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Evaluation of Dynamic Mechanical Properties of PALF and Coir Fiber Reinforcing Epoxy Composites†

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The growing interest in natural lignocellulosic fibers (NLFs) is associated not only with environmental benefits but also with technical, economical and social advantages. Among the NLFs, the coir fiber and pineapple leaf fiber (PALF) are low-cost and widely available waste materials. The present work investigated the dynamic mechanical properties, by means of DMA tests, of these two different fibers as reinforcement of epoxy matrix composites. The influence on DMA properties of composites by the volume fraction and the configuration form, mat or aligned coir fiber, were also evaluated. The results showed higher E' and E" moduli values and an increase in T_g with the incorporation of PALF in epoxy matrix, which indicate a superior dynamic mechanical properties for this composite in comparison to the neat epoxy resin. The experimental data also revealed a stronger interfacial interaction of PALF/epoxy when compared to coir fiber/epoxy. It was also found a lower tan δ value exhibited by higher volume fractions of aligned coir fibers in epoxy matrix.

Keywords: *PALF, Coir fiber, epoxy composites, DMA.*

1. Introduction

Due to an increasing environmental awareness, several studies have, in the past decades, been performed on natural lignocellulosic fibers (NLFs) as reinforcement of composites in substitution for synthetic fibers[1-11]. Among these NLFs, both the pineapple leaf fiber (PALF) and the coir fiber are extracted from leftover parts of corresponding plant and so considered as waste materials. This makes them interesting fibers to be used in composites, not only because of environmental sustainability as biodegradable and renewable resources, but also for economical reasons[12,13]. However, the use of NLFs in composites instead of synthetic fibers has its own drawbacks. NLFs exhibit a great dispersion in their properties, depending on several factors, such as age, chemical composition, diameter, and non-uniform dimensions[6,11]. Hence, the understanding of the correlation between the structure and properties is essential for the development of NFLs as reinforcement of new composites. Many thermal and mechanical techniques could be employed for this purpose. In particular the dynamic-mechanical analysis (DMA), especially with temperature variation, has been used to determine relevant viscoelastic properties of epoxy composites[14-16]. This analysis enables one to follow the elastic and viscous response of the material by varying the frequency, load or temperature[17]. The dynamic-mechanical properties are generally represented by the storage modulus (E'), loss modulus (E") and damping factor (tan δ) as a function of time and temperature. Furthermore, the DMA allows one to associate the viscoelastic behavior with macroscopic properties and it also provides relevant information about fiber/matrix interaction[18,19].

The evaluation of the dynamic mechanical behavior of composites reinforced with natural fibers has been the subject of several works[20-29]. In all these works it was found that the incorporation of a natural fiber into a polymer matrix tends to sensibly increase E' and E" as well as to promote a reduction in tan δ. As for the PALF and coir fiber, dynamic mechanical parameters of their epoxy composites have not yet been investigated. Therefore, in order to compare the dynamic mechanical properties of these two distinct fibers, PALF and coir, reinforcing epoxy composites, DMA tests were performed. Epoxy matrix was used due to its good properties such as, environmental, chemical and thermal resistance. It can also be cured at room temperature, which makes it attractive for several applications[30]. Moreover, the polar epoxy and hydroxyl groups may promote some interactions with lignocellulosic fibers, in spite of the hydrophobic character of this resin. An additional aim of this work was also to investigate how the volume fraction of coir fibers affects the dynamic mechanical properties of their epoxy composites.

