**Highlights in BioScience**
**ISSN:2682-4043**
DOI:10.36462/H.BioSci.20246

**Research Article**

**Open Access**

[1] Department of chemistry, Faculty of Education, University of Khartoum, Sudan.

*** To whom correspondence should be addressed: nawal.mahgoub@uofk.edu**

**Editor:** Alsamman M. Alsamman, *International Center for Agricultural Research in the Dry Areas (ICARDA), Cairo, Egypt.*

**Reviewer(s):**
Khaled H. Radwan, *National Biotechnology Network of Expertise, ASRT, Cairo, Egypt.*.

Tamer Ahmed Elakkad, *Faculty of Agriculture at Moshtohor, Benha University, Benha, Egypt.*

**Received:** May 3, 2024
**Accepted:** November 20, 2024
**Published:** December 27, 2024

**Citation:** Suleman NM, Eltyeb EM. The effectiveness of bentonite and esterified plant fibers on the biodegradability of 24 polypropylene biocomposites . 2024 Dec. 27;7:bs20246



**Data Availability Statement**: All relevant data are within the paper and supplementary materials.
**Funding:** The authors have no support or funding to report.
**Competing interests:** The authors declare that they have no competing interests.

# The effectiveness of bentonite and esterified plant fibers on the biodegradability of 24 polypropylene biocomposites

Nawal M. Suleman [1], Eiman M. Eltyeb[1]

## Abstract

This study aims to evaluate the biodegradability of 24 bio-composites synthesized from polypropylene and esterified plant fibers, with bentonite serving as a filler. The biodegradation process was conducted using *Aspergillus niger*. The study utilized standard laboratory equipment and the melt flow indexer. After three months of biodegradation, all biocomposites under investigation showed a significant loss in their weights. This is a logical observation because *Aspergillus niger* consumed parts of biocomposites as food during the biodegradation. The study also evaluated properties including density, melting point, melt flow rate (MFR), melt viscosity, molecular weight, and water absorption capacity. Densities, melting range temperature, and molecular weights were decreased (decreasing in molecular weights). The values of biodegraded composites after the biodegradation process for three months are in good agreement with the fact that the molecular chain breaks and the chain length shortens after any degradation process. Water absorption capacities were significantly increased (due to the holes resulting from the *Aspergillus niger* attack) for all biocomposites under study; this is considered very good evidence for biodegradation.

**Keywords:** Bio-composite, biodegradation, *Aspergillus niger*, bentonite, polypropylene

## Introduction

Biodegradable materials decompose within a year in their usual disposal environments, converting into non-toxic substances through the activities of living organisms [1]. The biodegradability of polymers is influenced by two main factors. The environmental conditions are exposed to (biotic or abiotic) and the polymer's characteristics (such as mobility, crystallinity, density, molecular weight, functional groups, and additives). Abiotic factors (e.g., pH, temperature, light, and moisture) can change the hydrolysis reaction rates. Biotic factors, such as extracellular enzymes produced by distinct microorganisms, can biodegrade certain polymers. For instance, *Aspergillus niger* and *Aspergillus flavus* fungi produce enzymes that more easily break down aliphatic polyesters composed of 6 - 12 carbon di-acid monomers than those produced from other monomers [2]. Most conventional plastics (polypropylene) are non-biodegradable, and their accumulation in the environment has endangered the planet. To resolve all these challenges, several strategies have been utilized. Firstly, it involves the degradation of some petrochemical-based plastics by biological processes. Secondly, the production of plastics with a high degree of degradability. The aliphatic polyesters are regular plastic polymers that show high potential for use as biodegradable plastics [3]. Polypropylene is a challenging substrate for biodegradation by microorganisms due to its high molar mass and inability to pass through microorganisms cell membranes [4]. Biodegradation of polypropylene/starch or polypropylene/cellulose composites has been observed, where organisms easily degrade starch or cellulose, producing polymer waste. These carbohydrates or fillers increase the adhesion of organisms to the surface of the polymer [5]. Protocols such as surface changes and changes in the mechanical and physical properties of the polymer were utilized to characterize the biodegradability of composites [6]. This study aims to test the biodegradability of 24 composites previously synthesized from polypropylene and esterified fibers extracted from Khimp (*Leptadenia pyrotechnica*) and date palm tree (*Phoenix dactylifera L.*) and esterified using citric and adipic acid (AAD).

