



Mechanical properties and biodegradation of biocomposites based on poly (hydroxybutyrate-co-valerate) and alfa fibers

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Abstract

In recent years, most of the scientific research had focused on developing environment-friendly biodegradable materials after being used. There are two ways to do that: the first consists of the use of biodegradable plastics, and the second is the incorporation of biodegradable fibers in polymers. The main objective of this paper is to develop eco-friendly biodegradable biocomposites and to make their characterization. We devoted significant attention to investigate the use of alfa fiber as reinforcement in Poly (hydroxybutyrate-Co-valerate) (PHBV) material. The effects of fiber content and fiber treatment, as well as the biodegradation behavior of elaborated composites, have been investigated. From the main results, we observed some morphological changes that occurred on the fiber surface after chemical treatment. The use of fibers in PHBV increases the modulus but not necessarily improves the tensile strength of resulting composites but remains dependent on the interfacial adhesion. Alkali treatment of alfa fibers leads to improve the tensile strength and the rigidity of PHBV/alfa composites. SEM micrographs highlighted differences in microstructure of untreated and treated composites indicating that fiber-treatment also enhances the quality of the interface between the thermoplastic matrix and alfa fibers. In terms of degradability characteristics, the treated composites were highly biodegradable compared to both neat PHBV and untreated composites when subjected to water absorption in distilled and seawater. Also, elaborated composites undergo significant degradation into seawater, indicating their environmentally friendly character.

Keywords Alfa fibers · PHBV biopolymer · Biocomposites · Mechanical properties · Biodegradation

Introduction

Polymer/natural fibers composites have become some essential materials for modern life and considered as alternative material living [1]. Polymer composites have practical processing techniques, appealing mechanical properties and are light [2]. In another way, the polymer also can evil the environment, particularly in the marine medium. Polymer

materials can fragment into smaller bits, but complete natural biodegradation can be awfully long in the time, consequently they accumulate in the environment [3]. 80% of the total litter in the oceans is the origin of Land-based sources [4]. According to Gewert et al., [5] plastic debris in the world's oceans is of particular environmental concern, more than 60% of all floating debris in the oceans is plastic, and amounts are increasing each year and 80% of the total litter in the oceans is the origin of Land-based sources.

Indeed, the polymer fragments pose huge worry to wildlife. By that, the use of degradable composites, based on a biodegradable matrix and natural fibers, is required as a solution to this problem and consolidates to the ecological awareness as their disposal could be easy after the end of life [6, 7]. For that purpose, Polyhydroxyalkanoates (PHA) are biodegradable polymers obtained from a whole variety of microorganisms, therefore, they are environmentally friendly materials [8–10].

Poly (hydroxybutyrate-co-Hydroxyvalerate) (PHBV) is biodegradable and biocompatible polyester of the family

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of polyalkenoates which is produced by bacterial microorganisms formed from 3-hydroxybutyrate units intercalated with a variable percentage of random segments of 3-hydroxyvalerate [11]. However, because of the higher production cost of PHBV as compared to traditional polymers, there are many attempts to replace a quantity of PHBV by low-cost fillers without affecting the properties of the resulting composites. Otherwise, to enhance the mechanical properties of PHBV and reduce its high cost, it is often blended with natural fibers with low market prices and synergy effects [12]. According to Ahankari et al. [13], the adding of 30 wt.% wheat straw fibers raises the tensile and storage modulus of PHBV by 256% and 308%, respectively.

Moreover, the effect of fiber content remains dependent on the quality of the fiber-matrix interface, which in turn, depends on the use of the coupling agent and the compatibility between the fiber and the matrix. For instance, good compatibility (better interfacial bonding) results in an improvement in the mechanical properties of composites and vice versa. Indeed, the hydrophilic character of natural fibers (e.g. alfa fibers) can lead to a poor fiber/matrix adhesion with hydrophobic matrices [14]. That is why chemical modifications have been considered for natural fibers to attenuate this problem of incompatibility. In this context, many works have been carried out to study the effect of incorporation of fibers-treatment on the mechanical properties of fiber-reinforced composites [15–21]. In general, chemical treatment of fibers was found to improve the mechanical properties (flexure, tensile, impact) of resulting composites compared to untreated composites. Alkali treatment of fibers improved strain at break and impact strength of PHBV composites by 35% compared to their untreated counterparts [13]. DMA studies indicated better interfacial interaction of PHBV with the fibers. Thus, the biodegradable composite can be taken as a judicious solution for the environment. Degradation starts by scission of the main chains of macromolecules, under thermal effect, oxidation, photolysis, radiolysis, or hydrolysis [20]. Hydrolytic degradation occurs after water absorption which leads to breaking the polymer backbone by random rupture of covalent bonds with the formation of oligomers and lastly monomers [21]. It is interesting to know about the effect of fibers on the degradation process of biopolymers in an aqueous medium.

