

# Biodegradation process of a blend of thermoplastic unripe banana flour—polyethylene under composting: Identification of the biodegrading agent

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**ABSTRACT**: Starch-based biodegradable polymers are obtained by incorporating plant-derived polymers into plastics. This blending allows for a reduction in the polymer's resistance to microbial degradation. Assessing biodegradability is a key step in the characterization of newly designed polymers. Composting has been taken into consideration in waste management strategies as an alternative technology for plastic disposal. This study analyzed the biodegradability of an injection-molded plastic material in which thermoplastic unripe banana flour (TPF) acts as a matrix (70%) and metallocene catalyzed polyethylene acts as a reinforcing filler (30%). This plastic was termed 70 TPF, and the structural, physical, and mechanical changes associated with its degradation were analyzed. The characterization of the microorganism that contributes to 70 TPF biodegradation was also performed. After composting, 70 TPF decreased in tensile strength and the TPF moiety in the blend was lost, greatly affecting the microorganism responsible for the degradation of the plastic, a finding that supports the role of fungal communities in the biodegradation of designed materials. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2015**, *132*, 42258.

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#### INTRODUCTION

Biodegradation of neat polyethylene is a heterogeneous process. The insolubility of polyethylene in water prevents microorganisms from incorporating directly into the plastic material. Thus, a feeding stimulant for microorganisms, such as a starch or cellulose, facilitates and shortens the biodegradation process. Starch-based biodegradable polymers are obtained by incorporating natural polymers derived from cereals and tubers into plastics. In a previous work, a polymer manufactured by blending metallocenecatalyzed polyethylene (mPE) with unripe banana flour was developed.<sup>1</sup> A thermo-plasticizing process was applied to the banana flour to form a plastic matrix that incorporated into mPE. The blend containing 70% of thermoplastic unripe banana flour and 30% of mPE (70 TPF) exhibited good mechanical properties and was likely to be highly biodegradable. The characteristics of this polymer made it an ideal candidate for industrial purposes for several reasons. The mPE is an easily processed synthetic polymer with a high degree of customization and chemical homogeneity.<sup>2</sup> The addition of several biofillers such as starch, flour, and cellulose is thought to release less environmentally toxic intermediate products.<sup>3,4</sup> The natural organic materials can be degraded by microorganisms in soil.<sup>5</sup>

The characterization of the biodegradation process of 70TPF was very important for determining the potential use of the new polymer. Materials based on renewable sources can be directly studied through composting.<sup>6</sup> This is an alternative technology for disposal of plastics<sup>7–9</sup> because microbial organisms present in the compost are able to transform biopolymers through metabolic or enzymatic action into smaller fragments that can be reintegrated into the soil.<sup>10,11</sup>

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Additional biodegradability tests have been developed to quantify the ability of microorganisms to degrade polymers.<sup>12,13</sup> For instance, studies have been performed to identify the microorganisms capable of degrading polyethylene.<sup>14</sup> In the case of flour, starch, and cellulose-synthetic polymer blends, a preparation of amylolytic enzymes containing amylase and amyloglucosidase from Aspergillus niger has been tested.<sup>15</sup> The biodegradability of polycaprolactone-starch blends were tested in a variety of environments including river and lake water, sewage sludge, soil, sediment, and compost.<sup>16</sup> The identification and characterization of the main microorganism capable of degrading a blend of unripe banana flour and mPE is very important because the main feature of this material is that it is mostly composed of natural polymers. The present work analyzed the biodegradability of an injection molded plastic made of 70 TPF. The structural changes associated with degradation were analyzed. Additionally, the main microorganism that contributed to the biodegradation was characterized.

#### MATERIALS AND METHODS

#### Preparation of the Biodegradable Blend

Thermoplastic unripe banana flour (TPF) and maleic anhydride (MA) grafted into mPE (mPE-g-MA) were prepared as previously described.<sup>1</sup> Metallocene-catalyzed polyethylene Engage 8402 (Dow Chemical, Midland Michigan USA) with a density of 0.902 g/cm<sup>3</sup> (ASTM D 792) and a melt flow index of 30 dg min<sup>-1</sup> (190°C/2.16 Kg, ASTM D 1238) was used. TPF, mPE, and mPE-g-MA were blended in the following proportions: 70% TPF, 25% mPE, and 5% mPE-g-MA (hereafter termed 70 TPF).

