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TENSILE, IMPACT, AND THERMAL PROPERTIES OF AN EPOXYNOVOLAC MATRIX COMPOSITES WITH CUBAN HENEQUEN FIBERS

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Composites based on an epoxy-novolac matrix and a Cuban henequen fiber reinforcement were investigated. A satisfactory reinforcing effect was observed for a composite material prepared by compression molding at room temperature using long random fiber mats. Tensile and impact experiments were run on the material, and its thermogravimetric analysis was performed. The breaking zone of the specimens tested in tension was examined by an electron sweep microscope. The results obtained are presented graphically and in photographs.

1. Introduction

The use of composite materials with a polymeric matrix and vegetable fibers as a reinforcement has increased in the last decades. Even though there have been several investigations into synthetic and semisynthetic biodegradable polymers [1-3], the conventional synthetics polymers, such as polyesters and epoxies, still remain the most used matrix materials in composites [4-6]. On the other hand, the popularity of substitution of glass and carbon fibers by natural, mainly

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Fig. 1. Dry Cuban henequen fibers.

vegetable ones is growing steadily. Their low cost and low density, together with the relatively high specific mechanical characteristics, offer a good renewable and ecofriendly alternative with a great number of applications [7, 8].

In the automotive and construction industries, which currently represent the most important sectors of employment of vegetable fiber-reinforced polymers, hemp, sisal, jute, cotton, and flax are generally used together with polyolefins, polystyrene, epoxy resins, and unsaturated polyesters. The intrinsic biodegradability of this alternative reinforcement increases the environmental friendliness of these materials [8, 9], which are suggested for lightweight structural parts, paneling elements in cars, temporary use in irrigation systems, or for various applications in furniture, sports, and leisure industries [10, 11].

It is well known that the main drawback of using vegetable fibers in reinforced polymers usually is the poor adhesion at the fiber-matrix interface due the different surface properties of the composite components. However, this drawback is not considered in this work, and natural fibers were used without any surface treatment.

The purpose of this work was to determine some mechanical properties of a Cuban henequen composite with epoxy-novolac as the polymeric matrix and to examine how they depend on the weight content of the natural fibers.

2. Experimental

2.1. Materials

The polymer used as the matrix was a Novola/Poliamina, PoliResinNovolac, epoxy resin developed and provided by the POLINOVA company of Rio de Janeiro, Brazil. This resin is a thermosetting polymer possessing good mechanical properties and a high resistance to some soft acids, alkalis, and solar light. The novolac groups added ensure its better coupling with natural fibers [12]. The use of polyamine as a curing agent allows the polymerization process at ambient temperature, convenient to avoid the degradation of vegetable fibers.

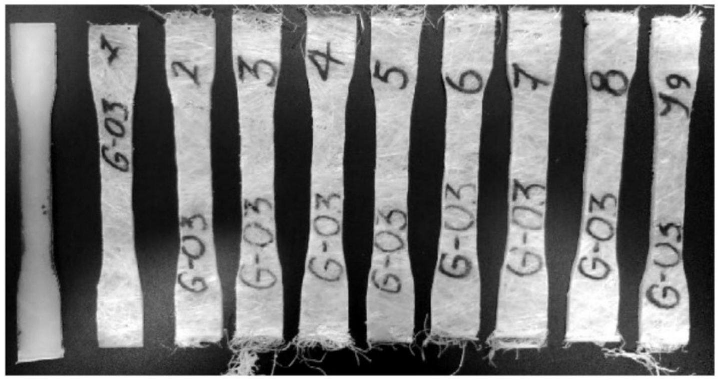

Fig. 2. Typical tension specimens of Cuban-henequen-reinforced epoxy-novolac composites.