Berthet et al. [21] evaluated the effect of the fiber exposed to wide moisture conditions (i.e. 0% and 98% RH) on the structure of composites by measuring the PHBV chain length using Gel permeation chromatography (GPC) technique. It was noticed that the molecular weight of PHBV decreases with increasing fiber moisture content, and this was assigned to the hydrolytic degradation of PHBV by chain scission reactions at ester linkages. Over this process, water molecule cleaves ester bond, leading to the formation of a carboxyl

and hydroxyl end group. It is considered that the hydrophilic nature of fiber fosters the occurs of hydrolyzing.

The main objective of this work is to investigate the effect of alfa fibers content and fiber surface treatment on the mechanical properties of PHBV based composites and their degradation process in distilled water and seawater solutions. The mechanical performance, in particular, the tensile strength and modulus of the composites were evaluated when studying different fiber contents, with and without fiber treatment. Finally, the biodegradation behavior of neat PHBV and its untreated and treated composites was studied in the aqueous environment.

Experimental details

Materials

PHBV copolymer used in this study was supplied by Tianah Biological materials Co. LTD. (China) with a molar ratio of 92:8 (HB: HV). It was commercialized in the form of pellets under the grade name ENMAT Y1000P. The properties of PHBV as given by the manufacturer were: density: 1.25 gcm⁻³, Tg: 8 °C, Tm: 165 °C and overage molecular weight Mw: 400 Kg mol⁻¹.

The alfa fiber was collected at M'Sila in Algeria and its average particle size was around 125 µm. The whole characteristics of alfa are listed in Table 1.

Chemical treatment of alfa fiber

The treatment was made by an in-house optimized method [22]. Firstly, the Olive husk flour (OHF) was pre-treated by immersing in a solution consisting of 38 ml of acetone and 62 ml of ethanol which was left stirring for 24 h at room temperature. After filtration, the OHF was washed with distilled water and dried under the hood for 24 h. Then, alfa fiber is treated with 2% NaOH-water for one hour and then washed many times with distilled water containing 1% acetic acid to neutralize sodium hydroxide. Finally, OHF is washed with distilled water until neutral pH and dried in an oven at 80 °C for 6 hours.

Table 1 Characteristics of alfa fiber

Chemical Composition (wt%)			Mechanical Properties		
Cellulose	Hemicellulose	Lignin	Young's Modulus (GPa)	Elongation at Break (%)	Tensile Strength (MPa)
45.00	20.94	29.80	21 ± 1.1	1.80 ± 0.9	145 ± 19

Preparation of composites

Composite samples are based on PHBV copolymer reinforced with various alfa contents of (10, 20, and 30 wt.% alfa fibers). To elaborate composite samples, alfa fibers and PHBV granules were dried separately in an oven at 80 °C of 24 h, before extrusion. After that, blends of fibers and matrix were performed using a co-rotating twin-screw extruder. The compounds were realized at a uniform temperature (170 °C) and a constant screw rotation speed (100 tr/min).

Finally, samples were prepared by injection molding using an Engel ES 8035 machine. The melt and the mold temperatures were kept at 160 and 50 °C, respectively.

Characterization techniques

FTIR spectra of alfa fiber before and after treatment were saved using an FTIR SHIMADZU FTIR-8400S in the range of 4000–400 cm^{-1} with a resolution of 4 cm^{-1} . The analysis was carried out using a KBr pellet method.

Mechanical properties of composites are obtained using an MTS Synergie RT1000 testing machine with a crosshead rate maintained at 5 mm min^{-1} . The dimensions of the calibrated part have a width of 4 mm and a length of 45 mm.

The morphology of tested samples was examined using a Hitachi S-2700 electron microscope. For composites, the compression-molded sheet was cryogenically fractured in liquid nitrogen. However, alfa morphology was assessed with the pellet method. The micrographs of composites were taken at a magnification of $\times 100$ while those of alfa fiber was at $\times 100$ and $\times 10$.

The biodegradation of neat PHBV and PHBV based composites, loaded with untreated and treated alfa fiber, was assessed by submitting samples in the aqueous environment according to ASTM F1635-04a specifications [23] and using tensile samples (ASTM D-638–02, 2002) [24]. The biodegradation process is carried out at 37 °C separately in distilled water (pH=7) and seawater (pH=8) over 15 days. Different samples are taken regularly after aging to be characterized. The mass evolution of the aged sample is assessed by the weight difference between the samples before and after immersion in water using the Eq. (1):

