EasyChair Preprint

№ 8428

Effect of Agave Americana Fiber Content on Properties of Poly(3-Hydroxybutyrate-Co-3-Hydroxyhexanoate) Biocomposites

Mustapha Kaci, Celia Idres, Nadjet Dehouche, Carole Lainé and Stéphane Bruzaud

July 10, 2022

3rd International Conference on Applied Engineering and Natural Sciences

https://www.icaens.com/

July 20-23, 2022, Konya, Turkey

Effect of Agave Fiber Content on Properties of Poly(3-Hydroxybutyrate-Co-3-Hydroxyhexanoate) Biocomposites

Mustapha Kaci[1*], Celia Idres[1,2], Nadjet Dehouche[1], Carole Lainé[2] and Stéphane Bruzaud[2]

[1]*Laboratoire des Matériaux Polymères Avancés (LMPA), Université de Bejaia, 06000, Algeria*
[2] *Institut de Recherche Dupuy de Lôme (IRDL), UMR CNRS 6027, Université de Bretagne-Sud, Rue Saint Maudé, 56321, Lorient Cedex France*

[*]*mustapha.kaci@univ-bejaia.dz*

Abstract **–** Biodegradable biopolymers are considered as potential candidate materials to substitute the synthetic polymers derived from fossil oil. In this regard, poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx), which belongs to the polyhydroxyalcanoates family exhibits good thermo-processability, ductility, biodegradability and gas barrier properties. However, the high hydrophobicity, lack of advanced functionality and high cost of PHBHHx with further a slow crystallization, have restricted further expansion of its utilization. The use of cellulosic fibers is now becoming essential in biocomposites not only in terms of performance, but also to preserve the biodegradability character of the materials. In this paper, the effect of Agave fibers (AF) content, i.e., 10, 20 and 30 wt % on the thermal stability and viscoelastic properties of PHBHHx biocomposites was investigated. The study shows that the incorporation of AF to PHBHHx displays a slight decrease in thermal stability but nearly no change between the different compositions. Storage modulus and complex viscosity of biocomposite samples are significantly improved, however more pronounced at 30 wt% of AF.

Keywords – Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate), Agave fibers, Biocomposites, Thermogravimetric analysis, Viscoelastic properties.

I. INTRODUCTION

Development of biodegradable polymers as engineering materials is an essential issue for many researchers aiming to elaborate advanced polymers with comparably good performance and moderate cost [1]. One of the alternatives to overcome this economic and technical barrier is the elaboration of composites with natural fibers [2]. In this regard, the use of polyhydroxyalcanoates (PHAs) and natural fibres for production of bio-based composites has attracted interest of many industrial applications involving mainly packaging, automotive components, building, thermal and acoustic insulation and other high value applications in the context of circular economy approach [3]. Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx), which belongs to the PHAs family, exhibits good properties in terms of ease of processability, lower melting point, ductility, biodegradability and biocompatibility. However, PHBHHx is facing some drawbacks such as slow crystallization rate, poor thermal stability, and high production cost, which restrict its wide fields of applications [4]. Different natural fibers have been used to reinforce PHAs, but their incorporation also presents some issues. One of them is poor fibers dispersion in the polymer and weak interfacial adhesion causing low stress transfer between the matrix and the fibers. So, the characterization of the fiber-matrix interface is essential to determine the final properties of the biocomposite materials [5].

The paper focused on the preparation of Agave fibers (AF) reinforced PHBHHx biocomposites at

various loading rates, i.e., 10, 20, and 30 wt% by melt compounding. The effect of AF content on the thermal stability and viscoelastic properties of biocomposites based-PHBHHx was investigated by thermogravimetric analysis (TGA) and dynamic mechanical analysis (DMA), respectively aiming to determine the optimized filler composition.

II. Materials and Method

A. *Materials Used*

PHBHHx having 10.5 mol.% of hydroxyhexanoate comonomer (HHx), was provided by Kaneka Co. (Japan) under the grade Aonilex 151X. The main properties of the resin, as provided by the manufacturer, are the following: melting temperature (T_m) = 126°C, density = 1.19 $g.cm^{-3}$ and glass transition (T_g) = 0°C.

