WATER ABSORPTION IN UNSATURATED POLYESTER/CAROÁ COMPOSITES: THE EFFECTS OF THE SIZE AND TEMPERATURE

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Abstract. Studies on composites reinforced with natural fibers show that are sensitive to influences from environmental agents such as water. The moisture causes degradation of mechanical properties of natural fiber reinforced composites to a larger extent when compared to synthetic fiber reinforced composites, as a consequence of the higher moisture sorption behavior, and the organic nature of the natural fibers. In this sense the purpose of this work is to study water absorption in unsaturated polyester composites reinforced with caroá fiber. The composites had weight compositions 30% caroá/70% unsaturated polyester with dimensions of 20x20x3 mm³ and 20x20x6 mm³. Water absorption tests were conducted by immersing specimens in a distilled water bath at 25, 50 and 70°C and the water uptake was followed gravimetrically for different elapsed time. Results of moisture content distribution inside these composites are shown and analyzed. Moisture induced degradation of composite samples was significant at elevated temperature

Keywords: composite, vegetable fiber, water sorption, experimental, caroá

1. INTRODUCTION

In recent years there has been a fast growth in using renewable vegetable fibers as reinforcements in composite materials. Reinforced plastics made with cellulosic materials, as fillers are low cost, light-weighted, have enhanced mechanical properties, and are free from health hazards.

Despite the attractiveness of vegetable fiber reinforced polymer matrix composites, they display lower mechanical properties and relatively poor moisture resistance compared to synthetic fiber reinforced composites.

Several studies show that mechanical properties improve significantly in vegetable fiber reinforced composites with high levels of fiber content (Carvalho *et.al.*, 2002; Joseph *et al.*, 2002; Wessler *et al.*, 2002; Dantas *et al.*, 2003). Their good environmental and mechanical properties, coupled to high synthetic fiber prices, resulted in an increased use of natural fibers as polymer reinforcers (Medeiro *et al.*, 2003). However, natural fibers show a weak adhesion on the polymer matrix-fiber interface, due their hydrophilic nature and to the hydrophobic nature of most polymers (Li et al., 2000; Wambua et al., 2003; Aziz et al., 2005).

Fibre moisture makes impregnation more difficult and leads to weak interface and fiber-matrix adhesion that causes internal tensions, porosity and the premature failure of the system. All these factors limit the field of application of vegetable reinforced polymer composites (Bledzki *et al.*, 1996; Clark and Ansel, 1996; Joseph and Carvalho, 1999; Carvalho *et al.*, 1999; Cavalcanti *et al.*, 2005).

Nevertheless, several works are reported in the literature about the development, mechanical properties and water sorption of polymer/natural fiber composites reinforced with jute, sisal, cotton, hemp, flax, pineapple, coir, and ramie fiber (Rao *et al.*, 1981, Sensarzadeh *et al.*, 1984; Sensarzadeh *et al.*, 1985; Sensarzadeh, 1986; Marcovich *et al.*, 1999; Soni and Soni, 1999; Cavalcanti *et al.*, 2001; Sreekala *et. al.*, 2002). In our group, we have started studies on the viability of using other regionally relevant vegetable fibers for these purposes and Caroá (neoglaziovia variegatta) is one of them.

In this work, the water absorption of compression molded unsaturated polyester/caroá fibers as a function of sample dimensions and temperature is reported. The driving force behind this study is the fact that the amount of water absorbed by a sample depends on its composition, dimensions, temperature and exposure time and also because although several works on the kinetics of water sorption and/or moisture polymer/natural fiber composites are reported in the literature, we have not found reports on the system investigated here.

The present work deals with the development of new polymer composites reinforced with short fibers from Caroá (*Neoglaziovia variegata*), from the Cariri region of Paraiba state (Brazil). The mechanical behavior of these systems as a function of fiber content was conducted and the water absorption as a function of sample dimensions and temperature is reported.

2. EXPERIMENTAL PROCEDURES

Unsaturated polyester was supplied by Resana S.A. and cured with 1% MEK supplied by VI Fiberglass S.A. Caroá fibers (Figure 1) were obtained at a farm in Pocinhos-PB, Brazil.The fibers were washed with running water, allowed to air dry at room temperature, combed and cut up to 5 cm.

Composites with varying fiber contents were compression molded. A fiber mat was produced by randomly distributing a pre-determined amount of fibers in a steel mold (220x180x3mm²) and compressing with 2 ton for 5 min at room temperature. The mats were removed from the mold for further use. An appropriate quantity of resin was mixed with the catalyst and a small amount was poured onto the mold. The fiber mats were placed in steel molds (220x180x3mm² or 220x180x6mm²) impregnated with more resin and the mold was closed (8 ton). The system was allowed to cure under pressure for 4h at room temperature before the composite plate was removed from the mold. The composite plates obtained were post-cured in an oven at 50°C for 48h. Tests for mechanical properties were conducted according to ASTM standards D-3039 for tensile and D-256 for impact.

For the water absorption test composites samples 20x20x3mm³ and 20x20x6mm³ were cut-off from the plates and their borders were sealed with resin to avoid water transport by capillarity.

The water sorption of the pre-dried (105°C to constant weight) samples was performed by immersion in water baths at 25, 50 and 70°C (Figure 20 and the water uptake was followed gravimetrically.



(a)



Figure.1. Caroá plant and fibers



Figure.2. Samples of the composite used in this work

3. RESULTS AND DISCUSSION

3.1. Morphology of Caroá fiber

The scanning electron micrographs (SEM) of caroá fiber are shown in Figure 3. The fibers exhibit fibrillar structure of long chains of molecules composed of cellulose and lignin with poliosis acting as ligands, with typical characteristics of vegetable fibers (Figures 3a-b). We can see the macrofibrilas (Figure 3c)united together by lignin and poliosis so as to form continuous filament in the direction of the length of the fiber which provides the stiffness of the caroá fiber.