#### Extrusion

TPF and the other components described above were extruded using a single-screw extruder (CICATA-IPN, Mexico City, Mexico) with three heating zones of 80, 120, and  $180^{\circ}$ C and an exit temperature of 200°C. Mass flow was set at 110 g min<sup>-1</sup> and screw speed was set at 30 rpm. The mass flow was controlled by a feeder (Baldor Electric, Arkansas, USA). After the extrusion, the material was milled to obtain a particle size of 4 mm<sup>2</sup>.

#### Mold Injection

Specimens (ASTM D638-08) were prepared by injection molding of the blend and neat mPE (as a control) into a Demag Ergotech 50-200 system (Düsseldorf, Germany) with 50 tons of clamping force. The injection temperatures were 140, 150, and  $160^{\circ}$ C for the heating zones and the speed of the screw was 100 rpm. The injection rate was  $30 \text{ cm}^3 \text{ s}^{-1}$ . Cooling time between cycles was 20 s.

#### Composting

The injected samples were fixed in a  $15 \times 20 \text{ cm}$  plastic grid. Five plastic grids were prepared (one for each time period). The grids were buried between two layers of Organodel compost (Agroformuladora Delta, Mexico) between a lower layer of 60 cm and an upper layer of 30 cm. To facilitate further localization, every plastic grid was settled according to the time when the sample would be withdrawn and a labeled flag indicated their location. The compost contained organic matter, humus, and humic acids. According to the producer specifications, the compost was supplemented with N, P, K, and Ca. Compost was conditioned to 50% humidity. After 25, 50, 75, 100, and 125 days beneath the compost, the grids were removed.

#### Weight Loss

Samples were retrieved from the grids and washed with deionized water. Retrieved specimens were dried for 24 h at 50°C and rested for 12 h at room temperature. All samples were weighed in an analytical balance and the percentage of weight loss was calculated as follows:

weight 
$$loss(\%) = \frac{(initial weight - final weight)}{initial weigh} \times 100$$

#### **Mechanical Properties**

The tensile properties of the injected samples were determined according to the standard ASTM D638-10 test method before and after composting. Samples were conditioned for 96 h at 23°C and 50% relative humidity prior to testing according to ASTM D618-08. The 70 TPF blend and the control samples were subjected to tensile resistance and elongation tests. Testing was performed in an Instron device (model 5583, Massachusetts, USA) with a load capacity of 150 kN and weight of 500 kg. A displacement rate of 50 mm/min was established. Bluehill software 3.11 (Illinois, USA) was used to analyze the results.

#### Measurement of Thermal Properties Using DSC

Thermal analysis was performed on a Pyris 1 Perkin Elmer differential scanning calorimeter (Norwalk, CT, USA). Dynamic scans of the neat mPE and 70 TPF specimens ( $\sim$ 8 mg) from the beginning to the final composting period were performed. Samples were placed in hermetically sealed aluminum pans and subjected to increasing heat from 30 to 400°C at a rate of 10°C min<sup>-1</sup>. Calibration was performed with indium and an empty pan was used as the reference.

#### Fourier-Transform Infrared Spectroscopy (FT-IR) Analysis

FT-IR spectra of the injected samples were obtained using an Attenuated Total Reflectance FT-IR spectrophotometer (Spectrum Two, Perkin Elmer, Massachusetts USA). The tested wavelength range was from 500 to  $4000 \text{ cm}^{-1}$ . The samples were pressed against the objective lens and analyzed directly.

#### **SEM Analysis**

For microscopic analysis of the fungi, samples  $(5 \times 5 \text{ mm})$  were cut from unwashed specimens and mounted on brass stubs with double-sided graphite-filled tape and were vacuum coated by sputtering with silver (Desk IV, Denton Vacuum). For the biodegradability analysis, specimens were washed with deionized water and then cut and mounted as described before.