TABLE 1. Critical Degradation Temperatures and Residues

Sample	T_{min}, °C	T_{max}, °C	Residues, wt.%
Pure resin	361,01	395,58	5,09
Henequen fibers	289,39	373,94	12,94
Composite with 9 wt.% fibers	355,97	398,89	5,37
Composite with 24 wt.% fibers	351,27	379,91	7,52

As a reinforcement, fibers of the Cuban henequen plant, traditionally employed for ropes and tissues, were used. They had an average length of 87 mm, an average length/diameter ratio of 289.4 and are shown in Fig. 1. The fibers were used without any surface treatment.

Composite plates were made using the cold molding process in closed cases. For comparison purposes, a neat resin plate and another one with a low percentages of fibers were also manufactured. We used the standard procedure described in [13]. From the plates obtained, test specimens were made (see Fig. 2).

2.2. Characterization methods

2.2.1. Mechanical measurements. The tensile properties of the composites were determined using an INSTRON 5567 universal tension machine. The specimens were conditioned at room temperature for 24 h before testing. Crosshead speeds of 1, 5, and 10 mm/min were used. Seven specimens of each sample were tested. Izod impact tests were also performed, using a CEAST impact machine, on unnotched specimens (60 × 12 × 3 mm) in accordance with ASTM D256-10.

2.2.2. Thermal analysis. Thermogravimetric (TGA) measurements were performed, using a TA Instruments High-Resolution 2950 TGA thermogravimetric analyzer, for material samples of weight between 10 and 20 mg placed in a platinum pan. Samples of fibers, matrix alone, and two samples of composites with 9 and 24 wt.% fibers were tested. The analysis was performed at a heating rate of 10°C/min from room temperature to 700°C under a N_2 flow.

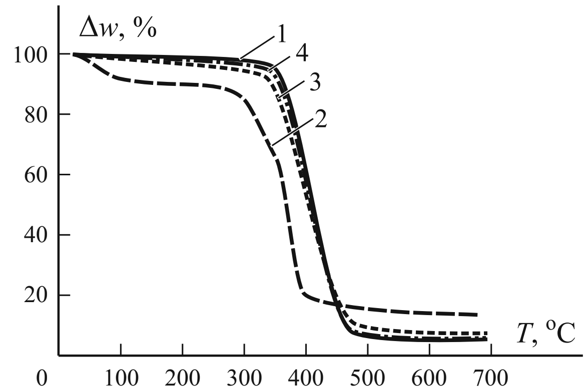

Fig. 3. TGA results as weight loss Δ*w* vs. temperature *T* for the pure resin (1), henequen fibers (2), and 24 (3) and 9 (4) wt.% composites.

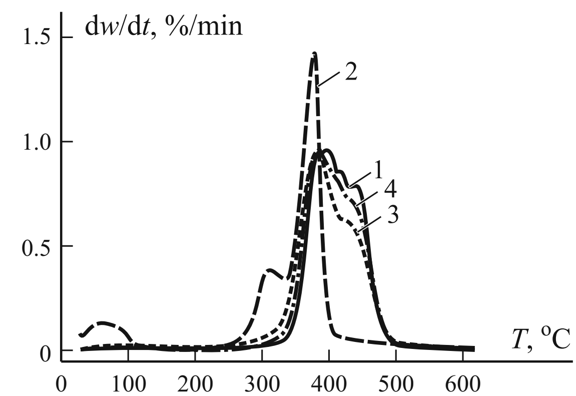

Fig. 4. DTGA data as *dw*/*dt* vs. temperature *T* for the same materials. Designations as in Fig. 3.

2.2.3. Microscopy. SEM observations were carried out using a Zeiss Digital scanning Microscope. The specimens were previously metalized with gold using an Emitech K560 metallizer. The fracture zones of specimens broken in tension were examined.

3. Results and Discussion

The general results of TGA are given in Table 1. As is seen the differences between results for the pure resin matrix and composites with different percentage of fibers are insignificant. Figure 3 shows that fibers caused an initial decrease in weight close to 10 wt.%, which was associated with loss of the water absorbed from the environment by the hygroscopic polysaccharide fiber components, followed by a single-step thermal degradation phenomenon centered at T_{max} around 395°C, which agrees with findings of other studies on similar fibers [5, 14]. The behavior of the pure matrix and the two composite samples differed slightly, see Fig. 4.