(a)

(b)



Figure 3. Micrographs of the caroá fibers a) (× 900), b) (× 200) and c) (×3000).

3.2. Mechanical Properties

Figure 4 show the results of tensile strength of the composites as a function of fiber content. It is verificad that tensile strength is lower that of the matrix for loadings up to 25% by weight for Caroá fibers. This behavior could be attributed to inefficient loading, so the fibers act as defects (stress concentrators) and effectively weaken the matrix, resulting in a lower composite mechanical strength. At low fiber content the matrix is not sufficiently anchored and high deformations imposed on it lead to a break of the matrix-fiber bond. As the loading increases, stresses are more uniformly distributed and the composite's strength increases.

At loadings above 40% by weight, the tensile strength of the composites decreases with a further increase in fiber content, which can probably be attributed to fiber agglomeration and poor impregnation, leading to an increase in their effective diameter, a decrease in their aspect ratio and void formation.

Figure 5 illustrates the effect of the fiber content on the elastic modulus of the composites. The results show that the inclusion of fibers increases the stiffness of the composite. This behavior was expected, since the strength and modulus of the fibers used are higher than the matrix. In addition, fiber slow the movement of polymer chains resulting in an increase in of elasticity modulus of the composite with fiber content (d'Almeida, 1987).

Figure 6 display impact resistance as a function of fiber content. Impact resistance increases with fiber content increases at all loading levels for type of fiber tested. In that case the caroá fiber is able to diverge crack propagation and delay breakage, thus increasing impact strength. It is worth noticing that impact properties are not as dependent on fiber/matrix adhesion as the tensile strength and, in fact, benefit from looser interfaces.



Figure 4. Tensile strength as a function of caroá fiber content.



Figure 5. Young's modulus as a function of caroá fiber content.



Figure 6. Impact resistance as a function of caroá fiber content.

Figure 7 shows the elongation of the composite as function of fiber content. It is verified that the incorporation of fiber increases the properties of elongation to fiber content above 23%. This is attributed to the greater stiffness of the fiber, increasing the stiffness of the composite. In general, the addition of fiber reduces the elongation at break in relation to the matrix mainly if the fiber bend is less than the matrix



Figure 7. Deformation as a function of caroá fiber content.

3.3. Water Absorption

Water sorption curves as a function of dimensions, temperature and exposure time of unsaturated polyester/caroá composites with 30% w/w fiber contents are shown in Figure 8. The data indicates water absorption to increase with fiber loading and to be higher than the matrix. While the water sorption in equilibrium for the unsaturated polyester is about 1%, those of the composites were around 15%. These results are coherent with those reported in the literature (Rao *et al.*, 1981; Marcovich *et. al.*, 1999; Cavalcanti *et al.*, 2001; Sreekala *et al.*, 2002) for similar systems and which attribute the increase in water sorption of vegetable fiber reinforced polymer composites to the hydrophilic nature and permeability of this type of reinforcement.



Figure 8. Effect of sample dimensions in the water sorption of caroá fiber reinforced unsaturated polyester composites

The curves depicted in Figure 9 also evidence that, despite the fact that the water sorption of all composites was shown to increase with the solid area/volume ratio (Table1) and temperature, this behavior was more pronounced at the lower experimental temperature. In the Table 1, Me represents the equilibrium moisture content, So/Vo is the initial area/volume relationships and Tw is water temperature.

Table 1 Moisture content and geometric data effects on the water sorption at equilibrium (M_e) of matrix and composites with 30% w/w fiber contents.

Sample	Thickness	T_{W}	Me	S _o /V _o
	(mm)	(°C)	(d. b.)	(m^2/m^3)
Matrix	3.00	25	1.255	433.3
Composites	3.00	25	14.49	433.3
	6.00	25	14.81	266.7
	3.00	50	15.16	433.3
	6.00	50	16.07	266.7
	3.00	70	15.61	433.3
	6.00	70	16.52	266.7

Figures 9a-b shows water sorption of unsaturated polyester/caroá fiber composites as a function of temperature and exposure time for thickness a) 3mm and (b) 6mm. The results indicate that, as expected, the water uptake of the composite sample submerged in a water bath at 70°C was faster than under the other (25°C and 50°C) experimental conditions. This behavior is attributed to the increased water mobility within the solid with increasing temperature. It is believed that higher water temperatures lead to thermal dilation of the composites and to increased composite porosity which would, in turn, cause quick moisture migration.

Other aspects such as water migration by capillarity in micro cracks inside the solid, mainly in the fiber-matrix interface where adhesion is of fundamental importance, was not analyzed.

4. CONCLUSIONS

Tensile strength of unsaturated polyester reinforced with caroá fiber increased at fiber loading levels above 25%, and with further addition of fiber up to a loading of 40% by weight. Elastic modulus of the composites show that the inclusion of fibers increases the stiffness of the composite. Impact resistance of the composites is higher than that of the matrix at all fiber content levels and that the incorporation of fiber increases the properties of elongation to content above 23% fiber.

The water absorption of the composites to increase with fiber loading and to be higher than that of the matrix. Higher water diffusion was obtained for the first stages of sorption, and the rate of sorption decreased at longer water immersion times. Sample thickness effects were pronounced at lower temperature and temperature effects were more relevant than those observed for changes in area/volume ratio.

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(b)

Figure. 9. Effect of the water temperature in the water sorption of caroá fiber reinforced unsaturated polyester composites

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