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Characterization and Degradation Analysis of Tapioca Starch Based Biofilms

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***Abstract*—**In this study, tapioca starch, which acts as natural polymer, was added in the blend in order to produce biodegradable product. Low density polyethylene (LDPE) and tapioca starch blends were prepared by extrusion and the test sample by injection moulding process. Ethylene vinyl acetate (EVA) acts as compatibilizer while glycerol as processing aid was added in the blend. The blends were characterized by using melt flow index (MFI), fourier transform infrared (FTIR) and the effects of water absorption to the sample. As the starch content increased, MFI of the blend was decreased. Tensile testing were conducted shows the tensile strength and elongation at break decreased while the modulus increased as the starch increased. For the biodegradation, soil burial test was conducted and the loss in weight was studied as the starch content increased. Morphology studies were conducted in order to show the distribution between LDPE and starch.

***Keywords*—**Biofilms, degradable polymers, starch based polyethylene, injection moulding.

I. Introduction

PLASTICS have been widely used all over the world but it has become a major problem when the plastic cannot degrade even after hundred years. The usage of excessive plastics in food packaging, plastics bags and plastics gardening film makes the landfill becomes worse once the plastics has become wastes. In Malaysia, plastic waste is the most common solid waste in the country accounting for 7-12 percent by weight and 18-30 percent by volume of the total residential waste generated. The characteristics of plastics that are strong, light-weight, inexpensive, easily processable and energy efficient make degradable being a difficult task [1]. The most common plastic that was used in many applications is polyethylene (PE) due to its good mechanical properties and low cost [2].

There are developments of biodegradable plastics that can reduce the municipal wastes and this development indirectly will prevent the world become waste disposal. Biodegradable polymers and plastics are material that was completely decomposed when exposed and under attack to microorganism, either bacteria or fungi with appropriate environmental condition. By blending polymers and starch the tendency for the product to degrade is become larger. The blending of biodegradable polymer which is starch with inert polymers such as polyethylene has received considerable attention because of the possible application in the waste disposal of plastics. The blends between starch and polyethylene also lead to poor mechanical properties and in order to improve the incompatibility between starch and polyethylene this is by addition a compatibilizers [3].

In this study, biodegradable LDPE and tapioca starch blends were formulate via injection moulding processes with addition of EVA as compatibilizers and glycerol as processing aid. The mechanical, thermal and biodegradability of LDPE/TS blends were discussed besides to optimize the injection moulding process parameters of the blends.

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II. Experimental

A. Materials and Sample Preparation

Low density polyethylene (LDPE) resin grade (Titanlene® low density Polyethylene LDI 300YY) supplied by Titan Polyethylene (M) Sdn. Bhd were used in this study. The density range is 0.91 – 0.925 g/cc. Tapioca starch food grade that were used in the blending process are as a natural polymer. Ethylene vinyl acetate, EVA (Cosmothene® EVA, Singapore) with vinyl acetate content of 18 wt % was used in this research as compatibilizers and glycerol (glycerin, $C_3H_8O_3$, molecular weight, MW = 92.10 g/mol) was used.

Tapioca starch was dried in the oven to remove the moisture content at 82°C for 24 hours. The mixing process was being carried out with LDPE and EVA added continuously by using high speed mixer. The glycerol as a processing aid added continuously during the mixing process and tapioca starch was added to the mixture as the process continuous mixed. Five formulations were prepared which consists of 30% until 60% of starch. The mixture was compounded by using Berstorff twin screw extruder ZE 25 at speed of 80 rpm and the temperature set at 120/125/130/140/135/125/120°C. The compounded pellet was dried in the oven at 60°C for 24 hours. The sample was put into the injection moulding machine to obtain test specimens for the measurement of tensile properties.

B. Melt Flow Index (MFI)

MFI of the samples was determined by using Zwick Werkstoff-Prufmashinen melt flow indexer according to the ASTM D1238. LDPE/TS blends will be loaded with load of 2.16 kg onto the piston and the temperature is 190°C was used in this measurement.