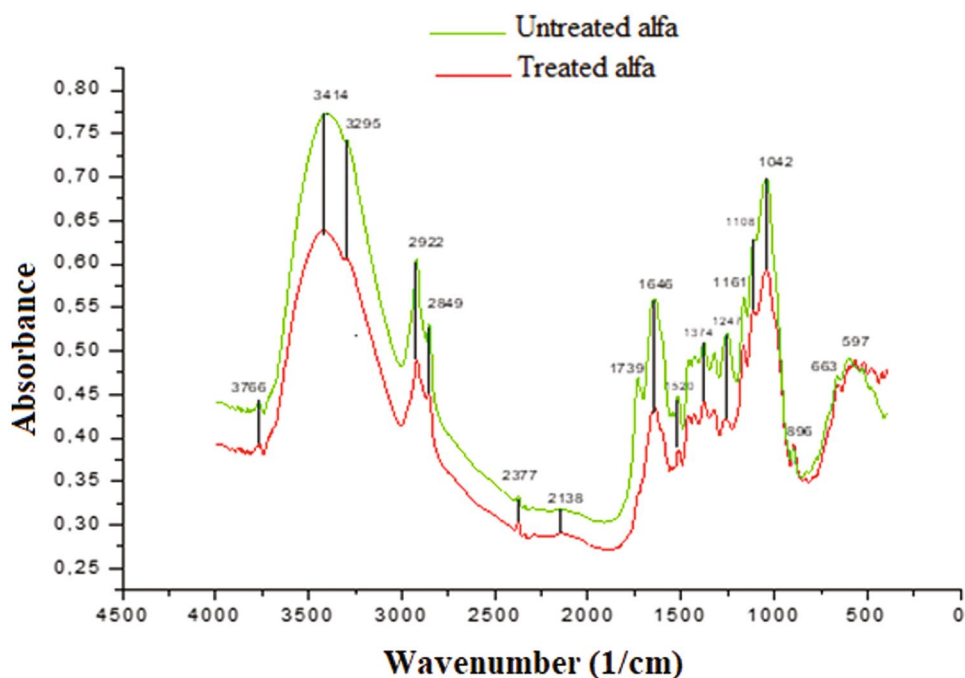
$$\% \text{ of weight} = \left[\frac{\text{final mass} - \text{initial mass}}{\text{initial mass}} \right] * 100 \quad (1)$$

Results and discussion

FTIR analysis of untreated and treated alfa fiber

Each absorption band that appears on the spectra (Fig. 1) corresponds to a chemical function or group. So, the band at 3414 cm^{-1} can be attributed to the vibration of hydroxyl groups (-OH) of cellulose and hemicellulose. The mid-intensity peaks at 2922 cm^{-1} and 2849 cm^{-1} reflect the symmetric and asymmetric elongation vibrations of the C-H bonds of the -CH₂ group of the cellulose and lignin segments. The absorption band at 1161 cm^{-1} corresponds to the C–O–C bond of the cellulose. The band at 1739 cm^{-1} is ascribed to the vibrations of carbon monoxide (CO) in hemicellulose which disappeared in the IR spectrum of treated alfa [25].

Fig.1 FTIR spectrum for untreated and treated alfa



According to previous studies [26, 27], the absence of this band affirms the removal of pectin from the surface of the fiber. The spectra also reveal an absorption band at 1042 cm^{-1} , associated with the elongation vibrations of the $\text{C}=\text{O}$ carbonyl groups of the acetyl groups of hemicellulose components found in alfa fiber. The absorption band at 1161 cm^{-1} can be assigned to the $\text{C}-\text{O}-\text{C}$ bond of the cellulose. It was noted the appearance of an absorption band at 1520 cm^{-1} which reflects the lignin $\text{C}=\text{C}$ deformation vibration, the symmetrical elongation of the $\text{C}=\text{C}$ bonds of the lignin having aromatic groups in its structure.

The 1247 cm^{-1} band corresponds to the vibration of the CO bond of acetyl groups of lignin and hemicellulose [28]. Besides the peaks of the cellulose, all the others decreased in intensity after the treatment thus justifying the elimination of hemicellulose and lignin.

Morphology of untreated and treated alfa fiber

Fig. 2 shows SEM micrographs of untreated and treated alfa fibers. As can be seen, the untreated fiber shows some irregularities of cell wall present on the fiber surface. The untreated fiber exhibits a roughened aspect on their surface which can prevent the anchoring of the polymer matrix on the surface of the fiber.

However, when looking at the SEM micrograph of the longitudinal surface of the treated fiber, we note that the defects which were present on the surface of the untreated fibers are removed, creating a clean and silk surface. This fact is due to the elimination and leaching out of waxy substances and pectin after treatment performed on the surface of alfa fiber. This observation agrees well with that reported by Rokbiet al. [19] when studying the effect of alkali treatment on the fiber morphology in the case of a natural

fiber-reinforced polyester composite. Additionally, similar morphological changes that occurred after the treatment of the fibers have been reported by Aziz et al. [16].

