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Table of contents

Periodical:
Advanced Materials Research

Volume:
Advanced Materials Science and Technology

Papers published in this volume:
The Role and Prospect of Nanomaterials in Polymeric Membrane for Water and Wastewater Treatment: A State-of-the-Art Overview
Ahmad Fauzi Ismail, Pei Sean Goh p.3

Synthesis of Low Fouling Porous Polymeric Membranes
Heru Susanto, Dwi Putri Julyanti, Anis Roihatni p.7

Mass Production of Stacked Styrofoam Nanofibers Using a Multinozzle and Drum Collector Electrospinning System
Muhammad Miftahul Munir, Ade Yeti Nuryantini, Iskandar, Tri Suciati, Khairurrijal p.20

Transparent and Conductive Fluorinated-Tin Oxide Prepared by Atmospheric Deposition Technique
Agus Purwanto p.24

Gas Sensing Using Static and Dynamic Modes Piezoresistive Microcantilever
Ratno Nuryadi, Lia Aprilia, Nuning Aisah, Djoko Hartanto p.29

One-Step Fabrication of Short Nanofibers by Electrospinning: Effect of Needle Size on Nanofiber Length
Indra Wahyudhina Fathona, Khairurrijal, Akihiro Yabuki p.33

Carbon Dioxide Permeation Characteristics in Asymmetric Polysulfone Hollow Fiber Membrane: Effect of Constant Heating and Progressive Heating
Muhamad Azwar Bin Azhari, Nooririnah Binti Omar, Nuzaimah Binti Mustaffa, Ahmad Fauzi Ismail p.37

Electrospinning of Poly(vinyl alcohol)/Chitosan via Multi-Nozzle Spinneret and Drum Collector
Ade Yeti Nuryantini, Muhammad Miftahul Munir, Muhamad Prama Ekaputra, Tri Suciati, Khairurrijal p.41

A Simple Way of Producing Nano Anatase TiO₂ in Polyvinyl Alcohol Fibers
Sabarmaha Harsojo, Kwat Triyanaha, Harina Sosiasi p.45

Synthesis of Hydroxyapatite Nanoparticle from Tutut (Bellamya javanica) Shells by Using Precipitation Method for Artificial Bone Engineering
Lenita Herawaty, Eti Rohaeti, Charlena, Sulistioso Giat Sukaryo p.284

Adsorption of Lead Ions onto Citric Acid Modified Rubber (Hevea brasiliensis) Leaves
Shariff Ibrahim, Megat Ahmad Kamal Megat Hanafiah, Faisal Fadzil  
Effect of Deacetylation on Characterization of pH Stimulus Responsive Chitosan-Acrylamide Hydrogels Using Radiation  
Kris Tri Basuki, Deni Swantomo, Sigit, Kartini Megasari  

Synthesis of Smart Biodegradable Hydrogels Cellulose-Acrylamide Using Radiation as Controlled Release Fertilizers  
Deni Swantomo, Rochmadi, Kris Tri Basuki, Rahman Sudiyo  

Structural Studies of 1,3:2,4-Dibenzylidene Sorbitol Gels  
Hiroyuki Takeno, Yuta Kuribayashi  

Effects of Surface Treatments on Nata de Cassava on the Tensile Strength and Morphology of Bacterial Cellulose Sheet  
Dini Cahyandari, Heru Santoso Budi Rohardjo  

Impact and Thermal Properties of Unsaturated Polyester (UPR) Composites Filled with Empty Fruit Bunch Palm Oil (EFBPO) and Cellulose  
Elmer Surya, Michael, Halimatuddahliana, Maulida  

Biodegradation of Low Density Polyethylene (LDPE) Composite Filled with Cellulose and Cellulose Acetate  
Halimatuddahliana, Ahmad Mulia Rambe  

The Treated Rice Straw as Potentially Feedstock of Wood and Rice Straw Fiber Blend for Pulp and Paper Making Industry  
Ariadne L. Juwono, Handoko Subawi  