Agave fibers were extracted from collected leaves in the region of Bejaia (Algeria). The fiber extraction process was carried out according to the procedure described in literature [6]. Before any use, the fibers were chopped into an average length of 3 mm.

B. *Preparation of PHBHHx-AAF Biocomposite Samples*

To prevent any clogging of extruder feeding by fibers aggregating, thin films of both neat PHBHHx and PHBHHx-AF biocomposites were first prepared by solvent-casting method in chloroform at various loading rates, i.e., 10, 20 and 30 wt. % according to Table 1. After solvent evaporation, the samples were cut into small pieces and placed in an oven overnight at 70°C. Then, the samples were melt compounded in a twin-screw extruder (Xplore MC 15), operating at 145°C and 60 rpm. The extruded materials were grinded and compressed in a Carver® hot press at 150°C according to the following procedure: the granules were preheated without pressure for 180 sec., followed by compression for 120 sec. under 10 metric tons to obtain samples for testing.

Table 1. Code and composition of PHBHHx-AF biocomposites.

Samples	Composition (wt%)	
	PHBHHx	AF
PHBHHx	100	0
PHBHHx-AF10	90	10
PHBHHx-AF20	80	20
PHBHHx-AF30	70	30

C. *Technical Characterization*

Viscoelastic properties of both neat PHBHHx and its biocomposites were investigated by using a dynamical mechanical analyzer DMA 2980 from TA Instruments.

The operating conditions were set as follows: temperature range: -40 to +80°C at 3°C min^{-1}. The dynamic tensile test was performed at a frequency of 1 Hz with amplitude of 10 mm.

TGA experiments were performed by a thermal analyzer (Mettler Toledo ATG/DSC1 STAR^e). Samples of about 10 mg were heated from 20 to 600°C at 10°Cmin^{-1} under a nitrogen flow.

The temperature values corresponding to mass loss at 10% (T_{10}) and the temperature at maximum degradation rate (T_{mdr}) were determined based up on three replicates.

III. RESULTS

Thermal stability of both neat PHBHHx and PHBHHx-based biocomposites was evaluated through degradation temperature values with respect to filler content ratio.

TGA/DTG data are provided in Table 2. Accordingly, it is observed a decrease in thermal stability of PHBHHx-based biocomposites with increasing AF content ratio. Indeed, the addition of AF to PHBHHx leads to a decrease of both T_{10} and T_{mdr} by approximately 7 and 10°C, respectively at filler content of 10 wt.%, which further remains almost unchanged at 20 and 30 wt.%.

Table 2. Degradation temperature values at T_{10} and T_{mdr} for neat PHBHHx and PHBHHx-based biocomposites.

Samples	Degradation temperature (°C)	
	T_{10}	T_{mdr}
PHBHHx	275.7±0.6	291.7±0.6
PHBHHx-AF10	268.0±1.7	283.0±1.0
PHBHHx-AF20	268.7±0.6	281.7±0.6
PHBHHx-AF30	266.7±0.6	280.0±0.0

Figure 1 depicts the evolution curves of loss modulus of neat PHBHHx and PHBHHx-based biocomposites as a function of temperature.

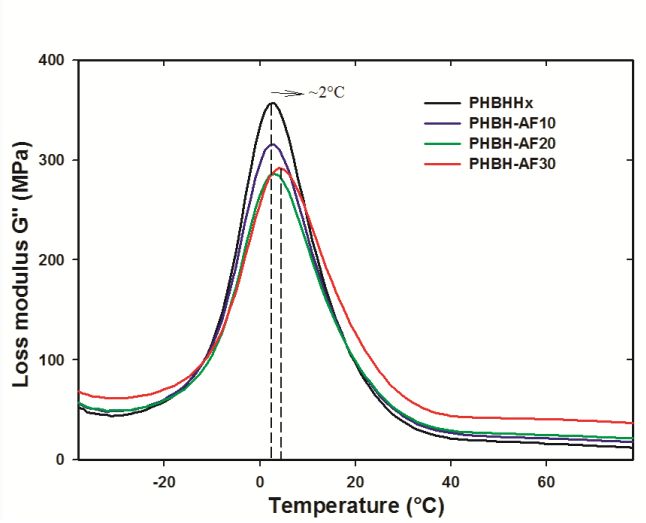

Fig. 1 Loss modulus curves of neat PHBHHx and PHBHHx-based biocomposites vs. filler content.

