Aging by burial in soil of polylactic acid biocomposites reinforced with Alfa fiber treated with dispersing agent

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Abstract

Soil burial test comprises of placing samples in soil/ compost for long durations and testing the mechanical properties/dimensional changes/ morphology before and after soil burial. In a bioreactor, samples are placed in a composting vessel containing a mixture of compost and the percentage of biodegradation is theoretically calculated by measuring the amount of CO2 evolved from the composting vessel for a period of 45 days. This present work is focused on the study of the durability of different PLA/Alfa biocomposite materials treated with BYK W-980 and untreated, prepared via burial in soil aging. The study of biodegradation by burial in soil showed that the biodegradability of biocomposites prepared with untreated and treated PLA/Alfa degrade in soil more than those prepared with BYK W-980 for the same burial conditions. Knowing that the PLA buried in the ground is biodegradable after 4 to 5 years, by consecrating in this time interval of burial, we only have the fibers which degrade.

Keywords: *Aerobic biodegradation, Alfa fiber, Aging, Burial in soil, Dispersing Agent, Polylactic Acid.*

I. Introduction

Environmental concerns and awareness have paved the way to the development of biodegradable composites as a replacement for petroleum-derived or non-degradable polymers. So, there is an increase in demand for natural fiber-based composites for commercial use in various industrial Sectors [1].

A variety of biopolymers such as polylactic acid (PLA), polyhydroxyalkanoate (PHAs), and polybutylene succinate (PBS) are reported to be used as matrixes in composites. These biopolymers are naturally sourced and can potentially be combined with various natural fibres/lignocellulosic to produce biodegradable composites [2].

Natural fibers are sustainable materials in nature with advantages like low cost, lightweight, renewability, and, most importantly, biodegradability [3,4]. Pretreatment methods can improve the interfacial bonding quality. Physical treatment methods include hydrothermal treatment [5], microwave processing [6-8], steam explosion method [9], etc. Chemical treatment methods include acid treatment [10], alkaline treatment [11], acetylation treatment [12], benzoylation treatment [13], etc. Recently, many investigators have studied different pretreatment methods; the reports indicate that treated fibers can improve the physical and mechanical properties of fiber-plastic composites [14]. The rough surfaces of the fibers can easily combine with matrix (plastic), whereby the mechanical and thermal properties of the resulting composites are improved.

Several studies measure the biodegradation in biocomposites by means of soil burial test and testing in a bioreactor. Soil burial test comprises of placing samples in soil/ compost for long durations and testing the mechanical properties/dimensional changes/ morphology before and after soil burial. In a bioreactor, samples are placed in a composting vessel containing a mixture of compost and the percentage of biodegradation occurrence [5].

The influences of fiber content on microbial development as well as the preferential localization of microorganisms at the composite interface were analyzed by Feng et al. [15]. These authors evaluated the development of molds on HDPE composites reinforced with different mass rates of wood and bamboo fiber flour, the results obtained showed a progressive increase in the development of molds with the rate of fibers, with a better resistance for composites with wood fibres. By scanning electron microscopy, the authors observed a preferential concentration of mycelium in the interfacial zone between fibers and matrix of the composites. In addition, it appears that microbial growth has allowed a greater uptake of water mass.

In another study [16], PLA composites containing oil palm empty fruit bunch fiber was compounded with a slow releasing fertilizer and was subjected to soil burial tests at a temperature of 30°C and relative humidity of 80%. The samples were recovered at different stages of degradation and weighed to ascertain the mass loss during soil burial. The surfaces of the samples were also analyzed using scanning electron microscopy. The biodegradation rate of the samples containing fibres and fertilizer was found to be lower than that of neat PLA. The scanning electron micrographs depicted the changes that occurred during the degradation period. The surface of the composite samples exhibited traces of shrinkage and roughness and exposed the natural fiber bundles. The scanning electron micrographs also revealed the presence of cracks and holes which were produced by the degradation of oil palm fibres. Soil burial test of biocomposites from wheat gluten and rubber wood sawdust were carried out by Bootklad et al. [17]. Compression molded samples were buried in soil for 15 and 30 days and the subsequent weight loss was measured. The authors observed that this type of green biocomposites could be degraded within 15 days. During the first 15 days, the weight loss was attributed to the leaching of glycerol which was used as a plasticizer in the system. The authors also observed that the biodegradation rate of composites containing 20 weight percent of rubber-wood waste was slower than that of wheat gluten biocomposites.