In the next section, we will study whether the morphological changes induced by the chemical treatment on the alfa fibers influence the mechanical behavior or whether the mechanical properties of the resulting composites remain unaffected by the fiber treatment.

Mechanical properties of PHBV and PHBV/alfa fiber without and with treatment

Fig. 3 reports the tensile properties of PHBV, and composites reinforced with 10, 20, and 30wt% alfa fiber. As it can be seen from Fig.3-a, the tensile strength for all PHBV/alfa fibers composites, with and without treatment, were found to be lower than that of neat PHBV.

This result could be due to the weak interfacial adhesion and low compatibility between alfa fiber and PHBV matrix. The presence of too many polar groups on the surface fibers and less of functional groups in the chemical structure of the PHBV matrix leads to a deficiency of interfacial interaction between fibers and matrix [29]. The loss of mechanical properties upon the incorporation of natural fibers was attributed to the lack of adhesion between polar fibers (hydrophilic character) and hydrophobic plastic matrix [30]. Finally, one can conclude that the application of natural fibers as reinforcements in composite materials requires an excellent fiber-matrix adhesion [16] because the macroscopic mechanical properties (for example tensile strength) strongly depend on the fiber-matrix interface [18].

Given the fiber-treatment effect, Fig.3-a shows equally higher tensile strength performance for treated composites as compared to untreated ones. This confirms that the applied fiber treatment can be a successful method and amply used to improve the interfacial adhesion between polymer and fibers. This result is again consistent with the more intensive chemical anchoring of the polymer matrix on the surface of the fibers, thereby allowing a better transfer of stresses from the matrix to the fiber.

On the other hand, when looking at the mechanical response in terms of rigidity (at little strains), Fig.3-b indicates that the addition of alfa fibers leads to improve Young's modulus of the PHBV matrix. The resulting composites became more rigid with the incorporation of alfa fibers, whatever the fiber content used. According to Singh et al. [30], the increased modulus of the composites is attributed to the uniform dispersion of fibers in the PHBV matrix. Fibers addition reduces the mobility of the polymer chains and became more stiffeners. It also weakens the ability of the composite material to deform by restricting the mobility of the polymer chains. As a consequence, it

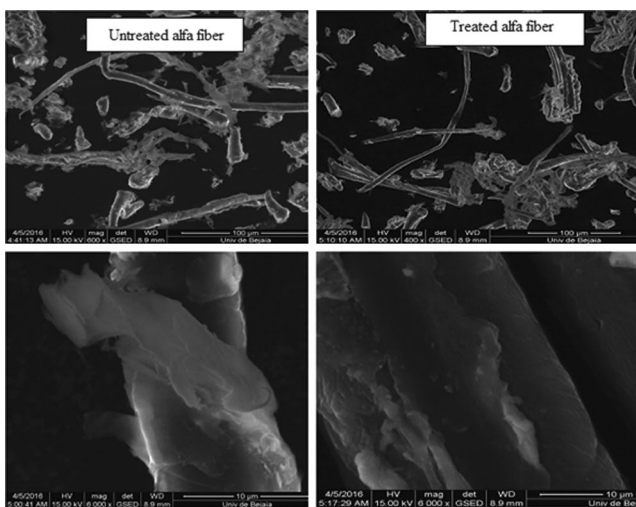
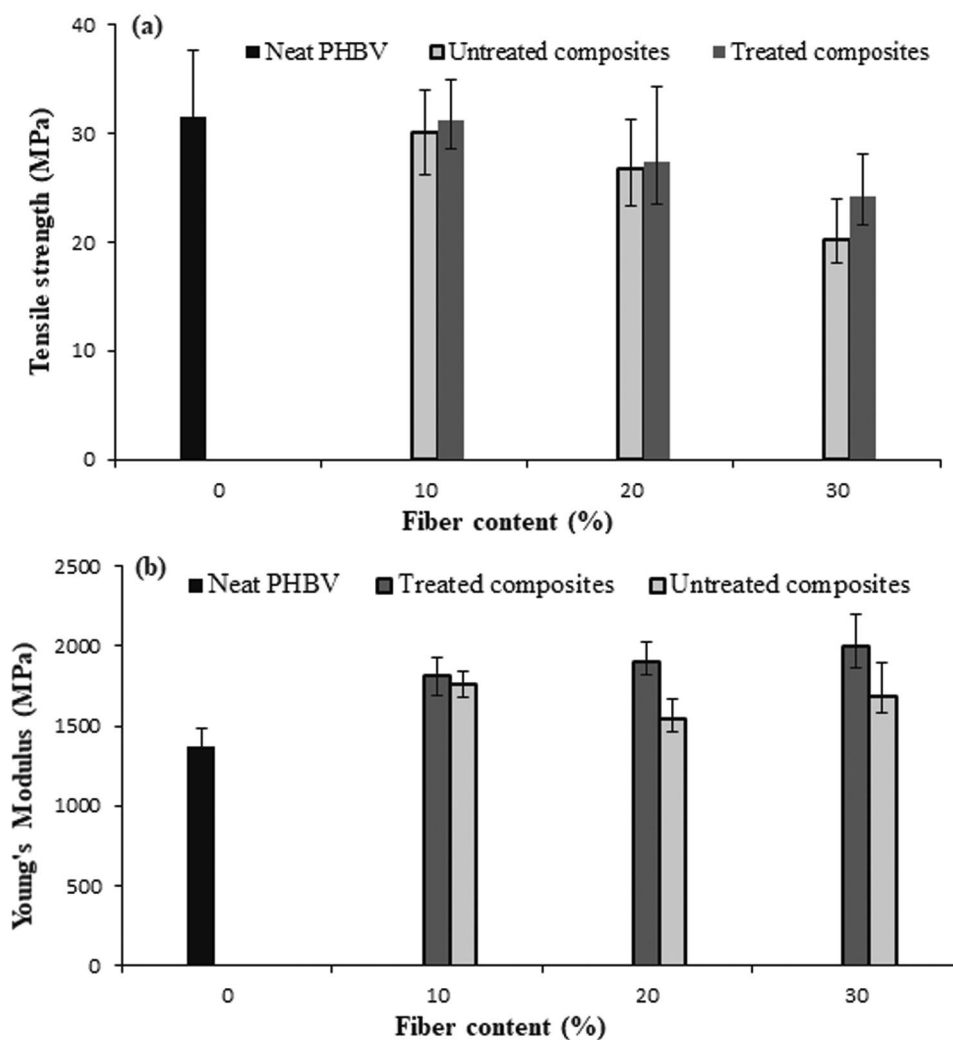


