Research Article

Development of bio-composite film based on high density polyethylene and oil palm mesocarp fibre

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Abstract
The aim of this study is to develop bio-film based on high-density polyethylene (HDPE) and oil palm mesocarp fibre (OPMF). Rheological and mechanical properties of the biofilm were investigated using melt flow index and tensile test respectively. The produced films were characterized using scanning electron microscopy and differential scanning calorimetry (DSC). In addition, biodegradation and absorption properties were studied. OPMF is dried and then added to HDPE to be compounded using a twin-screw extruder. Maleic anhydride was used as compatibilizer during the melt mixing. The film samples were prepared using hot compression moulding process. Young's modulus was found to increase with addition of OPMF and vice versa for tensile strength and elongation at break of the film sample. The reason behind the reduction of tensile strength and elongation at break is the low effective cross sectional area of HDPE matrix. Melting point value was increased after the addition of OPMF into the film as shown in DSC analysis. The rising of water absorption rate of the films is greatly relative to the content of OPMF as a result of high affinity of OPMF to water.

Keywords High-density polyethylene (HDPE) · Mesocarp fibre · Biofilm · Tensile test · Water absorption · Biodegradation

1 Introduction

Worldwide production of plastic is more than 78 million tons per year and almost half of that is discarded within short time, remaining in garbage deposits and landfills for decades [1, 2]. Petrochemical base plastics such as polyolefin, polyester, and polyamides have been increasingly used as packaging materials due to their desired properties such as good tensile and tear strength, good barrier to oxygen and aroma compounds and heat seal ability [3]. However, these plastics are petroleum-based materials that are not easy to be degraded naturally in the environment.

Biodegradable plastics are defined as plastics with similar properties to conventional plastics, but they decomposed after disposal to the environment by activity of microorganisms to produce end products of CO_2 and H_2O [3]. Many synthetic materials like polyolefin are not degraded by microorganisms in the environment, which contributes to their long-life of hundred years [4]. Polyethylene (PE) is one of the mass product non-degradable polymers and various types of PE such as high-density polyethylene are used extensively in many fields. The HDPE is an engineering thermoplastic used for several industrial applications due to low cost, desired mechanical properties and processing facility. Production and processing of synthetic polymer with natural polymer is interesting alternatives to reduce the cost of biodegradable polymers in market and enhance the biodegradation of petroleum-based polymers [5, 6]. Natural fibres have recently a widely attention of researchers due to advantages over traditional reinforcement materials such as glass fibre in terms of cost, density and biodegradability [5, 7]. Natural fibres have some disadvantage such as poor compatibility with

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hydrophilic matrix, tendency to form aggregates during processing and low resistance to moisture, which led to potential reduction to be used as polymer reinforcement [8]. On the other hand, various treatments are being used to improve fibres/matrix compatibility [4, 9, 10]. Natural fibre treatments have been studied in order to promote the chemical modification of fibres surface; in this case hydroxyl groups characteristically very reactive and susceptible to chemical reactions are substituted in direction to obtain a polar surface. Non-polar groups inserted on fibres surface, provides a hydrophobic surface characteristic increasing the compatibility of thermoplastic matrixes [8]. Blending HDPE with a cheap natural biopolymer such as mesocarp fibre is necessary method to enhance biodegradation and reduce the cost of final product. OPMF is a good choice since it is an abundant and low cost material; therefore it used in this study to reduce the production cost and enhance the biodegradation properties of HDPE film [11, 12].

2 Experimental

2.1 Materials

High-density polyethylene (HDPE) resin was used as a matrix and supplied by Titan Polyethylene (Malaysia) Bhd. The density HDPE was 0.96 g/cm^3 and melting temperature (T_m) range from 130 to 137 °C. Oil palm mesocarp fibre (OPMF) was supplied by oil palm mill at Labu, Negeri Sembilan, Malaysia. Maleic anhydride (MA) was supplied by Shenzhen Jindaquan Technology Co. and used as compatibilizer. Glycerol is provided by Hangsun Plastic Additives Co., Ltd. And used as plasticizer.

2.2 Preparation of bio-composite film

OPMF was ground using grinder to average particle size of 200 µm. High density polyethylene (HDPE) and OPMF were dried in an oven for 24 h at temperature of 70 °C. Then HDPE was mixed with OPMF and MA for 15 min at room temperature using high-speed mixer with 500 rpm. Twin-screw extruder was used for HDPE/OPMF compounding. The compounding process was carried out at a speed 50 rpm and temperature of 180 °C/185 °C/190 °C. Extradite was palletized using palletizer machine for each sample formulation as specified in Table 1. The composites were compression moulded into film with average thickness of 0.14 mm. A mould containing the required material were placed in the press machine and preheated for 5 min without applying any pressure, in order to ensure uniform heat flow through the material. The composites were pressed at 165 °C for 5 min and the resulting film was being cooled to the room temperature for 15 min using cold press machine. Then, the film was cut into required shape. In this study, 5 phr of glycerol was used as a plasticizer for each sample formulation.