2. Materials and Methods

For dynamic-mechanical analysis (DMA) samples were prepared with epoxy resin reinforced with two different natural lignocellulosic fibers. The pineapple leaf fibers (*Ananas comosus*), PALF for short, were supplied by Desigan Natural Fibers, Brazil, and the coir fibers extracted from the mesocarp of coconut (*Cocos nuciferaL.*) fruits were provided

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by "Coco Verde Reciclado", Brazil. The PALF was tested in continuous and aligned form, while the coir fiber was tested both in continuous and aligned form as well as a mat, which consists of random fibers, nonwoven and pressed. These untreated fibers were just cleaned and dried at 60°C in an air-oven. The composites were prepared by pressing the epoxy resin type diglycidyl ether of the bisphenol-A (DGEBA) mixed with the hardener triethylene tetramine (TETA) using the stoichiometric phr 13 and 30 vol % of aligned fibers or mat. Samples of pure epoxy resin were made as control. Figure 1 shows the specimens after cure at room temperature.

Three-point flexural mode was performed in a model Q 800 TA Instruments DMA equipment running at a frequency of 1 Hz under nitrogen atmosphere and heating rate of 5°C/min. Curves of loss modulus (E"), storage modulus (E') and tangent delta (tan δ) were recorded in the temperature range from -100°C to 180°C were recorded. Tests were also carried out for specimens with different volume fractions 10, 20 and 30 % of aligned coir fibers to evaluate the influence of the incorporated amount in the dynamic mechanical properties of composites.

3. Results and Discussion

The variation of storage modulus (E') as a function of temperature for the aligned/mat coir fiber and aligned PALF samples is given in Fig. 2. For comparison, the amount of fiber or mat was kept at 30 vol % for the different specimens. In this figure an abrupt modulus drop is observed at temperatures around 60-85°C. This zone corresponds to the respective relaxations in the polymer matrix associated with the glass transition temperature (T_g) from crystalline (rigid) to amorphous (rubbery) state[17].

The main result in Fig. 2 is that only the PALF composite exhibites improvement of its dynamic mechanical properties in comparison to epoxy resin up to 85°C, below the T_g. Indeed, this fiber produces high restriction on the molecular motions of the epoxy resin[20]. The decrease in E' for the coir mat composite, up to 60°C, is probably due to the low stiffness of the random distributed coir fiber, which tends to reduce the viscoelasticity of the epoxy matrix[5]. On the contrary, the incorporation of PALF in the epoxy composite increases the stiffness of the matrix[31]. This can clearly be seen in the loss

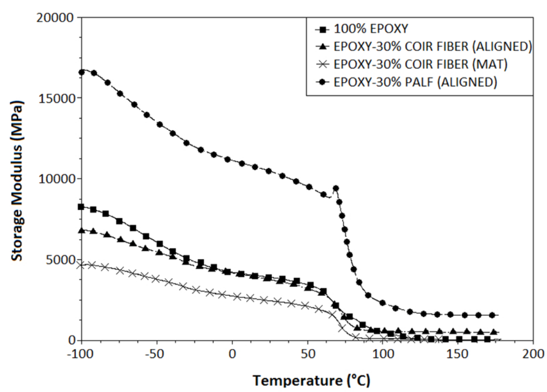

Figure 2. The variation of storage modulus as a function of temperature for the different composites