Fig.2 SEM micrographs of untreated and treated alfa fiber

Fig.3 Tensile properties of PHBV and composites with untreated and treated alfa fiber



is difficult for the segments of the material to easily slip past each other. This could be due to the better reinforcing action of the fiber [30].

In terms of chemical treatment impact, there was an increase in Young's modulus for composites based on treated fibers. So, the rigidity of the composites also increased with fiber surface-treatment. Also, the improvement of material rigidity remains dependent on the fiber content: it is around 7%, 27% and 26% for 10%, 20% and 30% of alfa fibers, respectively.

Similarly, the tensile strength was improved by 4–16% (depending on alfa fibers content) compared with bio-composites based on untreated fibers. This suggests that fiber surface-treatment improves fibers' dispersion in the PHBV matrix. The same trend was observed in Ref [15] in the case of composites based on LDPE/cellulosic fibers. Furthermore, fibers-treatment allows improvement of the mechanical interlocking and chemical bonding between the fiber and the matrix resulting in superior mechanical properties [16].

SEM observations of untreated and treated composites

The influence of the fiber treatment on the interface between the thermoplastic matrix and alfa fibers was investigated by examining the fracture surfaces of the composite samples after tensile tests (Fig. 4).

As can be observed from Fig. 4, SEM micrographs highlighted the differences in the microstructure of the composite samples. Indeed, SEM images of composites without treatment (on the right) show that the fiber was completely pull-out from the PHBV matrix leaving voids and cavities. This is due to the low interfacial adhesion, which indicates a poor affinity between PHBV and alfa fiber. The presence of greater voids, cavities, and holes was higher for composites loaded with 30% of untreated alfa.

Besides, on the surface of PHBV composites containing treated alfa fibers, a relatively homogeneous and regular surface is observed. The voids between treated fiber and matrix decreased, which can be explained by a good property of the

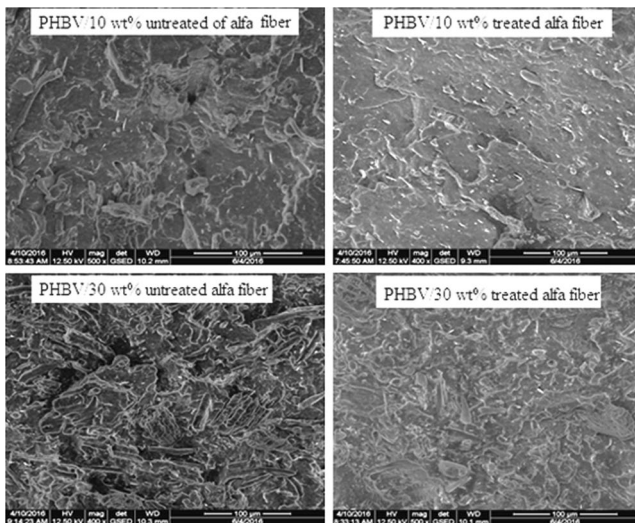


Fig.4 SEM photomicrographs of composites based on untreated and treated composites

interface, a better interfacial adhesion. These results are in good agreement with those obtained by Demir et al. [17] in the case of polypropylene-luffa fiber composites. The latter have attributed the decrease in voids between fiber and polymer matrix to the enhanced adhesion between fiber and matrix by the treatment.