Table 1 Sample formulation

Samples	Wt%		
	HDPE	OPMF	Maleic anhydride
S0	100	0	0
S1	90	5	5
S2	85	10	5
S3	80	15	5
S4	75	20	5

2.3 Sample testing

2.3.1 Melt flow index

The melt flow index test was used to analyse the melt flow behaviour. The test was conducted according to ASTM D1238, using an extrusion plastometer model of S. A. Associates. The mass of extruded material was determined and the results were expresses as g/10 min.

2.3.2 Tensile test

The tensile strength, elastic modulus, and elongation at break of samples were determined using Intron machine Lloyd. The test was conducted according to ASTM D882. The gauge length was set at average of 100 mm and carried out at a crosshead speed of 50 mm/min. Five specimens were tested for each formulation and the average value was reported.

2.3.3 Water absorption

Water absorption test was used to determine the amount of water absorbed under specified conditions. This test was carried out to study water resistance of HDPE/OPMF films. Samples were dried at 80 °C in a vacuum oven until a constant weight was reached prior to immersion in distilled water at 30 °C. The weight gain of the sample was reported after remove from distilled water at any time. The percentage of weight gain, (M_t) was determined using Eq. 1:

$$M1 = \frac{Ms - Mo}{Mo} \times 100 \quad (1)$$

where M1% is water absorption, Ms and Mo are weight of the samples after and before immersion in water respectively.

Table 2 Melt flow index for each sample

Sample	Melt flow index (g/10 min)
S0	8.6
S1	2.4
S2	1.3
S3	1.1
S4	0.6

2.3.4 Scanning electron microscopy (SEM)

SEM is used to study the interactions morphology between OPMF and HDPE. Hitachi Scanning Electron Microscope FlexSEM 1000 was used to study the surface topography of HDPE/OPMF films at magnifications of 500×.

2.3.5 Differential scanning calorimetry (DSC)

DSC was used to study the thermal properties of the compounded pellets of HDPE/OPMF. Perkin Elmer Instrument was used at range of temperature from 25 to 220 °C and scanning rate of 10 °C/min.

2.3.6 Biodegradability studies

Biodegradation of the films were determined according to ASTM G21 and used to evaluate the resistance of polymeric materials to fungi. Samples were tested in petri dish containing sterile nutrients salts agar and piece of each sample. Petri dish cover was sealed by wax to avoid any kind of contamination. The fungal pieces were tested with *Aspergillus niger* ATCC 9642. The samples were incubated at 27–37 °C for 9 days, and then the films were examined for evidence of colony growth and reweighed to determine the weight loss.

3 Result and discussion

3.1 Melt flow index

The results in Table 2 showed the MFI value for each sample. The results showed that MFI values decreased with increasing of OPMF content. The index value of S0 dropped drastically from 8.6 to 2.4 g/10 min with the addition of 5% OPMF. In the presence of 5% OPMF filler, the OPMF particles had restricted the mobility of HDPE chains. This is due to the grafting of OPMF particle onto HDPE chain through a reaction of maleic anhydride (MAH) with OPMF and HDPE. As consequence, the viscosity increased which led to less molten polymer could flow through the die. However, further addition of OPMF had imparted an exponential decreasing behaviour of index values. The viscosities of sample gradually increased and the molten samples are hard to flow.

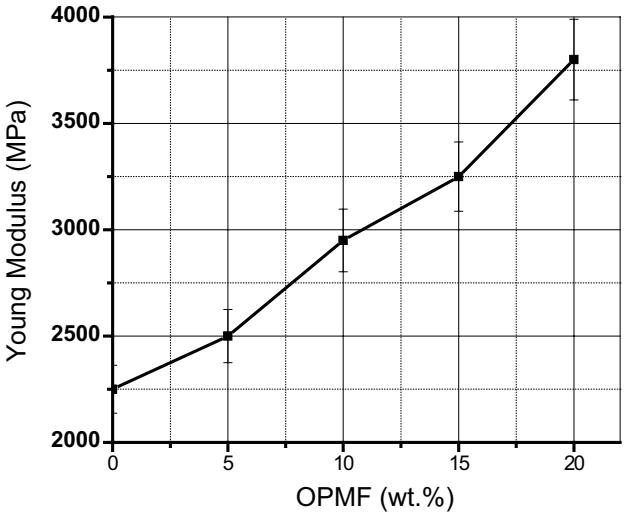

Fig. 1 The Young's modulus at different OPMF loading

3.2 Tensile test

3.2.1 Young's modulus

The results in Fig. 1 showed the Young's Modulus of the samples film at different OPMF content. Sample S0 has the lowest modulus of 2238 MPa. The modulus gradually increased with the increasing of fiber loading and S4 showed the highest value of 3796 MPa. Samples S1 and S2 showed 11% and 24% increment, respectively, meanwhile, sample S3 showed 30% incremnt. These results implied that the samples became more stiffer and had higher resistance to deformation. This was thought due to the reinforcing effect of OPMF fillers into HDPE matrix [12, 13].