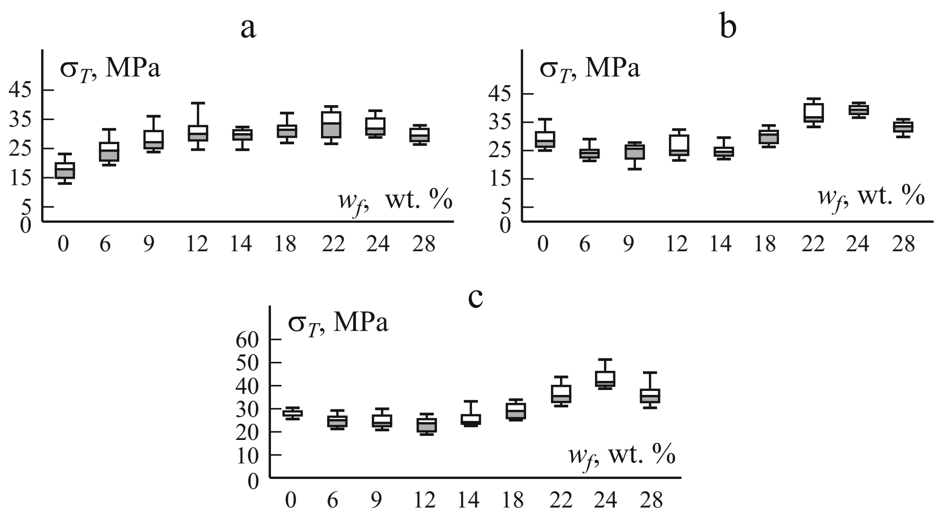

Fig. 5. Tensile strength σ_T of composite vs. weight content w_f of henequen fibers at deformation rates of 1 (a), 5 (b), and 10 (c) mm/min.

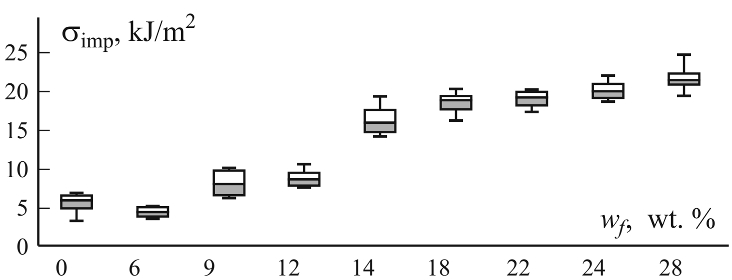

Fig. 6. Impact strength σ_{imp} of composite vs. weight content w_f of henequen fibers.

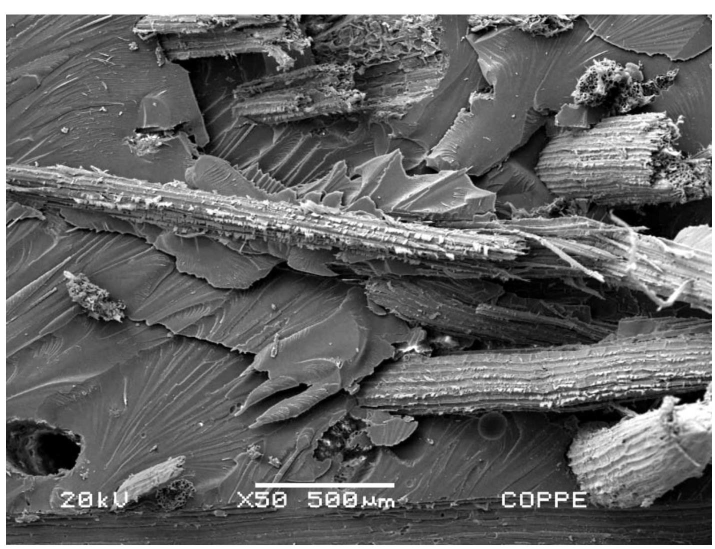

Fig. 7. SEM photograph of the fracture zone at a 50× magnification of a composite specimen with 18 wt.% henequen fibers broken in tension. Matrix breaks are clearly seen.