Samples were chronologically observed (JSM-6390LV, JEOL, Japan) at 0 days (as a control) and at 25, 50, 75, 100, and 125 days after their period under compost.

#### Morphological Characterization of Fungi

Visible mycelia from the surface of the composting samples were recovered in a 0.9% NaCl solution. The suspension was stirred vigorously and an aliquot of 900  $\mu$ L was placed in a test tube and centrifuged at 800 rpm. The dirt-free supernatant, which contained fungi without compost remains, was transferred to



Fungus	Primer sequences	Amplicon length (base pairs)
Mortierella elongata	Forward GGTGCTATGGGATCGTTCGG Reverse TCCTAAGTGTGGAAGAGCCG	85
Cladosporium cladosporioides	Forward GGTCTAACCACCGGGATGT Reverse TTTTACGGCGTAGCCTCCC	102
Aspergillus niger	Forward GAGACCCCAACACGAACACT Reverse GCATTTCGCTGCGTTCTTCA	102

 Table I. Primers Sequences for Fungal Identification by PCR

another tube. A  $200\,\mu\text{L}$  portion of the clean suspension was placed on a slide and air-dried. The slide preparation was stained with methylene blue and then examined under an optical microscope (Binocular Sehkraft M08 series, Brea, CA, USA).

## DNA Extraction and Fungus Identification by Polymerase Chain Reaction (PCR)

DNA was extracted from samples of the compost surrounding the plastic material using a Fast DNA Kit following the manufacturer's instructions (GE Amersham, Piscataway, NJ, USA). Primers were designed with the Primer-BLAST tool of the National Center for Biotechnology Information of the US National Library of Medicine (Table I), detecting the gene sequences of 28S ribosomal RNA, ITS2, 18S rRNA, ITS1, and 5.8S rRNA for the candidate fungi Mortierella elongata and Cladosporium cladosporidium. Sequences for Aspergillus niger were included to validate the specificity of the candidate fungi primers. All primers were synthesized using Integrated DNA Technologies (Coralville, Iowa, USA). The following final concentrations or total amounts were used for PCR amplification: 10 ng of DNA, 25 mM MgCl<sub>2</sub>, each deoxynucleotide triphosphate at a concentration of 10 mM, 40 ng forward primer, 40 ng reverse primer, and 1 U of Taq DNA polymerase. The PCR reaction was performed at the following conditions: 94°C for 2 min, 35 cycles of 94°C for 1 s, 56°C for 20 s, and 72°C for 30 s. The reaction was terminated at 72°C for 2 min followed by 4°C for 10 min. PCR was performed on an iCycler (BioRad, Hercules, CA, USA). The DNA product of PCR amplification was further purified by electrophoresis on 2% agarose gels containing GelRed Nucleic Acid stain (Biotium, Hayward, CA, USA) and visualized in a ChemiDoc MP imaging system (BioRad).

#### **Residual Polymer Estimation**

The rate of weight loss was determined using linear regression analysis for both the control and the blend. A graphical extrapolation of the weight loss rate was performed to estimate total biodegradation.<sup>17</sup> Because flour is the first carbon source consumed by microorganisms, the weight loss rate associated with polysaccharides content was modeled and plotted first according to:

 $ya = 100\% - (TPF weight loss rate \times days)$ 

where ya is the residual polymer (%) after the consumption of the TPF.

After the flour was consumed, only the mPE surface remained exposed to enzymatic attack. Thus, extrapolation modeling was

continued for the predicted consumption of pure mPE and the weight loss rate of the pure mPE was plotted according to:

 $yb = ya - (weight loss rate of neat mPE \times days)$ 

where *yb* is the residual polymer (%) after neat mPE consumption.