## Materials and Methods

The analytical chemicals used were polypropylene (9003-07-0, Sigma-Aldrich), bentonite clay (from India Mart), adipic acid, citric acid, sulfuric acid, acetic anhydride, sodium hydroxide, peptone, glucose, ethanol, agar, and hydrochloric acid (from BDH, India). Toluene (108-88-3, Sigma-Aldrich), xylene (1330-20-7, Sigma-Aldrich), and deionized water (the bio-composites of polypropylene were prepared previously) [7]. A computerized, fully automated melt flow rate (MFR) tester was sourced from India Mart.

### Biodegradation method

Biodegradation takes place according to the following equations:

$$\text{(A): } C_{\text{plastic}} + O_2 \rightarrow CO_2 + H_2O + C_{\text{residual}} + \text{Biomass}$$

$$\text{(B): } C_{\text{plastic}} \rightarrow CH_4 + CO_2 + H_2O + C_{\text{residual}} + \text{Biomass}$$

A: Aerobic biodegradation, B: Anaerobic biodegradation.

The ingredients of Sabouraud dextrose agar (SDA) (10g peptone + 40g glucose + 15g Agar) were combined in 900 ml of deionized water, then the pH was adjusted to 5.6 with HCl, and the final volume was adjusted to 1000 ml by adding deionized water. The mixture was boiled to completely dissolve the medium, sterilized at 121°C for 20 min by autoclave, then warmed to 50°C and distributed into petri dishes. The ingredients of Sabouraud dextrose broth (20g glucose and 10g peptone) were suspended in 1000 ml of deionized water, heated to dissolve the medium completely, and the mixture was sterilized by autoclave at 121°C for 15 min. The sterilization of polymer samples was conducted under UV light and transferred into the middle of petri dishes. The culture broth was replaced with 50 ml of sterile nutrient broth every 10 days to enhance healthy microbial growth. After three months, biodegraded composites were washed, dried, and weighed to determine the weight loss caused by microbial action during biodegradation. The composites were then subjected to characterization [8].

### Characterization of the Biodegraded Composites

Biodegraded composites were characterized by the same instruments and methods described for characterization of the synthetic composites and properties [7]. The measured properties that were characterized include density, melting points, melt flow rate (MFR), melt viscosity, molecular weight, and water immersion test.

## Results and Discussions

### Biodegradation of Composites

Providing sustainable alternatives to conventional plastics is indispensable. Therefore, researchers and manufacturers are attempting to integrate sustainable and biodegradable polymers into industrial processes [9]. Regarding the environmental concerns, this study was conducted. Biodegradation was carried out according to the literature review. (*Aspergillus niger*) were cultivated in order to provide a good culture for biodegradation (**Figure 1**) shows some petri dishes prepared for this purpose. During biodegradation, microorganisms start utilizing the polymer surface as a food source and grow on it. Various polymer characteristics influence biodegradation, such as their tactility, crystallinity, molecular weight, functional group types, the type of microorganism, and the pre-treatment method [10]. This clearly occurred for all samples under investigation. The esterified fibers used as reinforcement agents in the synthesis of the target composites introduced new functional groups to the polypropylene resin and hence enhanced the biodegradability in an acceptable way because the availability of functional groups like ester increases hydrophilicity. This ester group not only increases the hydrophilicity of the synthetic composites, but it is also an easily breakable bond compared to the carbon-carbon bonds in polypropylene. Biodegradation occurred differentially depending on the amount and type of esterified fiber.

