ACCELERATED ENVIRONMENTAL AGING IN HYBRID COMPOSITE WITH SYNTHETIC AND NATURAL FIBERS

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Abstract. The use of natural fibers as reinforcement in polymeric composites presents some advantages compared to synthetic fibers, such as glass fibers. The main are the low cost, low density, good tenacity, good thermal properties, biodegradability and reduced use of instruments for their treatment or processing. However, their mechanical performance is quite inferior to synthetic fibers hindering its use in structural applications. In this sense, hybrids composites, comprising natural and synthetic fibers have physical, thermal and mechanical properties quite different. This is very important when the composite is exposed to adverse environmental conditions (humidity, high temperature and UV rays), once each fiber will have a different response. This work reports the effects of accelerated aging on the tensile mechanical properties of two composites; a hybrid comprising glass-E and natural (curaua) fibers and a typical glass-E composite, both reinforcing orthophthalic polyester resin. The composites were tested in the original condition and after accelerated aging (UV rays and steam heated water in alternate cycles). It was also carried out fracture characteristics analysis of the testpieces. In the original condition the results of the hybrid composite were close to glass-E composite. After aging they presented different behaviors. The aging was more harmful to mechanical properties of the not glass-E composite.

Keywords: Accelerated aging, natural fiber, hybrid composite, mechanical properties.

1. INTRODUCTION

Fibers derived from renewable sources have stand out in the development of new materials, particularly the natural fibers used as reinforcement in polymeric composites. Natural fibers composites can be used in many industrial sectors, such as civil construction, furniture production and automotive industry. Their advantages include low cost, low density, high availability, biodegradability, etc (Rowell et al. 1997, Sudeell et al. 2002, Bledzik and Gassan, 1999). The main drawbacks are poor mechanical properties and high moisture absorption due to its hydrophilic nature. The humidity absorption can be an imperative problem when it is combined with heat, solar radiation, pollution and static or fatigue loadings. In this case the material can present degradation of the mechanical and physical properties.

The poor mechanical performance of the natural fibers composites can be improved by addition of small amounts of synthetic fiber, such as glass fiber, forming a hybrid composite. Besides to improve the mechanical properties, the glass fiber can act as a chemical barrier preventing the contact of the natural fibers with the water and reducing the water absorption of the composite (Thew et al. 2002, Seena et al. 2002, Kalaprasad and Kuruvilla, 1997).

In general, the works regarding hybrid composites highlight the cost reduction of the composite (low cost of the natural fiber), improvement of the mechanical properties, humidity absorption decrease and increase of the environmental aging resistance compared to the natural fiber composites.

Degradation is any destructive reaction that can be caused by chemical, physical or mechanical agents. The degradation causes a progressive deterioration of the composite properties. According to Woo et al. (2008) ultraviolet radiation (UV) and humidity exposure are amongst the most severe weathering conditions that can result in harmful effects on the mechanical properties of many polymers. At room temperature and light absence the polymers are stable for long time. However, under the sunshine the polymer oxidation is accelerated and this effect is increased by the pollutant atmospheric.

Natural fibers can present degradation due to biological agents, acid and alkaline environments, moisture absorption, UV radiation and temperature (Mohanty et al. 2000). Glass fibers are relatively inert, immune to the biological attack and with good resistance to environmental aging when compared to the natural fibers. In the case of the composites, the degradation can affect the polymeric matrix, fibers and even the interface causing interaction loss between matrix and fiber and disturbing the mechanical integrity of the composite.

The study of the composites degradation can be accomplished using natural or accelerated aging tests. In the natural aging test the composite is exposed to conditions close to real use condition. Usually, the samples are exposed in benches with inclination returned for the magnetic north (Silva, 2002). The test demands long exposition time so that the degradation factors can act in the material. The accelerated aging test presents the advantage of the speed, supplying data of the probable performance of the material during its useful life. These tests are usually realized in a chamber where are simulated the use conditions of the material, however with high intensities to accelerate the degradation process. The process can be monitored by the changes in the physical state and mechanical properties of the material. The conventional mechanical tests (tensile and flexural) and the thermal analyses techniques are quite useful for this purpose.

