction of the laminates, the process of failure in the thermosetting composites submitted to the loading in compression is very complex. Different failure mechanisms can occur and they are influenced by four main factors: the fiber characteristics, the polymeric matrix behavior, the angle lamination and the properties of the fiber/matrix interface.

Figure 6 presents representative photos of the samples tested in compression. All specimens present kink band failure. It is observed that the failure modes observed for the laminates submitted to the room conditioning are very similar to those ones observed for the specimens submitted to the humidity conditionings. It is also observed that the failure is perpendicular to the applied load, revealing interlaminar and translaminar failures.

The different environmental conditionings cause changes on the resin physical and chemical characteristics and on the fiber surface. The decrease on the compressive property indicates composite degradation. The fractography analyses were carried out to elucidate what are the probable causes that contribute to the decrease of the compressive strength.

The effects of moisture on the fracture surfaces of the specimens were examined by scanning electron microscopy (SEM). Figures 7 to 9 present a typical fracture surface of fractured com-

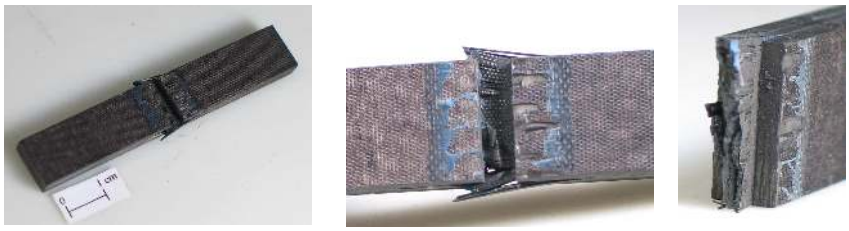

Figure 6: View of the samples after the compression test.

pression strength dry specimen of carbon/epoxy laminate. The smooth clean surface of fibers is caused by a fracture micromechanism involving an interfacial debonding in the carbon/epoxy fabric laminate. Cracks through a fiber tow, either in the warp or fill direction, can also be identified as a fracture micromechanism. The crack runs along the matrix between fibers. This feature suggests a brittle fracture micromechanism. In this material (carbon/epoxy) the microcracks initiated at the matrix at the interface growing along parallel planes under the shear load.

The transversal section analysis shows the occurrence of crushing (Fig. 7) and the significant presence of matrix residues on the failure surface, due to the carbon fiber rigidity and the fragile characteristic of the epoxy resin. Figure 8 presents the failure surface of the unidirectional carbon fiber composite with a clear fragile fracture, due to the overload for compression that leads to the fiber breaking in a fragile way (without plastic deformation). Franco [11] shows that this type of fracture involves possible aspects of flexure in the top of fibers. Aspects of cusps in the matrix are also observed (Fig. 9), revealing that the resin remains adhered to the fibers in large extension, disclosing a good interface between fibers and resin. The majority of the failure surfaces of the specimen is presented damaged, due to the compression effect, which promotes the surface crushing.

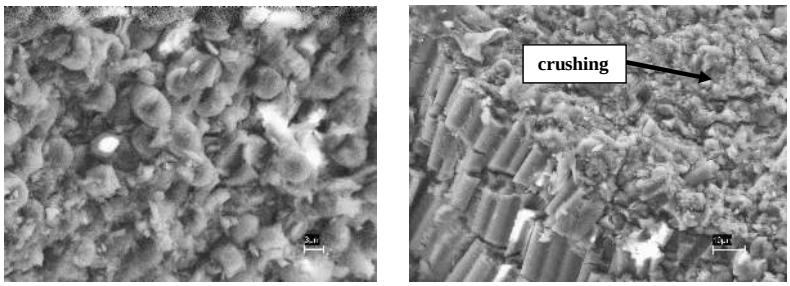

Figure 7: Micrograph of non-conditioned sample after compression test, at room temperature.

Figure 9 has been shown a decrease in compression strength for the carbon/epoxy laminates after the environmental conditioning. The water absorbed by the epoxy laminates, in general,

causes reversible plasticization of the matrix and lowers its glass transition temperature. Combined with temperature effects, these factors cause significant changes in the matrix toughness affecting the laminate strength. Moreover, humidity and temperature cause dimensional changes and induce stresses in the laminate that degrade the fiber matrix interface.