**Figure 1.** Petri dishes showing microbial colony diversity during biodegradation.

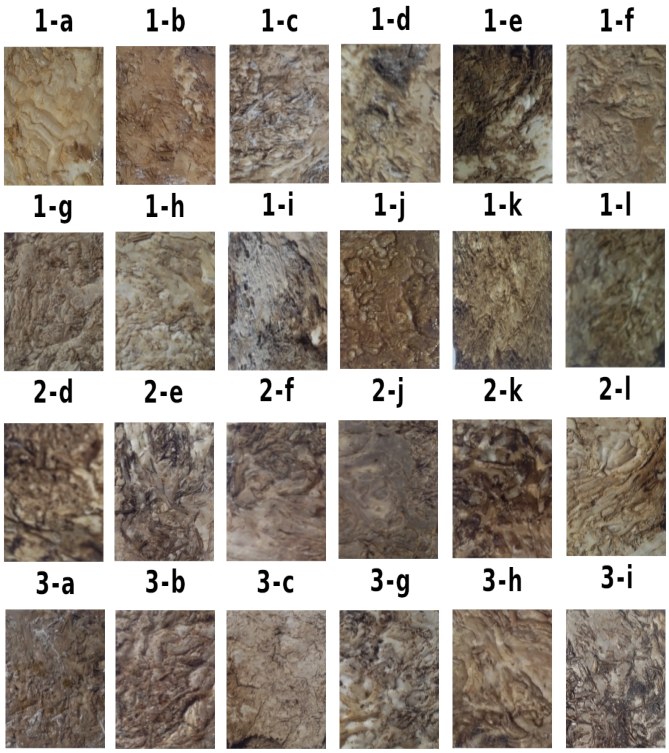


**Figure 2.** Biodegraded composites showing structural changes.

**Table 1.** Characteristics of 24 biodegraded composites under study

| No. | Composite entity code | Density (g/cm$^3$) | Melting range (řC) | Melt flow rate | Melt viscosity | M-W | Water absorption capacity (%) |
|---|---|---|---|---|---|---|---|
| 1 | 1-a | 0.84 | 165-166 | 5.02 | 0.86 | 1.8 x $10^5$ | 10.3 |
| 2 | 1-b | 1.00 | 170-173 | 8.3 | 0.82 | 1 x $10^5$ | 15.3 |
| 3 | 1-c | 0.92 | 169-173 | 4.68 | 0.87 | 0.8 x $10^5$ | 32 |
| 4 | 1-d | 0.89 | 170-175 | 4.91 | 0.87 | 0.8 x $10^5$ | 24 |
| 5 | 1-e | 0.93 | 168-169 | 1.54 | 0.90 | 0.3 x $10^5$ | 28.3 |
| 6 | 1-f | 0.82 | 175-177 | 6.9 | 0.81 | 0.9 x $10^5$ | 15.6 |
| 7 | 1-g | 0.82 | 169-172 | 3.55 | 0.89 | 0.6 x $10^5$ | 36.5 |
| 8 | 1-h | 0.87 | 160-163 | 6.2 | 0.85 | 0.9 x $10^5$ | 25.4 |
| 9 | 1-i | 0.95 | 164-170 | 2.69 | 0.90 | 0.5 x $10^5$ | 15.9 |
| 10 | 1-j | 1.00 | 168-171 | 10.6 | 0.80 | 1.1 x $10^5$ | 11 |
| 11 | 1-k | 0.87 | 165-167 | 2.11 | 0.93 | 0.4 x $10^5$ | 24.2 |
| 12 | 1-l | 0.83 | 175-178 | 6.06 | 0.84 | 0.9 x $10^5$ | 44 |
| 13 | 2-d | 0.84 | 160-163 | 10.5 | 0.80 | 1.1 x $10^5$ | 8.75 |
| 14 | 2-e | 0.80 | 165-168 | 2.71 | 0.91 | 0.5 x $10^5$ | 10 |
| 15 | 2-f | 1.00 | 161-165 | 4.72 | 0.87 | 0.8 x $10^5$ | 18.9 |
| 16 | 2-j | 0.82 | 167-171 | 5.37 | 0.85 | 0.8 x $10^5$ | 18.8 |
| 17 | 2-k | 0.95 | 170-174 | 6.17 | 0.82 | 0.9 x $10^5$ | 15 |
| 18 | 2-l | 0.92 | 170-172 | 8.5 | 0.82 | 1 x $10^5$ | 20 |
| 19 | 3-a | 0.94 | 174-178 | 8.2 | 0.82 | 1 x $10^5$ | 33 |
| 20 | 3-b | 0.92 | 172-177 | 5.41 | 0.86 | 0.8 x $10^5$ | 12.2 |
| 21 | 3-c | 0.86 | 169-171 | 4.9 | 0.86 | 0.8 x $10^5$ | 26 |
| 22 | 3-g | 1.00 | 169-172 | 6.4 | 0.84 | 0.9 x $10^5$ | 13.3 |
| 23 | 3-h | 0.80 | 167-170 | 3.34 | 0.89 | 0.6 x $10^5$ | 12.3 |
| 24 | 3-i | 0.90 | 170-172 | 1.29 | 0.97 | 0.2 x $10^5$ | 25 |