This work reports the effects of accelerated aging on the tensile properties of two composites; a hybrid comprising glass-E and natural (curaua) fibers and a typical glass-E composite, both reinforcing orthophthalic polyester resin. The composites were tested under original condition and after accelerated aging. The aging conditions were UV rays and steam heated water in alternate cycles. It was carried out tensile test and fracture characteristics analysis of the specimens.

Curaua fiber was selected for this work due to its good mechanical performance in relation to other natural fibers, such as jute and sisal, and also for social aspects (Leão et al. 2001, Oliveira, 2005). Curaua fiber, which originated in the Brazilian Amazon, represents a strong potential for agriculture because it offers an alternative to tropical forest exploitation, and also contributes to the relocation of man back to the countryside. Curaua fiber is rarely mentioned in scientific literature and the few references found are restricted to symposiums and journals within the national scope (Razera et al. 2008, Revista Agroamazonia (2004), Trindade (2000), Mothé and Araújo, 2004). No references regarding the association of curaua and glass fibers were found.

2. EXPERIMENTAL

The hybrid composite is a laminate with seven layers. The matrix is an orthophthalic polyester resin reinforced by glass-E fibers (mats of 450 g/m²), and curaua fibers (mats of continuous fibers).

Curaua fiber was supplied by Embrapa of the Amazon (Brazilian Enterprise of Agronomy Research). Mats of continuous fibers were manufactured manually. Firstly, the fibers were cut with exactly 65 cm in length, separated in strands and weighed. The approximate weight of each strand was 1.2 g that corresponds to 0.9 dtex. The strands were aligned and fixed with an adhesive tape forming a mat of continuous fibers with 65 cm length and 95 cm width. Curaua fiber properties can be found elsewhere (Silva et al. 2008, Oliveira (2005), Trindade (2000), Leão et al. 2001).

The hybrid composite was manufactured by a local industry utilizing the hand-lay-up technique. It is composed of four glass-E fibers layers alternated with three layers of continuous curaua fibers. The configuration is showed in Fig. 1. The final dimension of the plate was 95 x 65 x 0.56 cm. The external layers are of glass-E fibers what facilitates the lamination process and assures a more uniform thickness. A non-hybrid composite (60 x 30 x 0.67 cm), with seven layers of glass-E fibers mats (450 g/m²) was also manufactured using the same technique.



Figure 1. Configuration of the hybrid composite.

The density of the composites was determined according to ASTM D792-91. The obtained values were 1.39 and 1.47 g/cm³ for the hybrid and glass-E composites, respectively. The glass-E fibers volume fraction was determined by a burning test which obtained the value of 12.6 and 17.6% for the hybrid and glass-E composites, respectively. This low

volume is in agreement with the manual lamination process. In the burning test both resin and curaua fibers are burned and only the glass fibers remains, this way it is not possible to determine the volume fraction of the matrix, curaua fibers and voids separately. The volume fraction of matrix and voids of the glass-E composite were 78.2 and 4.4%, respectively.

Half of the tensile specimens were conditioned in an accelerated aging chamber. This chamber was built following recommendations of ASTM G53-96 and literature (Saron, 2000). The specimens were exposed to two conditions: 18 h of UV rays (lamps emitting UVA e UVB) and 6 h of steam heated water (95 % of relative humidity) in alternate and independent cycles until reaching 2016 h. Only one face of the specimen suffers the degradation effects as recommended by ASTM G53-96. The temperature in the interior of the chamber was 60 ± 3 °C during the UV radiation cycle and 61 ± 5 °C during the water steam cycle. The room temperature was 29 °C.