Biodegradation of PHBV and PHBV/alfa composites

Fig.5 illustrates the mass evolution over the immersion time in distilled water for the neat PHBV, untreated, and treated composites. Here, it is worth noting that *i*) there was a very distinguished behavior between the neat PHBV and PHBV/

alfa composites, and *ii*) there was a great difference between responses of composites manufactured without and with fiber treatment.

From Fig. 5 (on the right), there are two different trends: slight mass loss for the neat PHBV and mass gain for untreated composites. The diffusion of distilled water seems too fast for untreated composites. One of the possible reasons can be the hydrophilic temperament of alfa fiber which causes an important water absorption in the composites. Thus, PHBV/alfa composites with a high fiber content have the highest water absorption rate. The macromolecular structure and the hydroxyl groups of alfa fiber can also accommodate water molecules linked by hydrogen bonds. The insertion of the water molecules within the hydrophilic macromolecules causes the swelling of the material hence the mass gain [31].

The water absorption phenomenon relies on the composition of the fiber, fiber orientation, the area of the exposed surface, the permeability of fibers, and hydrophilicity of each constituent [32]. The chemical surface modification of the fiber supports the decrease of the water absorption rate of the composites. This behavior can be assigned to the reduction of the -OH groups after the treatment as it is confirmed by the FTIR analysis of the fiber. In this case, the decrease in mass due to the hydrolysis of the matrix is noted (Fig.6). The fibers acted as paths for the water diffusion in the biocomposite. The composite containing higher alfa content degraded more than all other materials.

In contrast, seawater involves a weight loss for both neat PHBV and resulting composites with and without fiber treatment (Fig. 7). One can say that the degradation rate is affected by the environment. The intense degradation of materials occurred in the saline environment can be

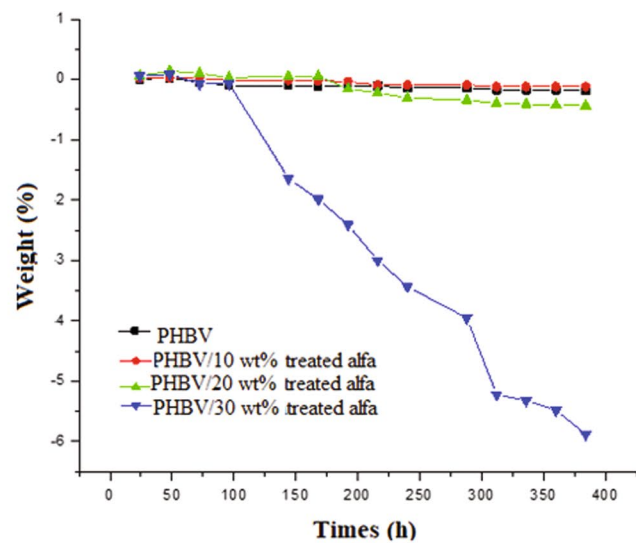
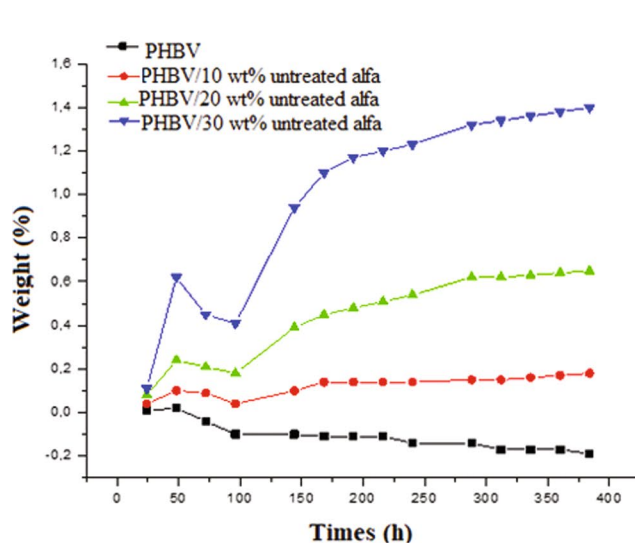


Fig.5 Evolution of weight samples in distilled water

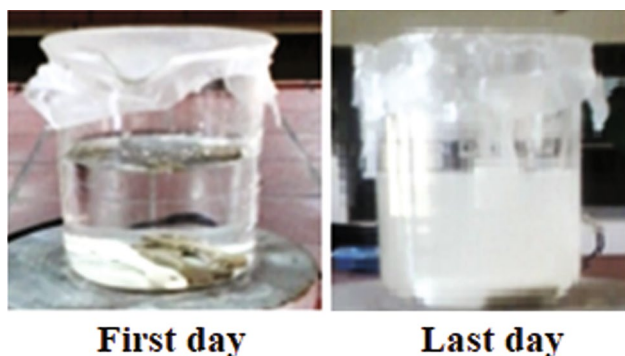


Fig.6 State of the water before and after immersion

assigned to the higher activities of microbial populations and metabolic [33]. As specified by Harmaen et al. [34], the biodegradation rate of polymers relies on various factors in the way that microbial activity, surface area, moisture, temperature, pH, and nutrients.