Additionally, using bentonite as a filler provides a good environment for microorganisms; all biodegraded samples showed an amazing change in their surfaces (**Figure 2**). In addition to the surface change, there are many physical properties that indicate biodegradation occurrence, such as density, water absorption capacity, molecular weight, melt flow rate, and melt viscosity. These properties will be discussed in detail after the characterization step. The loss of weight in 24 biodegraded composites is presented in (**Tables S1 to S4**), quantifying the level of composite biodegradation after three months of *Aspergillus niger* incubation. The weights of composites before and after three months were measured. Weights lost percentages were calculated according to the following equation:

$$\text{Weight lost (\%)} = \frac{(w_o - w)}{w_o} \times 100$$

$w_o$ = weight before biodegradation and w = weight after biodegradation.

A crucial aspect of our experiment is the impact of clay nanoparticles on biodegradation; the presence of clay increases microorganisms' attack, thus enhancing the biodegradation process due to microorganisms' preference for clay media for growth, as reported in the literature. Weights lost were clearly observed for all composites and increased with increasing ester content, either in the presence or absence of clay, except for samples 1-i, 1-k, 2-f, 2-k, and 3-b. This may have occurred due to the dispersion of either esterified fibers or clay among polypropylene resin during the injection molding process. Another important observation is that composites made from sample one, esterified by citric acid, showed significant increases in weight loss compared to their analogs made from the same sample esterified by adipic acid.

## Characterization of Biodegraded Composites

The densities of the biodegraded composites were determined based on the guidelines provided in [11]. Densities of composites decreased clearly after the biodegradation process because microorganisms consume a part of the composite for their growth; this is very good evidence of biodegradation occurrence (**Table 1**), and the melting range temperature of the 24 biodegraded composites was measured experimentally as described in [11]. The resulting melts of all composites under study were thick liquids. Biodegraded composites melted at lower melting ranges than those of synthetic composites.

The melt flow rate is a characterization method used to eval-

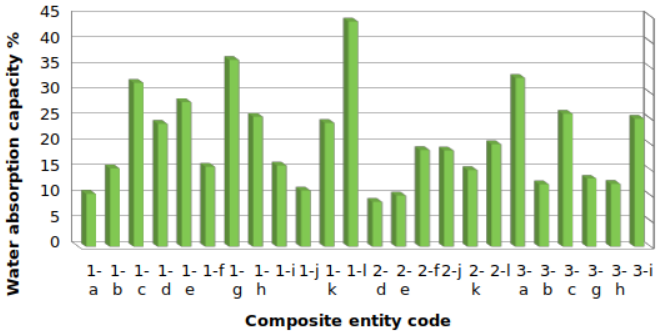


**Figure 3.** water absorption capacities for the biodegraded composites samples

uate the degradation process of biodegraded composites by calculating the reduction in molecular weight for each composite. (**Table 1**) represents the values of the melt flow rate for the 24 composites under investigation. For determining melt viscosities, the same equation described for synthesized composites was used as follows:

$$[\eta] = 0.996 - 0.189 \log(\text{MFR})$$

[η]: Intrinsic Viscosity, MFR: Molecular Flow Ratio or Melt Flow Rate. The average molecular weight of each biodegraded composite was determined using the same equation applied for the average molecular weight of synthesized composites.