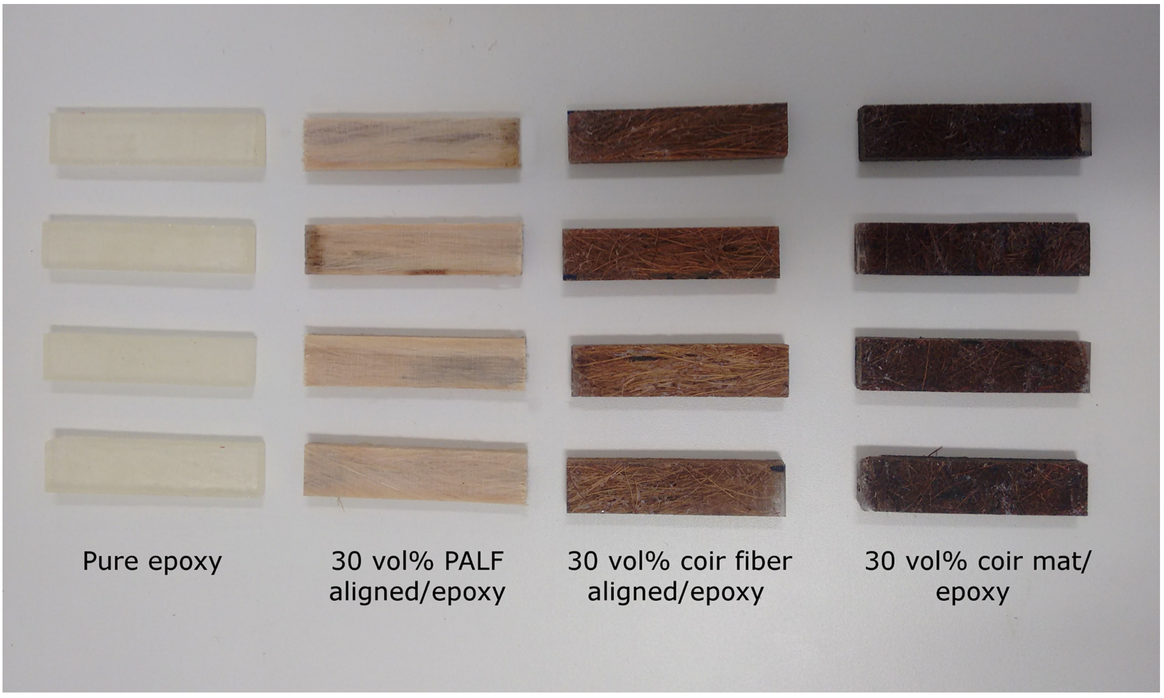

Figure 1. Specimens after cure at room temperature.

modulus curves, Fig. 3, which presents the highest E" peak for the PALF composite and lower values for both coir fiber and mat composites as compared to the neat epoxy. In Fig. 2, it should also be noted that the rubbery plateau, above 100°C, is slightly higher for the composites with both PALF and coir fibers aligned in relation to the neat epoxy resin. This indicates a reinforcing effect that improves the thermal mechanical stability of the material at high temperature, except for the coir mat composite.

Curves of tan δ for the composites reinforced with the different fibers are shown in Fig. 4. The results in this figure reveal a reduction in the damping capacity of all composites as compared to the neat epoxy. It is also observed a decrease in T_g values, given by the tan δ peak temperature. In other words, the incorporation of both coir fiber and PALF decreases the mobility of epoxy polymer chains and impairs their damping capacity[20,26]. By contrast, the decrease in tan δ is not so pronounced in the case of coir mat, probably due to the random distribution of fibers.

The effect of different volume fractions of aligned coir fiber incorporation in epoxy matrix was also investigated. Figure 5 shows the E" curves for the specimens reinforced with up to 30 % of continuous and aligned coir fiber. The results for the incorporation of 20 % and 30 % of coir fiber suggest a greater interaction with the polymer chains, which decreases the mobility and tends to increase the stiffness of the composite. Changes in the mechanical properties of coir fiber composites were also reported by Harish et al[32]. Moreover, a peak shift to higher temperature is observed in Fig. 5 for the composite with 30% of fiber incorporation. This behavior could be explained by the reducing in flexibility of the composite as a consequence of the interaction of coir fibers with the epoxy chains[20].

Figure 6 shows the tan δ variation with temperature for different volume fractions of aligned coir fiber reinforced epoxy composites. In this figure, it was identified peaks associated with the glass transition temperature, T_g, which are indicated in each curve. The reduction on the damping factor for epoxy composites with greater volumes of coir fibers was observed. This is related to the interaction between the coir fiber and matrix, reducing the ability of the epoxy to crystallize.