The stages of degradation under alkaline environment leading to hydrolysis, i.e., production of alcohol and carboxylic acid are schematized in Fig. 8.

Hydrolysis results in *i)* chain breaks, *ii)* Fastening of the hydroxide ion to the carboxyl group of the ester with the formation of a tetrahedral intermediate, *iii)* Fragmentation of the tetrahedral intermediate with the elimination of an ethanolate ion, and *iv)* Deprotonation of the carboxylic acid by the ethanolate ion with the formation of a carboxylate ion.

According to Rydz et al. [35], esters formed from acidic alcohols hydrolyze faster than ester formed from aliphatic alcohols. While Muhamad et al. [36] clarify that is due to the presence of greatly branched ester groups and the largest number of tertiary carbons which support hydrolysis and cleavage of the materials.

Also, to provide more insights on the material degradation, SEM observations were carried out on sample surfaces before and after degradation. Fig. 9 displays the surface of the composites loaded with 30 wt.% untreated and treated alfa fibers before and after immersion in seawater.

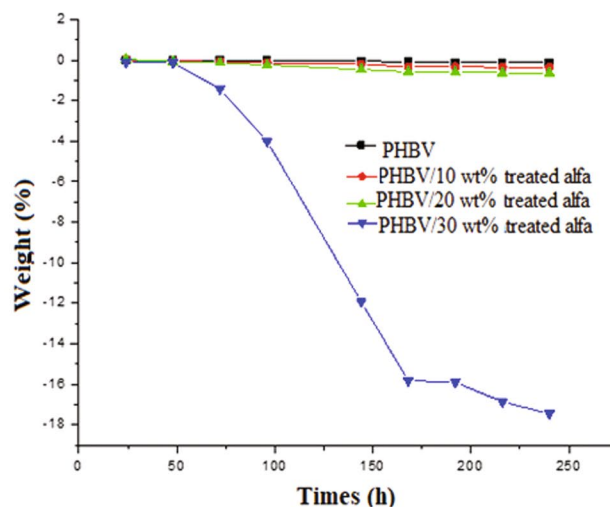
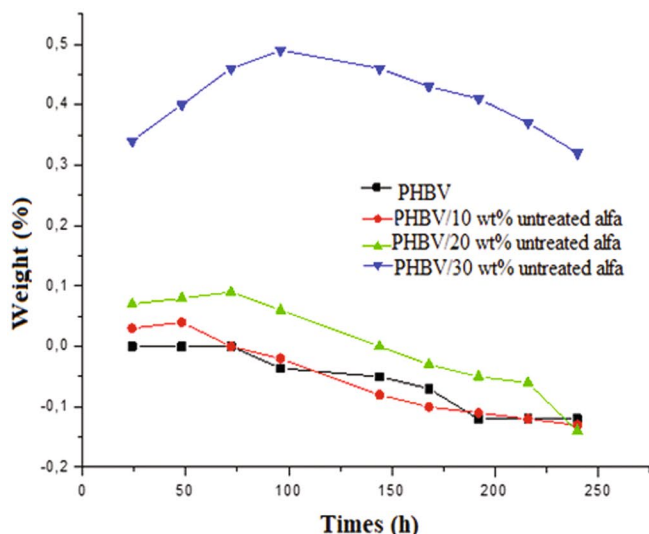