C. Water Absorption

The samples were cut in the rectangular form with 20mm length, 20mm width and 40mm thickness and dried at 70°C for 24 hours in the oven. The sample was cooled in the desiccator and weighed immediately to obtain initial weight (W1). The sample was placed in a container and immersed with distilled water. At regular time interval, each sample was removed and dried from container and subsequently weigh (W2) to determine its water absorption.

$$\% \text{ Water absorption} = [(W2-W1)/W1] \times 100$$

D. Soil Burial Test

Biodegradation of the samples was studied by the soil burial method. The samples with 20mm x 20mm x 40mm dimensions were buried in the open spaces. The samples were removed periodically after 1, 2, 3 and 4 weeks and washed thoroughly with water and dried at 70°C for 24 hours in the oven. The weight of the samples was recorded to calculate the extent of biodegradation of the samples.

III. Results and Discussions

A. Melt Flow Index (MFI)

The melt flow index (MFI) value of LDPE blends with TS is decreased as the starch loading is increased as shown in Table I. This is due to the increasing of viscosity of the blend which makes the polymer hard to flow, higher molecular weight and higher elasticity. The interaction among the granules was increased as the content of starch is increased thus lead to higher viscosity. The spaces between the particles were small as for higher loading content. If the particle-particle interactions are stronger than particle-matrix interaction, agglomeration of particle may occur and give immobilization of more matrix molecules [4].

B. Water Absorption Test

The melt flow index (MFI) value of LDPE blends with TS is decreased as the starch loading is increased as shown in Fig. 1. This is due to the increasing of viscosity of the blend which makes the polymer hard to flow, higher molecular weight and higher elasticity. The interaction among the granules was increased as the content of starch is increased thus lead to higher viscosity. The spaces between the particles were small as for higher loading content. If the particle-particle interactions are stronger than particle-matrix interaction, agglomeration of particle may occur and give immobilization of more matrix molecules [5].

TABLE I
Composition of the LDPE/TS Blends with MFI Value

Composition	MFI value (g/10 min)
LDPE: 100	5.18
LDPE/TS : 70/30	3.77
LDPE/TS : 60/40	2.50
LDPE/TS : 50/50	0.62
LDPE/TS : 40/60	0.084

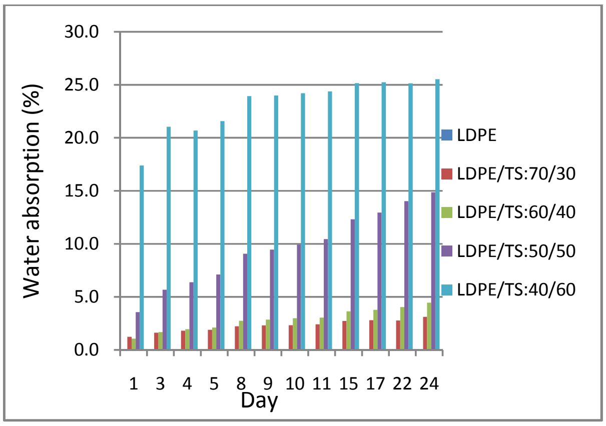

Fig. 1 Variation in water absorption of the LDPE and LDPE/TS with time of immersion

C. Mechanical Testing

Addition of starch into the LDPE reduced the tensile strength of the blends at all compositions. Young's modulus increase whereas the extension at break decreased as the amount of starch increased. Fig. 2 shows, the graph of tensile strength against tapioca starch loadings. While Fig. 3 shows the plot of the Young's modulus versus the percent of tapioca starch loadings. This shows that the LDPE/TS blends have higher modulus than the pure LDPE though, as the tapioca starch increased in the blends, the Young's modulus will also increase. This is due to the stiffening effect of the starch particles in the blends. It is because the modulus of starch is higher than semi-crystalline LDPE because of the presence of hydrogen bonding in starch and not in LDPE [6]. The elongation at break versus percentage of tapioca starch was plotted as shown in Fig. 4. The graph shows that the elongation at break decreased as the percentage in the starch increased. Elongation at break mainly depends on the interfacial adhesion between the polymers. Since the LDPE and starch are incompatible and their interfacial adhesion is very poor, so EVA as compatibilizer is expected to reduce the interfacial tension and increase the adhesion [7]. Thus, the elongation at break is decreased because the amount of EVA added in the blend is not compatible as the increasing of starch.