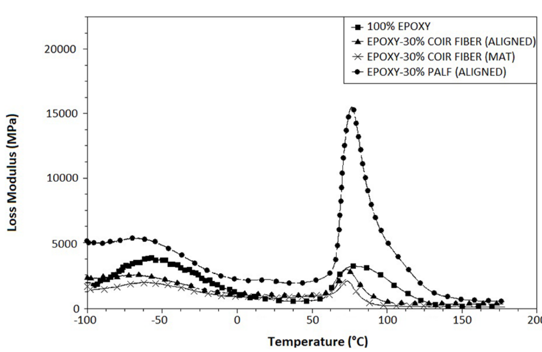

Figure 3. The variation of loss modulus as a function of temperature for the different composites.

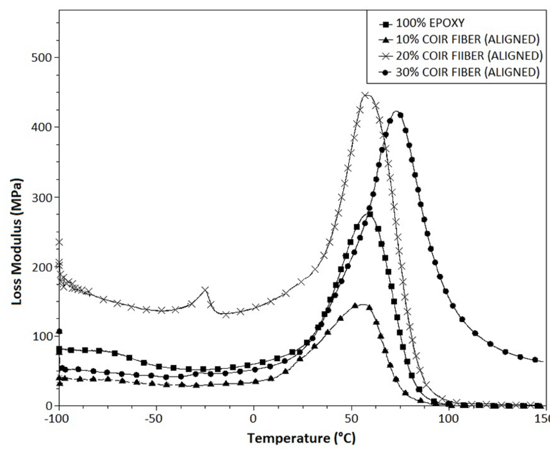

Figure 5. Effect of temperature on the loss modulus curves of the epoxy reinforced with different volume fractions of aligned coir fiber.

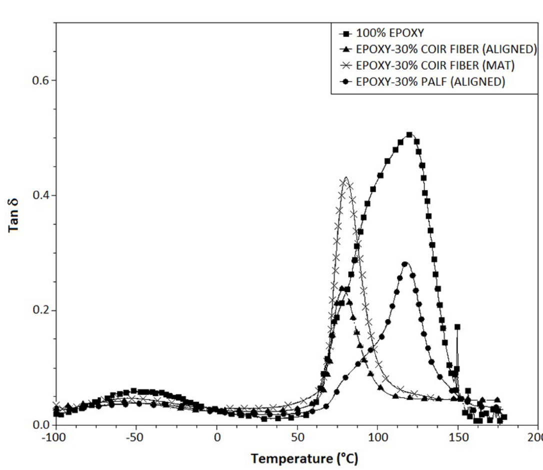

Figure 4. Effect of temperature on the tan δ curves of the composites reinforced with different fibers.

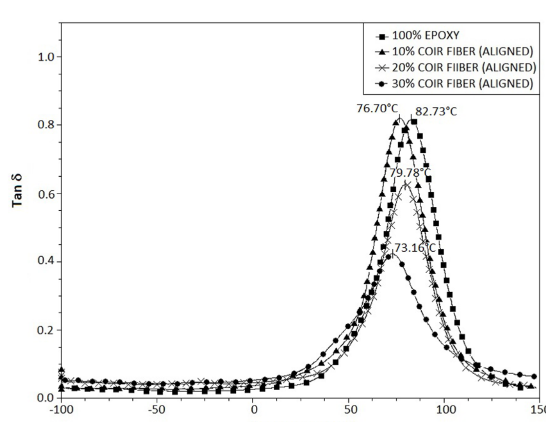

Figure 6. Damping curves for the epoxy reinforced with different volume fractions of aligned coir fiber.

4. Conclusions

- At high temperatures, DMA results showed a slight increase in the rubbery plateau for composites with both PALF and coir fibers aligned in relation to the neat epoxy resin. These composites display a reinforcing effect that improved the thermal mechanical stability above 100°C.
- The PALF incorporation increases the E', E" and T_g values due to the effective stress transfer from the epoxy matrix to the fiber up to about 60°C. This occurs as a consequence of the strong interfacial adhesion, which indicates superior dynamic mechanical properties for this composite in comparison to both coir fiber composites and the neat epoxy resin.
- The decrease in the T_g value and the highest damping factor attended by the coir mat composite reveals low restriction on the molecular motions of the epoxy resin and poor interfacial bonding of this composite.
- The low tan δ value and high glass transition temperature exhibited by higher volume fractions of aligned coir fibers in epoxy matrix suggests the increase of the stiffness of this composite, which was attributed to the interaction with the polymer chains.
- PALF/epoxy composite showed the highest reinforcing effect due to the increase of strength and stiffness verified in DMA curves. Therefore, these results emphasize the high potential of the PALF as reinforcement in epoxy composite and as a low-environmental impact alternative material.