IV. DISCUSSION

The decrease in thermal stability of the biocomposite samples is attributed to the holocellulose fraction contained in the natural fibers, which is less thermally stable than the neat polymer [7]. Moreover, the presence of fiber aggregates in the matrix may also be responsible for the decrease in thermal stability by causing defects in the polymer [8]. On the other hand, the decrease of T_{mdr} is more pronounced in the presence of AF natural fibers whatever the filler content ratio due probably to the thermal degradation of cellulose, which starts at 250°C.

From DMA data, it is observed a slight shift of the loss modulus (G") (~2°C) toward higher temperatures at 30 wt.% compared with the neat polymer and the other biocomposite samples. This is probably due to the restriction of polymer chains mobility indicating the occurrence of fiber-fiber and to some extent fiber-matrix interactions..

V. CONCLUSION

From this study, the following conclusions can be drawn: the reinforcing effect of AF on PHBHHx biocomposites evaluated by DMA is more pronounced at 30 wt.% compared with other biocomposite samples.

On the other hand, the thermal stability displayed a little decrease but nearly no change between the different formulations.

ACKNOWLEDGMENT

The authors would like to thank Campus France through PHC-Tassili programme N°19 MDU 204 for their financial support.

REFERENCES

[1] G. E. Luckachan and C. K. S. Pillai, "Biodegradable polymers- A review on recent trends and emerging perspectives," *J. Polym. Env.*, vol. 19, pp. 637–676, 2011.

[2] N. Gómez-Gast, M. D. R. López Cuellar, B. Vergara-Porras and H. Vieyra,"Biopackaging potential alternatives: Bioplastic composites of polyhydroxyalkanoates and vegetal fibers," *Polymers*, vol.14, p.1114(1-24), 2022.

[3] P. Cinelli, N. Mallegni, V. Gigante, A. Montanari, M. Seggiani, M. B. Coltelli, S. Bronco, A. Lazzeri, "Biocomposites based on polyhydroxyalkanoates and natural fibres from renewable byproducts," *Appl. Food Biotechnol.*, vol. 6, pp. 35-43, 2019.

[4] N. Dehouche, C. Idres, M. Kaci , I. Zembouai, S. Bruzaud, "Effects of various surface treatments on Aloe Vera fibers used as reinforcement in poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) biocomposites," *Polym. Degrad. Stab.*, vol. 175, p. 109131, 2020.

[5] N. Kaouche, M. Mebrek, A. Mokaddem, B. Doumi, M. Belkheir and A. Boutaous,"Theoretical study of the effect of the plant and synthetic fibers on the fiber-matrix interface damage of biocomposite materials based on PHAs (polyhydroxyalkanoates) biodegradable matrix," *Polym. Bull.*, 2021. DOI: https://doi.org/10.1007/s00289-021-03849-w.

[6] F. Mannai, H. Elhleli, and M. Ammar, "Green process for fibrous networks extraction from Opuntia (Cactaceae): morphological design, thermal and mechanical studies", *Ind. Crops Prod.*, vol. 126, pp. 347-356, 2018.

[7] A. A. Yussuf, I. Massoumi, and A. Hassan, "Comparison of polylactic acid / kenaf and polylactic acid/rise husk composites: the influence of the natural fibers on the mechanical, thermal and biodegradability properties." *J. Polym. Env.*, vol. 18, pp. 422–429, 2010.

[8] L. Hassaini, M. Kaci, N. Touati, et al. "Valorization of olive husk flour as a filler for biocomposites based on poly (3-hydroxybutyrate-co-3-hydroxyvalerate): Effects of silane treatment. *Polym. Test*., vol. 59, pp. 430–440, 2017.