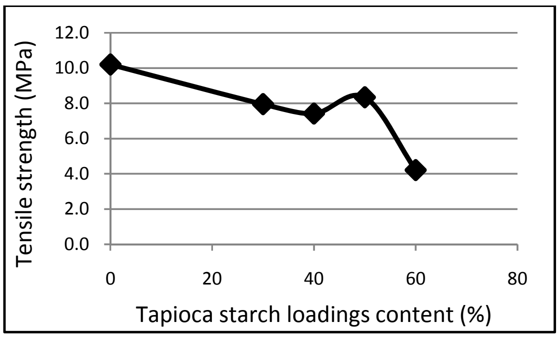

Fig. 2 Tensile strength of LDPE and of tapioca starch

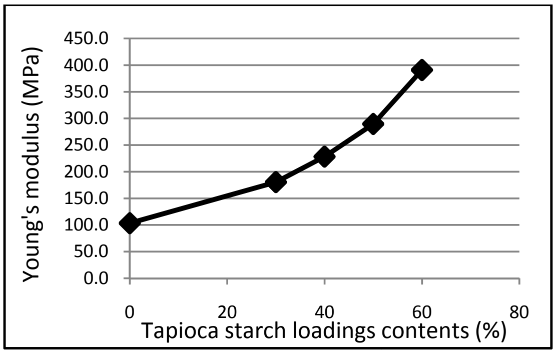

Fig. 3 Young's modulus of LDPE and LDPE/TS with varies percentage content of tapioca starch

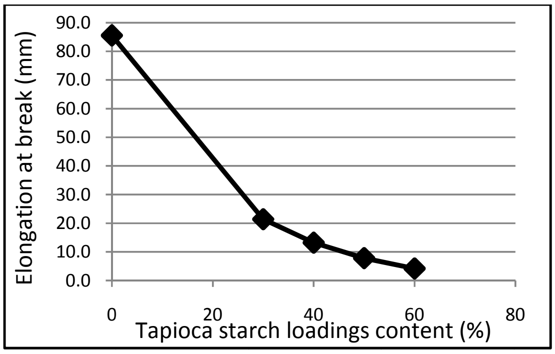

Fig. 4 Elongation at break of LDPE and LDPE/TS with varies percentage of tapioca starch

D. Soil Burial Test

Soil burial test is a common method for estimating the biodegradation rate by determined the weight loss of the samples after being buried at certain time. There are some changes in weight loss that can be clearly seen in Fig. 5. While Figs. 6-10 show the sample before and after being buried in the soil. The weight loss increased as the increasing of starch content in the blends. This can be observed from the formulation of LDPE/tapioca starch blends containing 60% of starch which shows the highest changes in weight loss.