Experimental Studies of Thermo-Induced Mechanical Effects in the Main-Chain Liquid Crystal Elastomers  
Supardi, Sabarman Harsojo, Yusril Yusuf
Biodegradation of Low Density Polyethylene (LDPE) Composite filled with Cellulose and Cellulose Acetate

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Abstracts. A comparative study was conducted for the biodegradation of low density polyethylene (LDPE) composites filled with cellulose (C) and cellulose acetate (CA). Composites were prepared with the content of each filler of 10\% (by weight) using an extruder at processing temperature of 125\degree C. Biodegradation processes were done by burying in the soil and by hanging in an open environment for four months. The percentage of weight loss of pure LDPE and composites due to the degradation was observed based on the weight reduction of the composites and supported by scanning electron microscopy (SEM). The results indicated pure LDPE was not susceptible to microbial attack as the percentage of weight loss was constant. However, the composites filled with cellulose were relatively more susceptible to degradation as compared with composites filled with cellulose acetate. Here, the percentage of weight loss of composites filled with cellulose was higher than the composites filled with cellulose acetate. On the other hand, the biodegradation processes by hanging in open environment were relatively faster than burying in soil for both types of composites. These results which were confirmed by SEM show that the composites have some cavities.

Introduction

Increasing the use of natural fibers such as jute, kenaf, banana, bamboo, and rice husk as fillers and reinforcements of composite materials continues to grow both in terms of industrial applications as well as fundamental research [1-5]. They are renewable, biodegradable, able to improve composite mechanical properties, and able to increase composite range of applications. If they are in contact with biodegradable polymers such as starch, cellulose plastics, poly(hydroxyalkanoate) (PHA), poly(lactic acid) (PLA), and polycaprolactone are they are fully or completely biodegradable [6-8]. If they are in contact with non-biodegradable thermosetting or thermoplastic polymers such as epoxy, polypropylene, polyethylene, they would not be able to be completely biodegradable [9-11]. However, due to the properties of thermoset polymer which include brittleness, lengthy cure cycles and inability to repair or recycle damages, the development of the thermoplastic matrix composite system could exist. Thermoplastic polymer, such as low density polyethylene (LDPE) is a thermoplastic polyolefin that is inexpensive, recycleable, easier to use for application, and easy to process at low temperature as compared to other polymers. However, it requires heat in the mixing process.

Meanwhile, cellulose is one of the most abundant biopolymers in nature which is renewable, sustainable and biodegradable. Recently, modified cellulose has been used as reinforcements for various composites due to its excellent mechanical performance and biodegradable in a wide variety of environmental conditions. Chemical modification of cellulose such as cellulose acetate which is prepared by acetylation cellulose is an important route for the production of multifunctional materials. Polymer matrix composite filled with cellulose or cellulose acetate is potential in the development of industrial materials especially due to the biodegradable issue. Therefore, the study on biodegradation behavior is important for the application of composites in the environment.
Experimental Procedure

Low density polyethylene (LDPE) used in this study has crystallinity about 50% and a density of 0.93 g/cm³. Cellulose contains 1.15% lignin, 73.91% holocellulose and 49.75% α-cellulose. On the other hand, cellulose acetate, as another filler, has 1.761 of degree of substitution and 32.098 % of acetyl.

To carry out this experiment 10% (by total weight of composite) of fillers (cellulose and cellulose acetate) were added with low density polyethylene (LDPE) and then mixed using an extruder at processing temperature of 125°C. Composites were then compression molded using hot press at 125°C. In hot press, composites were preheat for 5 minutes and followed by 5 minutes compression time at the same temperature. The specimens were allowed to cool under pressure for another 5 minutes. Biodegradation studies were carried out for specimen of each composite by burying in the soil and by hanging in the open environment. The specimens (40 x 10 mm) were weighed and then buried 150 mm beneath the surface of soil and it was also done for specimens by hanging in an open environment. The temperature and moisture were in accordance with the real condition at the time of the biodegradation process. After every 15 days until 4 months the specimens of each composite were weighed in order to determine the weight loss to calculate the percentage of weight loss as follows:

\[
\text{percentage of weight loss} (\%) = \frac{w_i - w_t}{w_i} \times 100\%
\]

where \(w_i\) is the initial mass and \(w_t\) is the remaining mass at any given time, \(t\). All results are the average of three replicates. The specimens of each composite were also analyzed by SEM.