a b

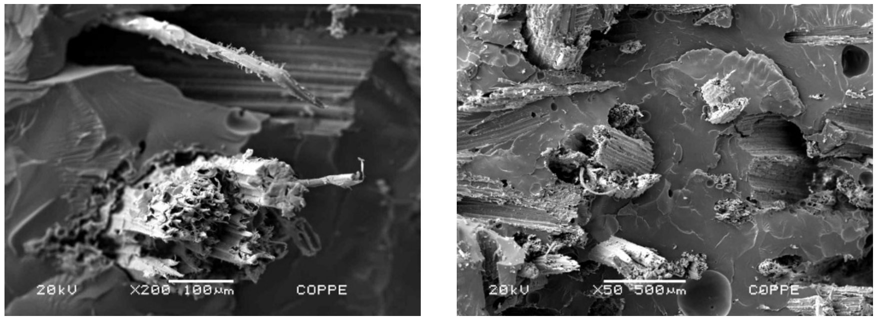

Fig. 8. SEM photographs of the fracture zone at 200× (a) and 50× (b) magnifications of a composite specimen with 18 wt.% henequen fibers broken in tension. Fibers both broken and pulled out from the matrix can be seen.

The possibility of reinforcing the epoxy-novolac blend with Cuban henequen fibers was verified. Figure 5 shows the results of tensile tests of composite specimens with different fiber concentrations deformed at rates of 1, 5, and 10 mm/min. At the three deformation rates, the maximum increments were reached in the cases with 22 and 24 wt.% fibers. In previous works, the best results were achieved at these or higher fiber concentrations [3, 15]. The increase in the tensile strength, compared with that of pure matrix, was 187.8% at the deformation rate of 1 mm/min and 22 wt.% fibers and 138% at 5 mm/min and 153.22% at 10 mm/min and 24 wt.% fibers.

At the highest fiber concentration, 28 wt.%, the tensile strength decreased, perhaps because of the reduced cohesive capacity of matrix. A similar effect was also observed in previous studies. For example, Sreenivasan [6] detected a decrease in the tensile and impact strengths at 40 wt.% fibers. Sitticharoen in [16] used bagasse fiber ash, untreated and treated with a solution of 1.0 wt.% silane (vinyltrimethoxysilane) as a coupling agent, detected a slight decreases in the tensile strength beyond the point of optimum of concentration of treated and untreated fibers, probably caused by the decreasing interfacial area in the matrix as the filler content increased.

The impact test was performed on a CEAST impact machine in accordance with ASTM D256-10. Ten unnotched specimens with different fiber concentrations were prepared and tested for the impact strength. Figure 6 shows results obtained, and they a similar to those found in other studies [15].

SEM was used to analyze the fracture surface of specimens broken in tension. In Fig. 7, the three main fracture modes of the composites are seen: pulled-out of fibers from the matrix, break of fibers, and break of matrix, which are typical of composite materials [1, 17].

Conclusion

It was found that the reinforcement of an epoxy-novolac resin with long Cuban henequene fibers increased its tensile and impact strengths. The highest tensile strength exhibited the composites with 22 and 24 wt,% of the fibers. The growth in the impact strength, on the whole, was directly proportional to the fiber percentage, except at the minor percentage of 6 wt.%. As a results it was concluded that this type of composite can be used for low-loaded products where the economy of reinforcement is required.