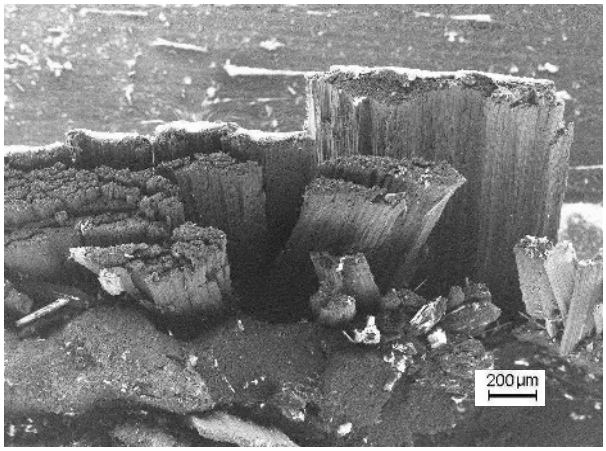

Figure 8: Surface fracture of non-conditioned sample after compression test, at room temperature.

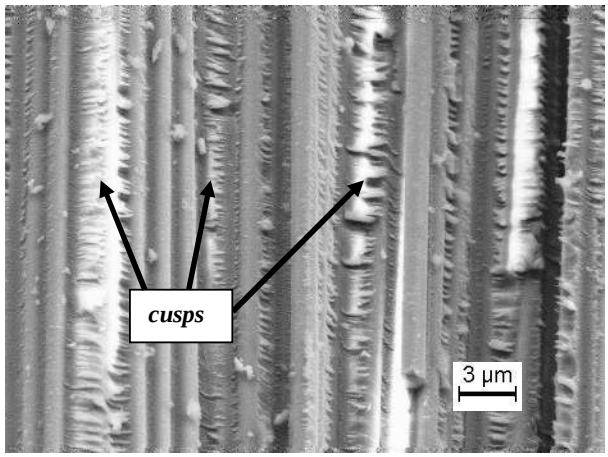

Figure 9: Longitudinal section of non-conditioned sample after compression test, at room temperature.

Fracture surfaces of carbon/epoxy laminate conditioned in environmental chamber, showed a similar fracture micromechanism observed for the dry specimen. However, the moisture absorption induced an increase of the interfacial debonding area, as shown in Figure 10. The effect

of debonding along the warp/fill yarn interface supersedes the effect of interfacial (fiber/matrix) debonding. Large debonding along resin rich areas resulted in significant degradation of the material toughness.

Figure 10 shows a typical aspect of the composite submitted to the hygrothermal chamber and tested at elevated temperature, presenting a fragile fracture promoted by the overload in compression. Figure 11 presents the longitudinal section of the same sample (failure area), disclosing a larger amount of fibers without resin, indicating that the hygrothermal conditioning affected the fiber/resin interface and/or promoted the matrix degradation. Figure 12 is representative of the composite submitted to the salt spray chamber and tested at elevated temperature, which shows a significant amount of matrix residues that difficult the analysis of the failure region.

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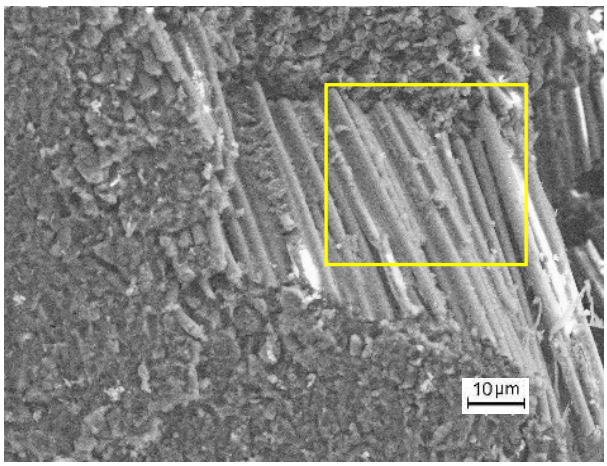

Figure 10: Transversal section of the fracture surface of the sample submitted to the hygrothermal chamber and tested in elevated temperature.