In another study, Pantyukhov et al. [18] investigated the biodegradation behavior of a range of lignocellulosic filler reinforced low-density polyethylene composites. The lignocellulosic fillers included flax shives, sunflower husk, hay, birch leaves, and banana skin. Soil mixture comprising of sand, garden soil, and horse manure were prepared and samples were placed in the soil for a period of 1year. The authors observed the greatest weight loss was in the case of hay filled composites followed by lignosulfonate, husk, banana, leaves, and shives. This was attributed to the chemical, fractional, and particle size composition of the fillers.

This present work is focused on the study of the durability of different PLA/Alfa biocomposite materials treated with BYK W-980 and untreated, prepared via burial in soil aging.

2. Material and methods

2.1. Materials

The polymer used in this work is Poly (lactic acid) (2003D grade) in the form of pellets. it was obtained from Nature Works LLC, USA. Alfa used as reinforcement was collected from the arid region of Algeria. The average particles size is <80 µm, obtained using a universal laboratory grinder for plastics and wood “VERDER”. The chemical composition of Alfa was reported previously [19].

Ethanol was 99% pure purchased from Changshu Yangyuan Chemical Company (Jiansu, China). The dispersing agent (BYK W-980) has been kindly given by BYK-CHEMIE whose properties are subjected previously [19].

The aging by burial in the ground of the samples prepared was carried out according to the ISO 14851 standard.

2.2. Methods

Fourier-Transform Infrared Spectroscopy (FTIR)

The IR spectra of olive husk powder and cellulose were analyzed with a Fourier transform infra-red (FTIR) spectrophotometer (SHIMADZU FTIR-8400S). The equipment was operated with a resolution of 4 cm^{-1} and scanning range from 4000 to 400 cm^{-1}. The samples were dried firstly at 80 °C for one hour before the FTIR analyzes.

Thermo-Gravimetric Analysis (TGA)

The thermogravimetric analysis (TGA) was performed on a DSC-LINSEIS calorimeter with a temperature range between 10 and 800 °C, with a heating rate of 10 °C/min under an inert atmosphere (nitrogen).

Aerobic biodegradation

The biodegradation of biocomposites was studied by burying different test specimens in the soil (compost) of a wild dump placed in pots of yoghurt. The latter was recovered from private compost in the Ouzellaguen/Béjaia region. Six specimens of each formulation were buried at a depth of 10cm for 3 months. All tests were carried out under aerobic conditions at a temperature of 20°C.

II. Results and discussion

Fourier-Transform Infrared Spectroscopy (FTIR)

Burial aging of no-charged and charged systems is accompanied by changes in their physical and mechanical properties. This is probably due to the structural changes that aging causes. Figure 1 shows the spectra of the different samples before and after 4320h buried in the ground. As shown in Figure 1, aged and unaged samples show the same absorption bands but with different intensities. The spectrum of aged PLA (Figure 1 (a)) shows the appearance of an absorption band at 35000cm^{-1} due to the vibrations of the hydroxyl groups of water molecules under the effect of humidity in the soil [20,21]. An increase in the intensity of the bands at 1543 and 719 cm^{-1} is also observed. This increase can be attributed to the presence of moisture during aging [22].

According to the spectra of aged samples, there is an increase in the absorption bands located in the 1800-1650cm^{-1} and 800cm^{-1} region due to the vibration of the water molecules. The increase in the hydroxyl absorption band can also be attributed to the hydrolysis of the ester [23] In addition, we observe on the spectra of the biocomposites (Figure 1 (b)) that after aging there is interference of the peaks of the hydroxyl groups with CH of CH_2, this can be explained by the fact that after aging, the humidity has diffused in the biocomposite, which created a broadening of the hydroxyl peak and its shift towards lower bands due to hydrogen bonds [24].

On the other hand, we can clearly observe a decrease in peak intensity in the OH stretch, the C=O stretch and the CO stretch indicating the absence of segregation between the PLA and the Alfa fiber, after 4320h of burial in soil, for

PLA/Alfa/BYK W-980 biocomposites (Figure 1 (c)), i.e., BYK W-980 reduced the hydrogen bonds between the matrix/fiber [25-27].