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REFERENCES

1. A. K. Bledzki, A. Jaszkiewicz, and D. Scherzer, "Mechanical properties of {PLA} composites with man-made cellulose and abaca fibres," Composites: Part A, Appl. Sci. Manufact., **40**, No. 4 404-412 (2009).
2. A. L. Duigou, C. Baley, Y. Grohens, P. Davies, J.-Y. Cognard, R. Crachcadec, and L. Sohier, "A multi-scale study of the interface between natural fibres and a biopolymer," Composites: Part A, Appl. Sci. Manufact., **65**, 161-168 (2014).
3. T. Gurunathan, S. Mohanty, and S. K. Nayak, "A review of the recent developments in biocomposites based on natural fibres and their application perspectives," Composites: Part A, Appl. Sci. Manufact., **77,** 1-25 (2015).
4. C. Gonzalez Murillo and M. P. Ansell, "Mechanical properties of henequen fibre and epoxy resin composite material," Proc. XIV Int. Conf. Mechanics of Composite Materials, Riga, Latvia, Vol. 1, p. 61 (2011).
5. I. M. D. Rosa, C. Santulli, and F. Sarasini, "Mechanical and thermal characterization of epoxy composites reinforced with random and quasi-unidirectional untreated phormium tenax leaf fibers", Materials & Design (1980-2015), **31**, No. 5, 2397-2405 (2010).
6. V. Sreenivasan, D. Ravindran, V. Manikandan, and R. Narayanasamy, "Mechanical properties of randomly oriented short sansevieria cylindrical fibre/polyester composites," Materials & Design, **32**, No. 4, 2444-2455 (2011).
7. E. Nassiopoulos and J. Njuguna, "Thermo-mechanical performance of poly(lactic acid)/ax fibre-reinforced biocomposites," Materials & Design, Pt B, **66**, 473-485 (2015), Lightweight Materials and Structural Solutions for Transport Applications.
8. A. Kiruthika, "A review on physico-mechanical properties of bast fibre reinforced polymer composites," J. of Building Engineering, **9**, 91-99 (2017).
9. Z. Azwa, B. Yousif, A. Manalo, and W. Karunasena, "A review on the degradability of polymeric composites based on natural fibres," Materials and Design, **47**, 424-442(2013). doi:http://dx.doi.org/10.1016/j.matdes.2012.11.025.
10. J. L. Changduk Kong and Hyunbum Park, "Study on structural design and analysis of ax natural fiber composite tank manufactured by vacuum assisted resin transfer molding," Materials and Design, **130**, 21-25 (2014).
11. K. Pickering, M. A. Efendy, and T. Le, "A review of recent developments in natural fibre composites and their mechanical performance," Composites: Part A: Appl. Sci. Manufact., **83**, 98-112 (2016), Special Issue on Biocomposites.
12. W. Jinyan, L. Rui, and J. Xigao, Handbook of Epoxy Blends, Springer Int. Publ., 1-29 (2016).
13. A.-A. Rodríguez-Soto, L.-M. Sanabio-Alves-Borges, J.-L. Valín-Rivera, D. Díaz-Batista, "Fabrication of a composite with epoxy matrix and henequen long fibers as reinforcement and with random orientation," Ingeniería Mecánica, **18**, No. 2, 122-128 (2015).
14. A. R. Martin, M. A. Martins, L. H. C. Mattoso, and O. R. R. F. Silva, "Caracterização química e estrutural de fibra de sisal da variedade agave sisalana," **19**, 40-46 (2009).
15. H. O. Maurya, M. Gupta, R. Srivastava, and H. Singh, "Study on the mechanical properties of epoxy composite using short sisal fibre," Proc. 4th Int. Conf. Materials Processing and Characterzation, **2**, No. 4, 1347-1355 (2015).

16. W. Sitticharoen, A. Chainawakul, T. Sangkas, and Y. Kuntham "Rheological and mechanical properties of silica-based bagasse-fiber-ash-reinforced recycled HDPE composites," Mech. Compos. Mater., **52**, No. 3, 421-432 (2016).
17. K. H.-R. M. Ramesh and K. Palanikumar, "Comparative evaluation on properties of hybrid glass fiber- sisal/jute reinforced epoxy composites," Procedia Engineering, **51**, 745-750 (2013).