#### **RESULTS AND DISCUSSION**

#### Weight Loss

Weight loss is one of the indicators of biodegradation.<sup>18</sup> Figure 1 shows the monitoring of sample weight loss samples during composting. The neat mPE control had a minimum weight loss after 125 days (1.2%). This insignificant effect of composting on mPE is probably due to the polymer's natural resistance to microbial attack.<sup>19</sup> The 70 TPF blend lost 40% of its weight in only 75 days out of a total of 45.23% of its weight lost after 125 days under compost. Thus, the observed rate of weight loss revealed that 70 TPF had a good tendency to deteriorate considering that the thickness of the samples was 4 mm. It is interesting to note that the 70 TPF sample lost more than half of the total weight lost in the first 50 days. These results indicated that the higher flour content increased the microbial enzymatic attack and therefore facilitated biodegradation.<sup>20</sup> This may be related to the higher content of starch and cellulose expected for the blend. Total starch in banana flours prepared from peeled bananas ranges between 61 and 76% of the dry weight, whereas the fiber content ranges between 2 and 12%; although it may be lower in our blends because the addition of the peels, it still contains a high amount of naturally biodegradable polymers.21



Figure 1. Percentage of weight loss after 125 days of composting.



Figure 2. Mechanical properties of the 70 TPF blend and neat mPE: (a) tensile resistance and (b) elongation at break.

#### **Mechanical Properties**

The loss in mechanical properties is also considered to be another factor indicating biodegradation.<sup>22</sup> As shown in Figure 2(a), the tensile strength for 70 TPF was almost half of the value for the neat mPE before composting. After 125 days of composting, the tensile resistance of the 70 TPF blend decreased with respect to its original value while the tensile resistance of neat mPE was preserved. The decrease in tensile resistance for the biodegradable material was not uniform over time; for instance, values at day 25 and day 75 were lower than the value obtained after day 125. It is possible that the distribution and size of fractures were not uniform over the time of composting as a consequence of the biodegradation process. Moreover, the functionalization of maleic anhydride contributed to the homogeneity of the 70 TPF blend prior to the composting process, but when biodegradation advanced, the localization of the microorganisms over the surface of the samples altered the conditions of sample homogeneity, resulting in variations in the tensile strength. When polysaccharides such as starch are blended with nonfunctionalized synthetic polymers, clusters of the material are formed, producing fragile blends. If these blends are composted, the reduction in tensile resistance is noticeable after a few weeks.9 Whenever a grafting process was performed, such as in mPE-g-MA, a greater tensile strength was observed.<sup>23</sup> Thus, modification of the main components prevents the formation of heterogeneous blends and enhances the plasticization effect,<sup>24</sup> which may explain the variation of the tensile strength result.

As for elongation, no substantial changes were observed in the neat mPE during the 125 days of composting [Figure 2(b)]. This result was consistent with the absence of weight loss and the undamaged condition of the sample after composting. In the case of 70 TPF, the percentage of elongation uniformly decreased from the beginning to the end of the 125 days, in contrast to the result for the sample's tensile resistance. This result suggests that the blend reduces elongation capacity while increasing biodegradation. Variations in results are also likely to occur because of uneven distribution of the biodegrading microorganisms over the sample.

#### **Thermal Properties**

The thermal behaviors of the neat mPE and the biodegradable material are depicted in Figure 3. Comparing the thermograms of each material from the beginning to the end of the composting process yielded information regarding their biodegradation. At day 0, the expected peak of the melting temperature of the neat mPE was observed. Peaks corresponding to the neat TPF (45°C) and the mPE (68°C) appeared in the 70TPF blend. The mPE peaks were slightly displaced probably because they were chemically affected by the functionalization of the mPE, which created new bonds and influenced its thermal behavior. Above 250°C, the TPF blend thermogram displayed a peculiar behavior produced by the thermal degradation at this temperature. At day 125, the neat mPE thermogram remained identical to the thermogram obtained at day 0. The thermal behavior of the TPF blend sample at temperatures above 250°C decreased.





Figure 3. Comparative DSC thermograms obtained at the beginning (0 days) and end (125 days) of the composting process.