$$\log[\text{MFR}]_{\text{composite}} = [0.454 \times 10^{-6}]\text{M}_{\text{w}} - 0.1273$$

Values of the average molecular weight for the biodegraded composites are shown in Table 5. According to the above table, molecular weights of biodegraded composites were decreased significantly. The water immersion test demonstrates that all biodegraded composites absorbed some amount of water over time; however, they were insoluble in water. Water absorption capacity (WAC) was calculated according to the equation clarified by [12].

$$\text{WAC}\ (\%) = \left(\frac{(m - m_o)}{m_o}\right) \times 100$$

WAC (%): Water absorption capacity, $m_o$: weight of dry sample, and $m$: weight of sample after 24-hour immersion in distilled water. There is a strong correlation between molecular weight and water absorption capacity of all biodegraded composites; biodegraded composites with lower molecular weight have more water absorption capacities than those with comparatively high molecular weight. This is considered a reasonable correlation because when microorganisms attack the composite surface in the biodegradation process, many holes occur as a result of this action, allowing water to penetrate. The decrease in molecular weights and the increase in water absorption capacities provide strong evidence for the occurrence of biodegradation **(Figure 3)**.

## Supplementary

Table S1: Weights lost for composites of Khimp fiber esterified by adipic acid. Table S2: Weights lost for composites of Khimp fiber esterified by citric acid. Table S3: Weights lost for composites of date palm leave fiber esterified by citric acid. Table S4: Weights lost for composites of date palm fiber esterified by adipic acid.

## Reference

1. Leja K, Lewandowicz G. Polymer biodegradation and biodegradable polymers-a review. Polish Journal of Environmental Studies. 2010;19(2).

2. Devi RS, Kannan VR, Natarajan K, Nivas D, Kannan K, Chandru S, et al. The role of microbes in plastic degradation. Environ waste manage. 2016;341:341-70.

3. Tokiwa Y, Calabia BP, Ugwu CU, Aiba S. Biodegradability of plastics. International journal of molecular sciences. 2009;10(9):3722-42.

4. de Carvalho CL, dos Santos Rosa D. Polypropylene biodegradation. PROPERTIES, USES AND BENEFITS. 2016:141.

5. Arutchelvi J, Sudhakar M, Arkatkar A, Doble M, Bhaduri S, Uppara PV. Biodegradation of polyethylene and polypropylene. 2008.

6. Markl E, Grünbichler H, Lackner M. PHB-bio based and biodegradable replacement for PP: a review. Nov Tech Nutr Food Sci. 2018;2(5):206-9.

7. Eltyeb EM, Mohammed AA, Suleman NM. Polypropylene and some Esterified Plants' Fibers Bio-based Composites. 2023.

8. Ebadi-Dehaghani H, Barikani M, Borhani S, Bolvardi B, Khonakdar HA, Jafari SH, et al. Biodegradation and hydrolysis studies on polypropylene/polylactide/organo-clay nanocomposites. Polymer Bulletin. 2016;73:3287-304.

9. Mysiukiewicz O, Barczewski M, Skórczewska K, Matykiewicz D. Correlation between processing parameters and degradation of different polylactide grades during twin-screw extrusion. Polymers. 2020;12(6):1333.

10. Luthra P, Vimal KK, Goel V, Singh R, Kapur GS. Biodegradation studies of polypropylene/natural fiber composites. SN Applied Sciences. 2020;2:1-13.

11. Suleman NM, Eltyeb EM, Mohammed AA. Polypropylene and some Esterified Plants' Fibers Bio-based Composites. IJFAC (Indonesian Journal of Fundamental and Applied Chemistry). 2023;8(1):48-54.

12. Kudahettige-Nilsson RL, Ullsten H, Henriksson G. Plastic composites made from glycerol, citric acid, and forest components. BioResources. 2018;13(3):6600-12.