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 °C. Biodegradation processes were done by burying in the soil and by hanging in open environment for four months. The percentage of weight loss of pure LDPE and composites due to the degradation were 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 were constant. However, the composites filled with cellulose were relatively more susceptible to degradation as compared with composites filled cellulose acetate. Here, the percentage of weight loss of composites filled cellulose were higher than the composites filled 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 were conformed by SEM which have showed the 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, improve the mechanical properties, increase their range of applications, and biodegradable. If it deals with the compounding of natural fibers with biodegradable polymers including starch, cellulosic plastics, poly(hydroxyl alkanoate) (PHA), poly(lactic acid) (PLA), and polycaprolactone are called fully or completely biodegradable [6-8]. The other way is by mixing natural fiber with thermosetting or thermostatic polymers such as epoxy, polypropylene, polyethylene [9-11]. These composites cannot be taken a completely biodegradable materials due to the fact that the matrix is a non biodegradable polymer. However, due to the properties of thermoset include brittleness, lengthy cure cycles and inability to repair or recycle, damages, therefore, the development of the thermoslastic matrix composite system are exist. Thermoslastic polymer, such as low density polyethylene (LDPE) is a thermoplastic polyolefin that is, inexpensive, recycleable, easy processing, low temperature processing as compared to other polymers as well as more applicable in their use, however, in the mixing process requires heat.

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 acetylated 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 behaviour is important for the application of composites in environment.
Experimental Procedure

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

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 open environment. After every 15 days until 4 months the specimen of each composite were weighed in order to determine the weight loss to calculate the percentage of weight loss as follows:

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\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. Specimen of each composites were also analyzed by SEM.

Results and Discussion

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

![Graph showing biodegradation of LDPE and LDPE composites](image)

Fig. 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

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), 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 caused by the chain termination of hydrolysis and oxidative. 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 caused hydrolysis. Whereas an oxidative terminations occurs because of the presence of oxygen and UV light [13].

SEM Analysis. Fig. 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.

Fig. 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 are almost have no change whether it was buried in soil or hanged in open environment (Fig. 2a and 2d). Whereas on composite filled 10% C after buried in soil (Fig. 2b) contained cavities left by the cellulose undergoes biodegradation. However, composite filled...
10% C after hanged in open environment (Fig. 2e) had more cavities indicate more biodegradation 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 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 exposed environment were faster than burying in soil.

References