Results and Discussion

Biodegradation of LDPE composites filled with cellulose (C) and cellulose acetate (CA). Figure 1 shows the percentage of biodegradation of LDPE and LDPE composites filled with cellulose as well as cellulose acetate.

![Figure 1. Percentage of weight loss LDPE and LDPE composites filled with cellulose (C) and cellulose acetate (CA) during burying in soil and hanging in open environment for 4 months](image)

Results have shown the addition of fillers in LDPE composites have less impact on a material’s degradation. It can be seen that the percentages of weight loss is relatively very small which are below 0.2%. For pure LDPE almost nothing happened during biodegradation over time that has been set for either the specimen cultivated in the ground or hung. This is related to the hydrophobic backbones consisting of long carbon chains that gives high resistivity against hydrolysis [12]. Therefore, pure LDPE showed almost resistance to microorganism attack whether in the soil as well as in open environment.
On composite filled 10% cellulose (C), for the first 45 hours the specimen burying in the ground, the percentage weight loss had a negative value means that there is weight gain a specimen on the first 45 hours. This may be caused by the absorption of water by the specimen in the soil so as to add weight composites. Along with time, the composites then losing their weight and is getting bigger. This happens because the cellulose has been decomposed by microorganisms in the soil. Cellulose is biodegraded by microorganism that utilize cellulase enzymes [13]. As the microorganism attacks, the composites lose their integrated structure.

On the other hand, the composite filled 10% cellulose acetate (CA) showed that the percentage of weight loss due to biodegradation is smaller compared to the composite filled 10% C. This is due to the acetyl groups in cellulose acetate requires the presence of esterases on the first phase of biodegradation. Previous studies identified that the first step mechanism of degradation in the process is deacetylation by chemical hydrolysis and acetylesterases [14].

Meanwhile, the percentage of weight loss composite by hanging on open environment was higher compared by burying in soil. The main step of degradation in open environment is the termination on the main chain to form fragments with low molecular weight (oligomers) that can be assisted by microbes. Decrease in molecular weight has been caused by the chain termination of hydrolysis and oxidation. Hydrolysis occurs in the environment with the presence of enzymes or non-enzymatic conditions. Some fungi can secrete enzymes that catalyze oxidation reactions of either cellulose itself or the lower molecular weight oligomers produced from the enzymatic hydrolysis of cellulose. In this case heat can also cause hydrolysis. Whereas an oxidative termination occurs due to the presence of oxygen and UV light [13].

**SEM Analysis.** Figure 2 shows the SEM of LDPE and composites LDPE filled with cellulose (C) and Cellulose acetate (CA) after buried in soil and hanged in open environment.

![SEM images](image_url)

a. Pure LDPE (buried in soil)  
b. Composite 10% C(buried in soil)  
c. Composite 10% CA (buried in soil)  
d. Pure LDPE (hanged in open environment)  
e. Composite 10% C (hanged in open environment)  
f. Composite 10% CA (hanged in open environment)

**Figure 2.** SEM of LDPE and LDPE composites filled with cellulose (C) and cellulose acetate (CA) after buried in soil and hanged in open environment (mag. 100x)

It can be seen that pure LDPE have almost had no change whether it was buried in soil or hanged in an open environment (Figs. 2.a and 2.d). Whereas on composite filled 10% C after buried in the soil (Fig. 2.b) contained cavities left by the cellulose which undergoes biodegradation. However, composite filled 10% C after hanging in open environment (Fig. 2.e) had more cavities indicates
more biodegradation of cellulose. Whereas, the 10% CA in composite (Fig. 2c) is still clearly visible even after 4 months of burying, while for composite after hanged in an open environment (Fig. 2f) there is a cavity due to the degradation of cellulose acetate.

**Conclusion**

It can be concluded that on the biodegradation test, the percentage of weight loss of LDPE composite filled with 10% cellulose showed higher as compared to the composite filled with 10% cellulose acetate. Also, the degradation process by hanging on the exposed environment was faster than burying in soil.

**References**