5. Acknowledgements

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6. References

1. Pickering KL, Efendy MGA, Le TM. A review of recent developments in natural fibre composites and their mechanical performance. *Composites Part A: Applied Science and Manufacturing*. 2016;83:98-112.
2. Güven O, Monteiro SN, Moura EAB, Drelich JW. Re-emerging Field of lignocellulosic Fiber - Polymer Composites and Ionizing Radiation Technology in their formulation. *Polymer Reviews*. 2016;56(4):702-736.
3. Thakur VK, Thakur MK, Gupta RK. Review: Raw Natural Fibers-Based Polymer Composites. *International Journal of Polymer Analysis and Characterization*. 2014;19(3):256-271.
4. Thakur VK, Singha AS, Thakur MK. Ecofriendly Biocomposites from Natural Fibers: Mechanical and Weathering Study. *International Journal of Polymer Analysis and Characterization*. 2013;18(1):64-72.
5. Faruk O, Bledzki AK, Fink HP, Sain M. Biocomposites reinforced with natural fibers: 2000-2010. *Progress in Polymer Science*. 2012;37(11):1552-1596.
6. Monteiro SN, Lopes FPD, Barbosa AP, Bevitori AB, Silva ILA, Costa LL. Natural Lignocellulosic Fibers as Engineering Materials - An Overview. *Metallurgical and Materials Transactions A*. 2011;42:2963-2974.
7. La Mantia FP, Morreale M. Green composites: A brief review. *Composites Part A: Applied Science and Manufacturing*. 2011;42(6):579-588.
8. Monteiro SN, Lopes FPD, Ferreira AS, Nascimento DCO. Natural-fiber polymer-matrix composites: Cheaper, tougher and environmentally friendly. *JOM*. 2009;61(1):17-22.
9. John MJ, Thomas S. Biofibers and biocomposites. *Carbohydrate Polymers*. 2008;71(3):343-364.
10. Crocker J. Natural Materials. *Materials Technology*. 2008;23(3):174-178.
11. Satyanarayana KG, Arizaga GGC, Wypych F. Biodegradable composites based on lignocellulosic fibers - An overview. *Progress in Polymer Science*. 2009;34(9):982-1021.
12. Luz FS, Tommasini FJ, Nascimento LFC, Figueiredo ABS, Monteiro SN. Critical length and interfacial strength of PALF and coir fiber incorporated in epoxy resin matrix. *Journal of Materials Research and Technology*. 2018. Submitted.
13. Asim M, Abdan K, Jawaid M, Nasir M, Dashtizadeh Z, Ishak MK, et al. A Review on Pineapple Leaves Fibre and its Composites. *International Journal of Polymer Science*. 2015;2015:950567.
14. Niedermann P, Szebényi G, Toldy A. Characterization of high glass transition temperature sugar-based epoxy resin composites with jute and carbon fibre reinforcement. *Composites Science and Technology*. 2015;117:62-68.
15. Stark W. Investigation of the curing behaviour of carbon fibre epoxy prepreg by Dynamic Mechanical Analysis DMA. *Polymer Testing*. 2013;32(2):231-239.
16. Deng S, Hou M, Ye L. Temperature-dependent elastic moduli of epoxies measured by DMA and their correlations to mechanical testing data. *Polymer Testing*. 2007;26(6):803-813.
17. Ferry JD. *Viscoelastic Properties of Polymers*. 3rd ed. New York: Wiley; 1980.
18. Vajrasthira C, Amornsakchai T, Bualek-Limcharoen S. Fibre-matrix interaction in aramid-short-fibre reinforced thermoplastic composites. *Journal of Applied Polymer Science*. 2003;87(7):1059-1067.
19. Bikiaris D, Matzinos P, Prinos J, Flaris V, Larena A, Panayiotou C. Use of silanes and copolymers as adhesion promoters in glass fibre/polyethylene composites. *Journal of Applied Polymer Science*. 2001;80(14):2877-2888.
20. Costa CSMF, Fonseca AC, Serra AC, Coelho JFJ. Dynamic Mechanical Thermal Analysis of Polymer Composites Reinforced with Natural Fibers. *Polymer Reviews*. 2016;56(2):362-383.
21. Cândido VS, Oliveira MP, Monteiro SN. Dynamic-Mechanical Performance of Sponge Gourd Fiber Reinforced Polyester Composites. *Materials Science Forum*. 2016;869:203-208.