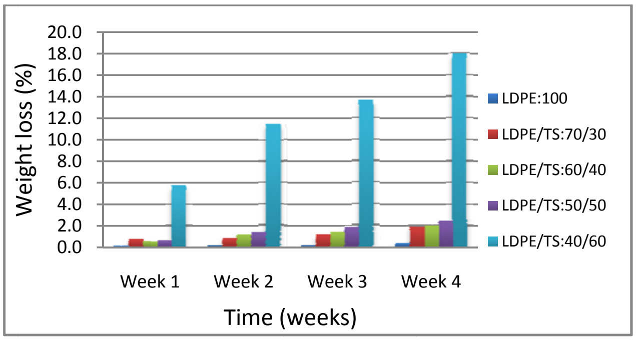

Fig. 5 Variation in weight loss of the LDPE and LDPE/TS with time of burial

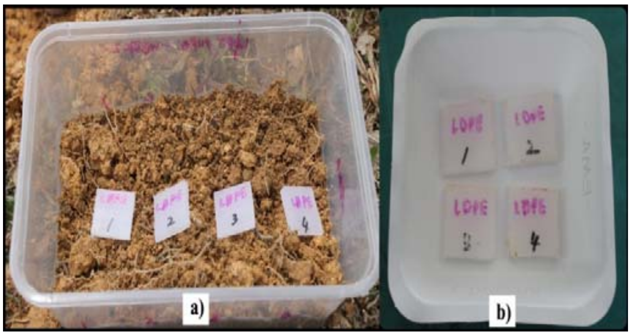

Fig. 6 Sample of pure LDPE a) before and b) after the soil burial test

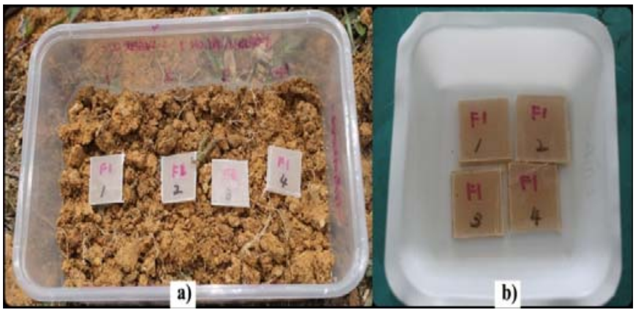

Fig. 7 Sample of LDPE/TS: 70/30 a) before and b) after the soil burial test

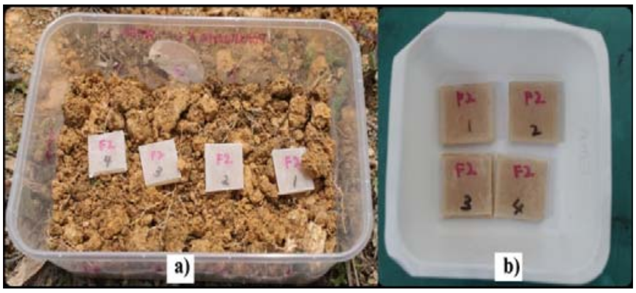

Fig. 8 Sample of LDPE/TS: 60/40 a) before and b) after the soil burial test

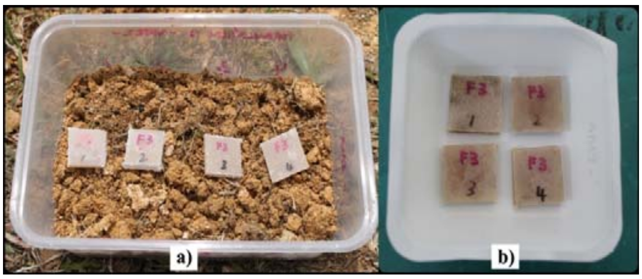

Fig. 9 Sample of LDPE/TS: 50/50 a) before and b) after the soil burial test

Fig. 10 Sample of LDPE/TS: 40/60 a) before and b) after the soil burial test

IV. Conclusion

The blends of LDPE and tapioca starch containing 30, 40, 50 and 60% of starch were prepared. The addition of starch to LDPE reduced the MFI values, tensile strength and elongation at break, whereas the modulus increased. For morphology studies, the micrographs shows agglomerate between the blends as the content of starch become higher. As for biodegradable testing, LDPE/TS shows signed of degradation after undergo soil burial test for 4 weeks. Formulation containing 60% starch shows the most changes in weight and the surface of the sample have darker spots which shows that the microorganism starting to react with the sample and makes the degradation process have higher possibilities.

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