The peak at 45°C corresponding to the pure TPF and the peaks above 250°C almost disappeared. Taken together, this behavior could be associated to the biodegradation and loss of the TPF moiety after 125 days of composting. It was not likely that our material suffered thermal degradation during the manufacturing process, because our blend reached the maximum temperature at extrusion (200°C) and injection was performed at 160°C. The thermal degradation of banana starch, measured by TGA analysis, occurs at 240°C<sup>25</sup>; the thermal degradation of small amounts of starches mixed with glycerol may occur before  $230°C^{26}$ ; cellulose, the main constituent of the banana peels, degrades at 290°C.<sup>27</sup> Biodegradation explains better the thermal behavior of our material.

#### FT-IR Analysis

Infrared spectroscopy is considered to be an important technique for assessing the biodegradation of polymers.<sup>28</sup> Figure 4 shows the FTIR spectra in the range of  $4000-400 \text{ cm}^{-1}$  for the 70 TPF sample. For the blend, day 0 is depicted in black and day 125 is in red. The characteristic peaks of the TPF were as follows: a broad absorption band was due to the stretching



Figure 4. Infrared analysis of the 70 TPF sample before and after composting.

vibration of the hydroxyl group (–OH) in the cellulose, starch, and glycerol contained in the flour  $(3307-3292 \text{ cm}^{-1})$ ; the hemiacetal bonds arose from the starch flour  $(1638-1650 \text{ cm}^{-1})$ ; the vibration peak corresponded to the cyclic –C–C– bond of the glucose  $(1030-1018 \text{ cm}^{-1})$ ; and the 1151, 924, and  $860 \text{ cm}^{-1}$  peaks corresponded to the fingerprint region of the banana starch. The mPE in the blends contributed to additional peaks in the spectra consisted of the following: the symmetric and antisymmetric stretching bond vibrations of the –CH<sub>2</sub> and CH<sub>3</sub> groups (2918 and  $2850 \text{ cm}^{-1}$ ) in mPE<sup>29</sup>; the characteristic –CH<sub>2</sub> stretching indicating the oxidation of synthetic polymers (1460 cm<sup>-1</sup>); a band at  $1375 \text{ cm}^{-1}$  indicating the angular bend of the –CH<sub>3</sub> group; and the presence of a 719 cm<sup>-1</sup> band typical of polyethylene.

Once the sample was buried in compost for 125 days, the infrared spectra for the sample changed. The broad peak of the group  $-OH (3423 \text{ cm}^{-1})$  markedly softened. Flour constituents such as



Figure 5. Chronological microstructural analysis by SEM (1000×) of the composted samples over 125 days.

### Applied Polymer



Figure 6. Unwashed specimens. Filamentous fungi were observed as whitish fluff over the samples.

starch and cellulose are excellent source of carbon for microorganisms,<sup>30</sup> and it was assumed that microorganisms, specifically fungi, consumed the flour moiety of the blends as a carbon source. This was evidenced by the reduction in the -OH band in the spectrum. The two peaks corresponding to the bond vibrations of the  $-CH_2$  and  $CH_3$  groups in mPE (2970–2860 cm<sup>-1</sup>) were intensified and sharpened due to the resistance of mPE to biodegradation. Hemiacetal bonds bands remained unchanged. The peak corresponding to the cyclic -C-C- of glucose (1010 cm<sup>-1</sup>) in the starch was also weakened.

#### SEM Microstructural Analysis

SEM visualization of the composted samples was performed to track the changes associated with biodegradation because of material surface irregularities along with fissures and cracks that usually arise from environmental microorganism-mediated erosion.<sup>31,32</sup> As expected, neat mPE remained unchanged for the first 100 days of the composting process (Figure 5); by day 125, however, some deterioration of the surface was observed. The conservation of the microstructure correlated with the lack of weight loss. The compact structure of the 70 TPF sample started deteriorating at day 25, and by day 125, the microstructure of the sample was greatly affected.