Moisture absorption can also induce swelling deformation. This phenomenon is almost linear with respect to the absorbed amount of moisture. When the specimens are removed from the environmental chamber in the beginning of the hygrothermal conditioning process and submitted to the compression test, the fibers provide some lateral restraint in the transverse direction of the polymer composite specimens, but they do not restrain the swelling of the matrix along the loading direction. This effect is reflected in very low longitudinal deformation in the polymer composite specimens. This restraining effect is necessarily offset by internal stresses within the matrix phase (mainly compression) and along the fiber/matrix interfaces (shear) [13].

Moisture penetration through the fiber/matrix interfaces causes also interfacial debonds causing the rupture or degradation of the interface, exposing the carbon fibers, as depicted in

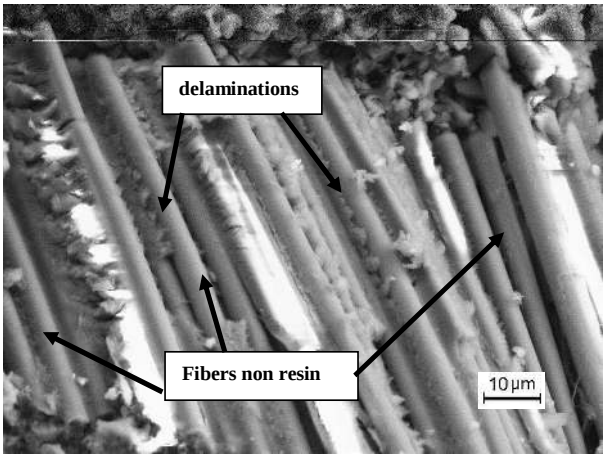

Figure 11: Longitudinal section of the fracture surface of the sample submitted to the hygrothermal chamber and tested in elevated temperature.

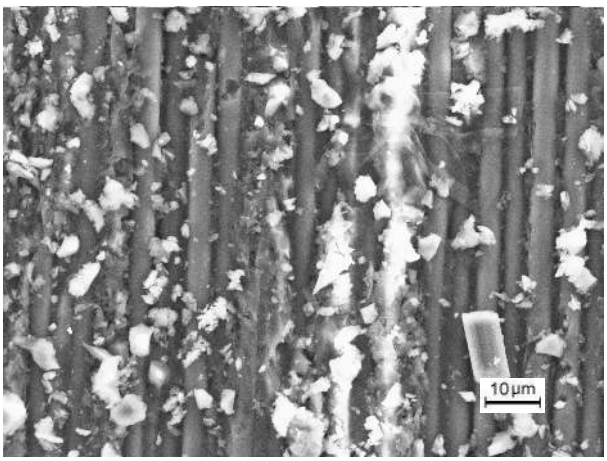

Figure 12: Longitudinal section of the fracture surface of the sample submitted to the spray chamber and tested in elevated temperature.

Figures 10-12. The absorption of the water molecules accelerated by temperature used in the hygrothermal conditioning weakens the fiber/matrix interface exposing the fibers.

It has been shown that the sorbed moisture acts as a plasticizer and a crazing agents into the thermoset resins. Absorption of moisture induces also swelling of the matrix. Because the swollen matrix forms a shell around the dry matrix, the dry matrix will be placed under a state of triaxial stresses. When the swelling stresses reach a critical value, the fracture of the matrix occurs. This mechanism can lead to the rupture of the fiber/matrix interface. Knowledge of the mechanisms that drive moisture sorption, as well as the influence of sample dimensions, temperature, and relative humidity, becomes crucial when long-term properties of the material are needed.

4 Conclusion

The study of the hygrothermal effect on the compressive strength of unidirectional carbon fiber impregnated with resin epoxy 8552 composites showed that the polymeric matrix is severely affected in the tests at high temperature.

The composites non-conditioned and tested at the room temperature present a decrease of 16.2% in relation to the tested at high temperature. On other hand, the samples conditioned in chamber of salt spray and tested at the high temperature show a reduction of 25.8% in the resistance in relation to the samples tested at room temperature. This fact confirms that the effect of the high temperature of test reduces the compressive strength, due to degradation of the polymeric matrix and/or the fiber/matrix interface.

Acknowledgements. The authors are indebted with SENAI-SP, EMBRAER, FAPESP 02/01808-7 and CNPq (303528/2003-6).

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