Tensile tests were carried out for aged and unaged specimens. Tests were carried out at room temperature according the ASTM D3039-00 and a minimum of eight specimens were tested for each condition. In the case of the hybrid composite the fibers are oriented in the loading direction. After the tests it was realized the fracture characteristics analysis through optical microscopy and scanning electronic microscopy (SEM).

To facilitate the understanding the following denomination will be used for the composites: GC (glassfiber composite), HC (hybrid composite), AGC (aged glassfiber composite), AHC (aged hybrid composite).

3. RESULTS AND DISCUSSION

3.1. Tensile test in the original condition

Stress x Strain curves for both composites are shown in Fig. 2. In general, the curves presented almost linear behavior up to fracture. This behavior is typical of composites with thermoset matrices and synthetic fibers, such as glass/epoxy. In fact, the behavior is close to matrix due to its high volume fraction.

The tensile mechanical properties are presented in Fig. 3. The properties of the hybrid composite were very close to that of the glass-E composite. This result can be considered excellent once that the glass-E fibers were partially substituted by natural fibers whose mechanical properties are inferior. Of course, this behavior is also related to the use of natural fibers oriented to loading direction. In this situation, the load transfer matrix/fiber is maximized.



Figure 2. Stress x Strain curves from the tensile test of the composites in the original condition.

Figure 4 shows some specimens after the tensile test. The fracture was quite localized, which means, the specimen did not show damage evidence in the areas distant from the fracture. Figure 5 shows a fracture surface (hybrid composite) where it is easily identified the layers of glass and curaua fibers. The fracture is flat (in macroscopic way) and it wasn't observed delamination between the layers.

These characteristics are very important in the composite evaluation, once they show a good adherence between the layers of glass and curaua fibers despite the strong mismatch (in the mechanical and physical properties) between them. All this is directly related to interlaminar stress distribution (between layers) what depends of the composite configuration. Similar behavior was observed for glass-E composite what was not surprised once the same one is just formed by fiberglass mats.



Figure 3. Tensile mechanical properties of the composites in the original condition. (a) Tensile strength, (b) Young's modulus, (c) Elongation.



Figure 4. Specimens after the tensile test.



Figure 5. Fracture surface of a tensile specimen (hybrid composite).

3.2. Tensile test after aging

Tensile properties of the aged and unaged composites are showed in Tab. 1, as well as the percentage comparison unaged/aged properties. After aging the glass-E composite (AGC) presented decreased of the tensile strength and elongation; however there was an expressive increase of the Young's modulus (17.8 %). The tensile strength decrease is mainly related to matrix degradation on the external layers of the composite once the glass fibers are relatively inert (Segovia et al. 2001). It is important to say that some fibers sticking out of the surface (in the face that suffer the UV aging) losing its reinforcement function. The Young's modulus increase and elongation decrease is due to the matrix brittleness after UV exposure.

Composite	Tensile Strength	Young Modulus	Elongation
	(MPa)	(GPa)	(%)
GC	93.89	2.21	4.0
AGC	87.34	2.69	3.12
%	- 7%	+ 17,8 %	- 22 %
HC	92.15	2.34	3.68
AHC	71.53	2.31	3.06
%	- 22.4 %	- 1.3 %	- 16.9 %

Table 1. Tensile properties of the aged and unaged composites (%, comparison unaged/aged properties)

According to Rabek (1995) and Kumar et al. (2002), mechanical degradation leads to excessive brittleness by chain crosslinking that can result in microcracking, but would also increase the tensile strength. Chain scission can also happen so that the molecular weight and strength of the polymer are diminished. This way, the load bearing capability as well as the long term durability are affected.

From Tab. 1 one can see that the aging was more harmful to HC composite than to GC composite, mainly the tensile strength that showed a reduction of 22.4 %. Probably the degradation reached the internal curaua fibers layers that are highly sensitive to UV radiation and humidity, mainly the humidity. Therefore, the largest reduction of the AHC tensile strength it is probably related to humidity absorbed by natural fibers. The Young's modulus of the AHC didn't show relevant alteration. In this case the stiffness increase caused by matrix brittleness was compensated by degradation of the natural fibers.