## Identification of the Fungus Growing Over the Plastic Samples

The active component involved in the biodegradation and conversion processes during composting is the resident microbial community. Fungi play a critical role in these communities because they can use many carbon sources primarily consisting of lignocellulosic polymers and can survive in extreme conditions. Fungi are also responsible for compost maturation. The genera with the highest load and number of species in composts are *Penicillium*, *Aspergillus, Cladosporium, Acremonium*, and *Mortierella*.<sup>33</sup>

After day 50, filamentous fungi were observed as a whitish fluff spread over the TPF blend samples buried in the compost



Figure 7. SEM micrographs. Fungi formed biofilms directly on the surface of the composted TPF-mPE blend materials.





Figure 8. Optical microscope micrographs. Fungi were obtained from the biofilm on the surface of the TPF-mPE blended material after the composting period.

(Figure 6). A mass of branching hyphae, called mycelia, were present over the TPF blend but not over the neat mPE. Because the mycelia were visible on the TPF blend, the question of whether the fungi formed a biofilm directly on the surface of the material was also investigated by SEM analysis of unwashed samples (Figure 7). A single fungus was found growing over the plastic material throughout the composting time. Figure 7(a–d) shows the fungus growing over the sample after 75 days of composting. The fungal spores (conidia) were evident inside the cavities of the partially degraded polymer [Figure 7(e,f)]. By day 125, there were no visible filaments (hyphae) and only



Figure 9. Agarose gel electrophoresis. Af: *Aspergillus fumigatus*, Cc: *Cladosporium cladosporioides*; Me: *Mortierella elongata*, amplified as an 85 bp band. Unused primers appear at the front.

spores were observed, which was probably due to the loss of humidity in the compost [Figure 7(g,h)].

To identify the fungus responsible for the degradation of the 70 TPF sample, further morphological description was required. The conidia and hyphae morphology are genus-restrictive and would provide a candidate fungus. Under light microscopy (Figure 8), the conidia exhibited a small cavity and were thickened, darkened, and refractive, probably caused by diminished turgidity and dehydration. Under SEM, hyphae were thin and unbranched or sparingly branched, approximately  $1-5 \,\mu m$ wide, septate, smooth, and thin-walled. Conidiophores were solitary, macronematous or semimacronematous, narrowly cylindrical, non-nodulose, unbranched or occasionally branched with mostly short branches, and smooth toward the base. Conidia were numerous; catenate; in long branched chains with up to 10 conidia in the upper unbranched part; branching in all directions; with small, terminal conidia subglobose; ovoid to limoniform; aseptate; approximately between 3 and 7  $\mu$ m; occasionally had a small cavity in the cells; and occasionally had roughwalled irregularities. Intercalary conidia were ellipsoid-ovoid; aseptate; and smooth.

The microscopic morphology suggested two candidates: *Cladosporium cladosporidium* and *Mortierella elongate.*<sup>34</sup> The identity of the fungus was corroborated using PCR with specific primers for the two candidate fungi species. Sequences for *Aspergillus niger* were included to validate the specificity of the candidate fungi primers. We found that *Mortierella elongata* was the fungus growing over the plastic samples (Figure 9). Because the material was to be analyzed by several properties, the sample



Figure 10. Total biodegradation by weight loss extrapolation. (a) Graphical extrapolation; (b) sixfold factor mathematical model.

retrieved for DNA identification contained small amounts of DNA (lane 4) and the amplification band appear less bright than the size marker bands, but it still indicates specificity of amplification; therefore, *Mortierella* was positively detected.

Although they are considered nonbiodegrading microorganisms, soil fungi such as Aspergillus fumigatus, Aspergillus terreus, and Fusarium solani have been reported to degrade synthetic polymers, especially by stimulating other microbial communities.35-39 However, additional factors or a limited carbon source are prerequisites for stimulating microbial communities. Mycelia growing over neat mPE were not observed, indicating that the Motierella elongata fungus responsible for the degradation of the plastic samples was not degrading mPE but was degrading the organic component of the blend. Amylases produced by soil microorganisms hydrolyze starch<sup>40</sup> and the activity of fungal species improves with humidity, gas concentrations, and degree of penetration into the substrate.<sup>41</sup> The deep penetration of the fungus into the blend probably greatly contributed to its degradation by digesting the plant-derived moiety. Therefore, the addition of plant-derived compounds into designed biodegradable plastics would accelerate the degradation of thick materials as well as films.