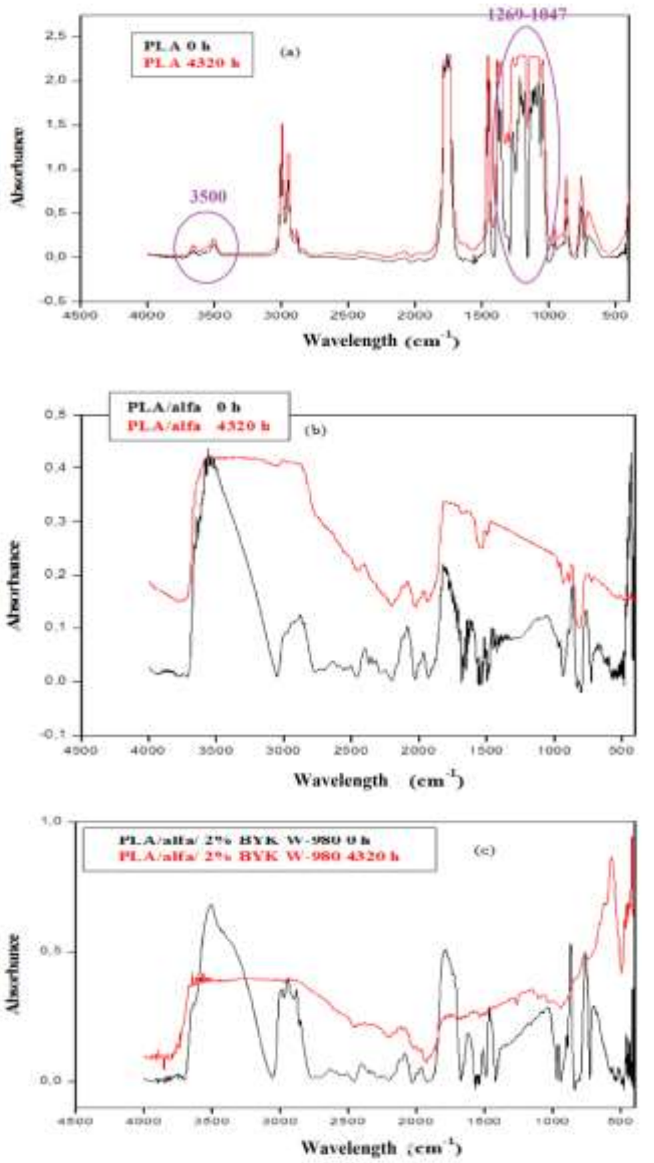

Figure 1: IRTF spectra of (a) PLA, (b) PLA / Alfa and (c) PLA / Alfa / 2% BYK W-980 before and after 4320h of burying.

Thermo-Gravimetric Analysis (TGA)

The thermal decomposition of charged and uncharged PLA before and after aging was carried out by thermogravimetric analysis (TG/DTG). From the TG thermograms of the different materials, it was possible to derive the values of the decomposition temperatures at 5, 50 and 75% ($T_{5\%}$, $T_{50\%}$ and $T_{75\%}$ respectively) of mass loss (Table 1).

Modification of PLA/Alfa biocomposites using 2% BYK W-980 influences the thermal degradation behaviour of the biocomposites. The degradation temperature at 50% mass loss ($T_{5\%}$) the TG curve of the modified biocomposite is shifted by approximately 120 °C. towards a higher temperature than that of the unmodified biocomposites. Thus, modification of PLA/Alfa biocomposites with BYK W-980 improved thermal stability. This effect is due to the stronger interaction between the fiber and the matrix with the formation of covalent bonds at the fiber/matrix interface [28].