Fig.7 Evolution of weight samples in seawater

Fig.8 Hydrolysis steps of polyesters under alkaline conditions

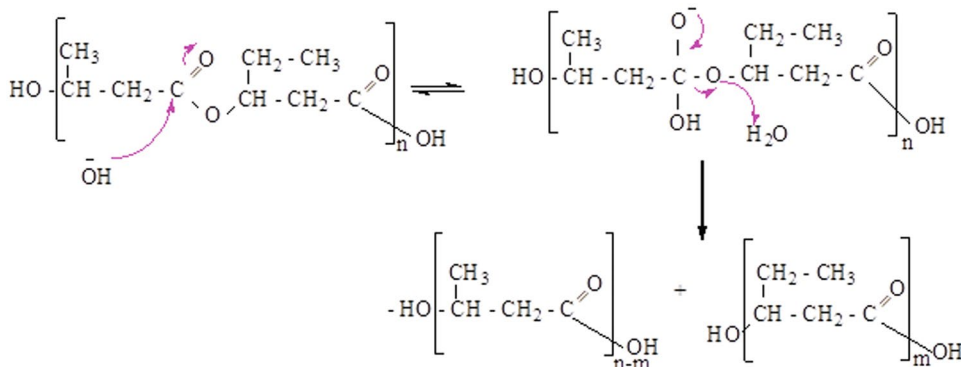
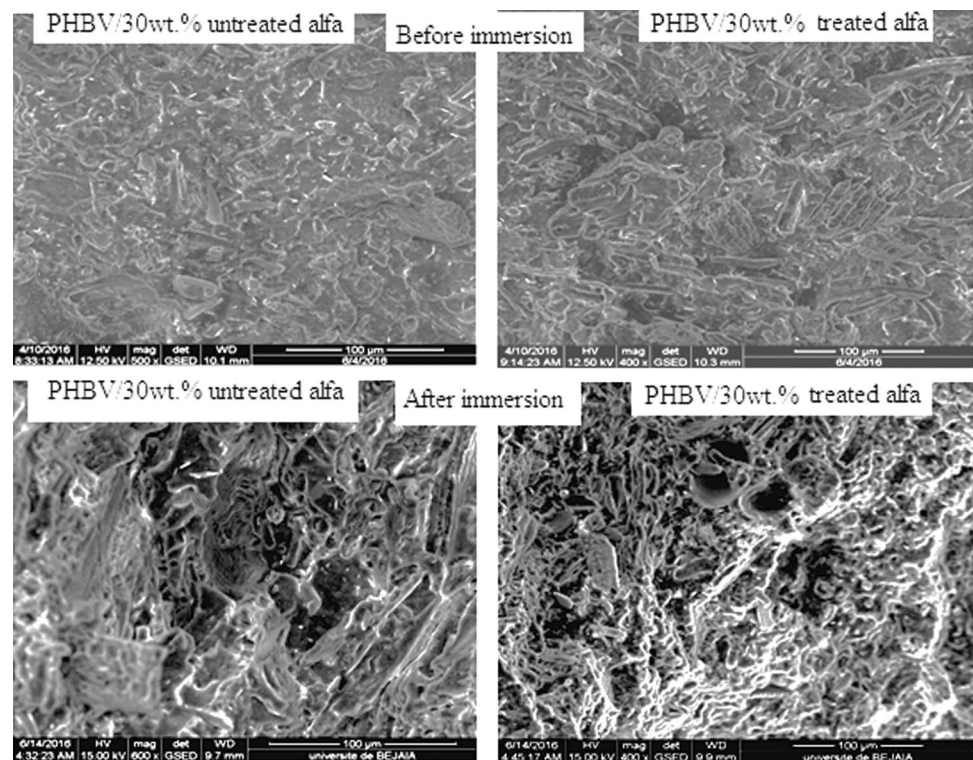


Fig.9 Morphology of PHBV/alfa composites before and after immersion in seawater



The saline medium induces the formation of more and larger gaps on the surface of the composites. These holes become a refuge for the larger amount of water which can speed up the hydrolysis process.

The SEM study of Altaee et al. [37] confirmed various changes that had taken place within the surface of the polymer after degradation including alterations in the appearance of pores, cavity, grooves, incisions, slots, and pointers. Such changes were due to the biopolymer films degradation.

These results join the approaches proposed by Bond et al. [38] about what may help mitigate the problems of plastic litter. This strategy to reduce plastic pollution, indeed, is to replace established polymers with those which degrades relatively quickly in the environment and which are often described as green and biodegradable.

Conclusions

In this paper, we assessed the effects of the alfa fiber content and the fiber surface treatment on the mechanical properties as well as the degradation of composites based on PHBV in an aqueous medium. The idea behind is to develop eco-friendly composites by exploiting the use of natural fibers and PHBV biopolymer. Based on the obtained results, the main findings are summarized below:

Morphological changes occurred on the fiber surface after chemical surface treatment.

Improvement of mechanical performance of composites reinforced with lignocellulosic fibers needs strong adhesion.

Alkali treatment of alfa surface fibers enhances both of the tensile strength (by 4–16% according the percentage of alfa fibers) and the rigidity (by 7–25%) of PHBV/alfa composites.

SEM micrographs highlighted differences in microstructure of untreated and treated composites confirming the enhancement of the quality of the fiber-matrix interface owing fiber surface-treatment.

The treated composites were highly biodegradable compared to both neat PHBV and untreated composites.

All elaborated composites undergo degradation more pronounced into seawater (compared to distilled water) that is wealthy in microbial populations indicating their environmentally friendly character.

Acknowledgment I would like to dedicate this paper to my father whose sudden and untimely death provoked his leave. Thanks for always being there for me. You not gone; you remain here beside me. Just in a different form, in my heart forever.

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