22. Oliveira GC, Neves ACC, Simonassi NT, Pereira AC, Margem FM, Barbosa AP, et al. Dynamic-Mechanical Characterization of Polyester Matrix Composites Reinforced With Eucalyptus Fibers. In: Ikhmayies SJ, Li B, Carpenter JS, Hwang JY, Monteiro SN, Li J, et al., eds. *Characterization of Minerals, Metals and Materials 2016*. Hoboken: John Wiley & Sons; 2016. p. 377-383.

23. Monteiro SN, Margem FM, Martins LBS, Loiola RL, Oliveira MP. Dynamic-Mechanical Analysis of Polyester Composites Reinforced with Giant Bamboo (Dendrocalmus giganteus) Fiber. *Materials Science Forum*. 2014;775-776:302-307.

24. Monteiro SN, Margem FM, Margem JI, Martins LBS, Oliveira CG, Oliveira MP. Dynamic-Mechanical Behavior of Malva Fiber Reinforced Polyester Matrix Composites. *Materials Science Forum*. 2014;775-776:278-283.

25. Margem FM, Monteiro SN, Bravo Neto J, Rodriguez RJS, Soares BG. The dynamic-mechanical behavior of epoxy matrix composites reinforced with ramie fibers. *Matéria (Rio J)*. 2010;15(2):164-171.

26. Pothan LA, Thomas S, Groeninckx G. The role of fibre/matrix interactions on the dynamic mechanical properties of chemically modified banana fibre/polyester composites. *Composites Part A: Applied Science and Manufacturing*. 2006;37(9):1260-1269.

27. de Paiva JMF, Frollini E. Unmodified and Modified Surface Sisal Fibers as Reinforcement of Phenolic and Lignophenolic Matrices Composites: Thermal Analyses of Fibers and Composites. *Macromolecular Materials and Engineering*. 2006;291(4):405-417.

28. Ray D, Sarkar BK, Das S, Rana AK. Dynamic mechanical and thermal analysis of vinylester-resin-matrix composites reinforced with untreated and alkali-treated jute fibres. *Composites Science and Technology*. 2002;62(7-8):911-917.

29. Mohanty S, Verma SK, Nayak SK. Dynamic mechanical and thermal properties of MAPE treated jute/HDPE composites. *Composites Science and Technology*. 2006;66(3-4):538-547.

30. Mittal V, Saini R, Sinha S. Natural fiber-mediated epoxy composites-a review. *Composites Part B: Engineering*. 2016;99:425-435.

31. Mishra S, Mohanty AK, Drzal LT, Misra M, Hinrichsen G. A Review on Pineapple Leaf Fibers, Sisal Fibers and Their Biocomposites. *Macromolecular Materials and Engineering*. 2004;289(11):955-974.

32. Harish S, Peter Michael D, Bensely A, Mohan Lal D, Rajadurai A. Mechanical property evaluation of natural fiber coir composite. *Materials Characterization*. 2009;60(1):44-49.