Table1: Degradation temperature of PLA, PLA / Alfa and PLA/Alfa/2% BYK W-980 biocomposites before and after 4320h of burying

Temperature (°C)/ Formulations	PLA	PLA/Alfa	PLA/Alfa/2% BYK W-980
Before Burial			
$T_5\%$	317	240	251
$T_{50}\%$	356	306	315
$T_{75}\%$	366	322	327
After Burial			
$T_5\%$	286	213	225
$T_{50}\%$	332	296	295
$T_{75}\%$	342	317	310

Aerobic biodegradation

Biodegradation is followed by loss of mass from samples buried in soil, which is due to assimilation of the material by microorganisms [29]. Figure 2 shows the mass loss of PLA and the various biocomposites during 180 days of burial in the ground.

According to figure 2, it is noted that the loss of mass of virgin PLA is almost zero whatever the time of burial in the ground, in this chosen interval. Knowing that PLA buried in the ground is biodegradable after 4 to 5 years, only the fibers degrade [30]. We can also note that the mass loss of PLA/Alfa is much higher than PLA/Alfa/2%BYK W-980. It went from 2.39% for the biocomposites in the presence of the dispersing agent to 11.53% for the biocomposites in the absence of BYK W-980, which the microorganisms easily assimilate [31,32]. This was attributed to the reduction of the hydroxyl groups during the fiber pretreatment. The hydrophobicity of the Alfa fibers was reduced when compared to the untreated fibers. The BYK W-980 was easily coated by the PLA matrix, which improved the interfacial bonding that led to less mass loss. In contrast, biocomposites prepared with the dispersing agent BYK W-980 experience fiber distribution and scattering to the point of making their assimilation difficult [33]. In fact, the presence of BYK W-980 at the PLA/Alfa interface inhibits the penetration of water and consequently it causes interference with the action of microorganisms.

In general, the studies encountered in the bibliography are carried out on wood fiber composites and are limited to highlighting microbial development with an evaluation of changes in color or change in mass of the composites. For example, Naumann et al., [34] studied the resistance to Trametes versicolor fungi of PP composites reinforced with 55% by mass of solid beech wood. Growth of fungal mycelium was observed by light microscopy and mass loss was determined after 16 weeks of incubation at 21.5 °C on samples previously dried at 103 °C. The authors estimated a mass loss of 2.2% for composites, whereas it is 45% for solid beech wood.

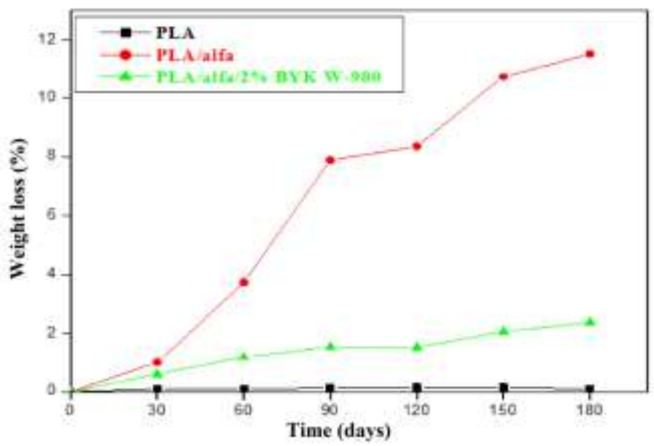

Figure 2: Mass loss rate of PLA, PLA/Alfa biocomposites and PLA/Alfa biocomposites/ 2% BYK W-980 after 180 days of burial in the ground.

The authors observed a surface fungal development following aging but which has no consequences on the chemical and mechanical properties of the composites. Other authors [35] have shown that PVC/wood fiber composites are resistant to Serpula lacrymans, this fungus is responsible for the degradation of half of the buildings constructed of wood in Europe. Thus, they recorded a mass loss after 16 weeks of aging of about 0.8% against a water mass gain of 9.2%, the authors do not specify the incubation conditions.

II. Conclusions

In this work, poly lactic acid reinforced with Alfa fibers with and without the dispersing agent which is BYK W-980 underwent burial in soil aging for duration of 4320 hours (180 days). Considering all the results, we were able to draw the following conclusions:

The study of biodegradation by burial in soil showed that the biodegradability of biocomposites prepared with untreated and treated PLA/Alfa degrade in soil more than those prepared with BYK W-980 for the same burial conditions. Knowing that the PLA buried in the ground is biodegradable after 4 to 5 years, by consecrating in this time interval of burial, we only have the fibers which degrade.

Conflict of interest. The authors report no conflict of interest.

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