The HC configuration had a fundamental role in its degradation resistance. The natural fibers layers are internal and relatively protected from direct action of environmentally. Otherwise the degradation would be worse.

Figure 6 shows some specimens (AGC and AHC) after the tensile test. The fracture was quite localized and the face of the composite that suffer the UV aging shows exposed glass fibers due to matrix degradation. The fracture is flat and it was not observed delamination between the layers (the same was observed to unaged composites, GC and HC).



Figure 6. AGC and AHC specimens after tensile test. It is observed exposed glass fibers in the face of the composite that suffered the aging.

Figure 7 shows the face exposed to aging of two specimens of the AHC and AGC composites, respectively. In the AHC composite one can observe some depressions evidencing the fibers loss due to matrix degradation (Fig. 7a). Figure 7b shows fiberglass bunches without adherence to matrix characterizing the adhesive fracture due to matrix degradation of the AGC composite. Some bubbles are also observed in the Fig. 7b (black arrows).

The specimens fracture surfaces of the AGC and AHC composites are showed in Fig. 8. In both composites exposed fibers can be observed in larger number and length characterizing fiber/matrix debonding followed by pull-out mechanism. The "holes" (white arrows) were caused by fiber pull-out. In Fig. 8b it is also observed some glass and curaua fibers that were broken without sliding (black arrows). No delamination (between the layers) was found.



Figure 7. Specimens after aging (the face exposed to aging). a) AHC composite with depressions evidencing the fibers loss. b) AGC composite with fiberglasses bunch without adherence to matrix.



Figure 8. Fracture surfaces of the AGC (a) and AHC (b) composites. White arrows indicate fiber pull-out and black arrows indicate fiber fracture without sliding.

Figure 9 presents the fracture region of an AGC specimen where transverse cracks are observed along its whole length. The number of these cracks increases with the test progression. As they are perpendicular to the load direction, they are typical of tensile test (Scida et al. 2002). These cracks were not observed in the unaged composites.

Figure 10 presents a micrograph of the fracture region of an AHC specimen. Microcracks perpendicular to the loading direction are observed. In this case, the crack propagates through the curaua/matrix interface not breaking the curaua fiber. It is also observed fiber/matrix debonding.



Figure 9. Final fracture region of an AGC specimen.



Figure 10. Photomicrograph of the final fracture region of an AGC specimen (200X).

4. CONCLUSIONS

In the original condition the properties of the hybrid composite were very close to that of the glass-E composite. This result can be considered excellent once that the glass-E fibers were partially substituted by natural fibers whose mechanical properties are inferior. This behavior is also related to the use of continuous fibers oriented to loading direction as well as to configuration of the laminated hybrid.

After aging the GC composite presented decreased of the tensile strength and elongation; however there was increase of the Young's modulus. The tensile strength decrease was mainly related to matrix degradation on the external layers of the composite. The Young's modulus increase and elongation decrease was due to the matrix brittleness after UV exposure.

The aging was more harmful to HC composite than to GC composite. There was tensile strength and elongation decrease and the Young's modulus didn't show relevant alteration. The degradation probably reached the internal curaua fibers layers that are highly sensitive to UV radiation and humidity, mainly the humidity.

For both composites and conditions (aged and unaged) the fracture was quite localized. The specimens didn't show damage evidence in the areas distant from the fracture and it was not observed delamination between the layers. These characteristics show a good adherence between the layers of glass and curaua fibers despite the strong discrepancy between them.

After aging the composites showed exposed glass fibers in the face that suffered the UV aging due to matrix degradation. Transverse microcracks perpendicular to the load direction were also observed.

5. ACKNOWLEDGEMENTS

The authors acknowledge the financial support provided by CNPQ and the mechanical tests accomplished by CT-GÁS (Natal/RN/Brazil).

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7. RESPONSIBILITY NOTICE

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