*Mortierella* has been reported to be present not only in soils but also in compost formulations.<sup>42</sup> Moreover, *Mortierella* has proved to be useful for bioremediation of polluted soils because it ensures the elimination of pesticides and allows bacterial communities to spread over polluted soil, enhancing degradation of contaminants.<sup>43</sup> The introduction of specific degrading microorganisms into polluted soil or landfills is a promising remediation method provided that the organisms survive and spread in the environment. Our results suggest that *Mortierella elongata* is a promising candidate microorganism for use in degradation processes of biodegradable plastics such as the one designed in this study.

#### **Residual Polymer Extrapolation**

Because plastics require years, decades, and even centuries to reach complete degradation, it would be practically impossible and unaffordable to design an experiment over that time span. Thus, a graphical-mathematical model can help to establish an approximate biodegradation time for new materials. In this study, the samples studied were predicted to follow the 125 days pattern for longer periods until complete degradation. To calculate the complete degradation time, a weight loss rate was calculated and then extrapolated. Figure 10(a) initially shows a pronounced negative slope for 70 TPF, indicating the fast consumption of the natural component in the material by the microorganisms. Then, the change in the slope marks the slow rate biodegradation of the remaining mPE. A continuous curve was observed for neat mPE, indicating a very slow degradation. According to Figure 10(a), a 4-mmthick mPE sample is expected to reach complete degradation in 11,364 days (31.133 years). This period of time seems short for an inert material, and could be due to the type of compost and the advantageous properties of the polyethylene type. According to the sixfold mathematical method, the sample containing 70% of TPF would need 3567 days (9.77 years) to reach complete degradation. Because the samples are not films, the thickness of each sample must be considered in calculating the time they require for total biodegradation. The result is encouraging and desirable because companies that produce existing biodegradable plastic materials would shorten their estimated times of degradation with a highly positive impact on marketing.

A previous study<sup>44</sup> reported that when the natural polymer in a biocomposite is completely degraded by microorganisms, the weight loss rate of the remaining synthetic polymer is expected to increase sixfold when the original natural polymer concentration was at least 50% of the total blend. The newly exposed



surface generates this phenomenon. Mathematical arrangements for modeling this situation were made as follows:

 $yb=ya-(weight \ loss \ rate \ neat \ PE \times \ days \times \ six \ fold \ factor)$ 

where yb is the percentage of biodegradation (%). The new graphical extrapolation is depicted in Figure 10(b). The total biodegradation time is as follows: the sample containing 70% TPF would need 732 days (2 years) to reach complete degradation. This is an outstanding and promising result that would help to solve industrial and ecological issues.

#### CONCLUSIONS

A biodegradable material consisting of a polyethylene blend containing 70% thermoplastic unripe banana flour (70 TPF) was developed. The physical and mechanical properties of 70 TPF allowed for the manufacture of 4-mm-thick samples. The blend was subjected to 125 days of composting in order to assess its biodegradability. The blend exhibited a noteworthy behavior, losing 40% of its weight in only 75 days. Mathematical modeling predicted that 70 TPF would need approximately 2 years to reach complete degradation. After composting, the tensile resistance of 70 TPF decreased at maximum load, the TPF moiety in the blend was lost, and the microstructure of the sample was greatly disorganized. Together with these indicators of degradation, the fungus Mortierella elongata grew over the samples and inside the fractures of the samples, supporting the role of fungal communities in the biodegradation of the designed material. The neat mPE remained unaffected by the composting and was devoid of fungal colonization. Both the utilization of unripe banana flour in the preparation of polyethylene blends and the inoculation of landfills and compost with Mortierella elongata are promising technologies that would help to solve industrial and ecological issues surrounding plastic disposal.

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