3.2.2 Tensile strength

Figure 2 shows the results of tensile strength of the samples. The result indicated that the tensile strength of samples decreased with the increasing of OPMF contents. Samples S1 and S2 decreased by 26% and 42% respectively, meanwhile, sample S3 decreased by 47%, while S4 showed the lowest tensile strength at 14.3 MPa.

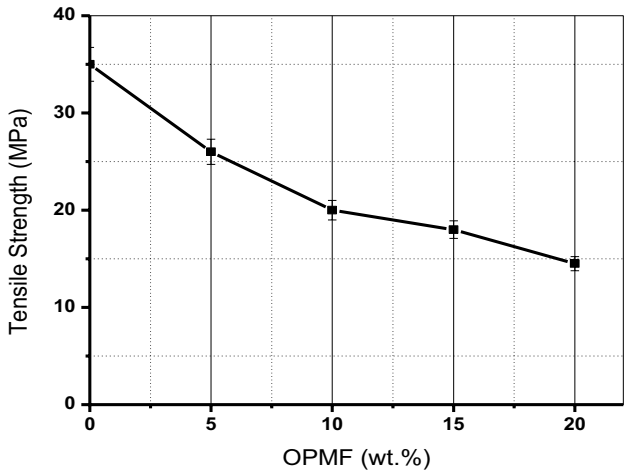

Fig. 2 Tensile strength of biofilm samples

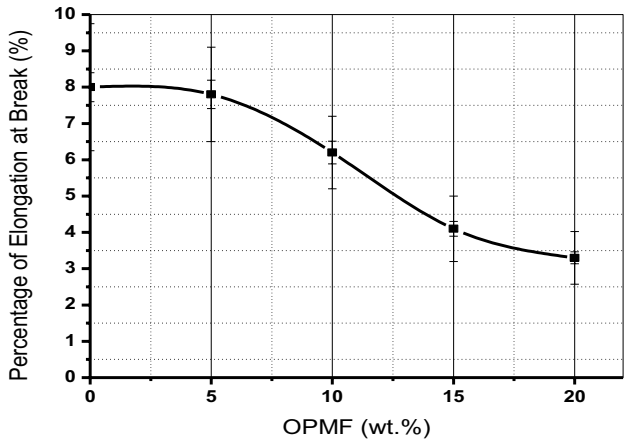

Fig. 3 Percentage of elongation at break of biofilm samples

The decreasing of tensile strength occured due to interfacial adhesion weakness between fiber-polymer [14, 15]. Although grafting agent such as MAH was introduced in the system at fixed amount of percentage of weight but the amount might be not enough to accommodate the increasing of OPMF content, thus producing weak connection with HDPE matrix. Besides that, the presence of bubbles in the sample may also cause the reduction of tensile strength [12].

3.2.3 Elongation at break

The plot of elongation at break versus OPMF loading is illustrated in Fig. 3. As expected, percentage of elongation at break decreased when OPMF contents increased. The elongation at break of S0 was about 8.1%, while S1 was 7.8%. The values kept decreasing, and S4 has lowest elongation at break of 3.6%. This reduction is due to intervention of OPMF particles on the continuity of HDPE matrix, which resulting in discrete phase and affecting the flexibility of the samples [12, 16].

Table 3 The percentage of water absorption for biofilm samples

Sample	Percentage of water absorption (%)
S0	0.21
S1	0.82
S2	1.21
S3	1.32
S4	1.52

3.3 Water absorption

The test was conducted to determine water sensitivity of HDPE/OPMF films. The percentage of weight gain (M1) is indicated in Table 3. It can be seen that, the percentage of water absorption of the sample increased with the increasing of OPMF content in each sample due to the tendency of OPMF to absorb water as a result of the existence of hydroxyl groups. Hydroxyl groups can form hydrogen bonding with water, which enhance the affinity toward water, and allowing water molecules to be absorbed [17, 18].

3.4 Differential scanning calorimetry (DSC)

Figure 4 shows the DSC curve of heat flux versus temperature for the samples at different loading of OPMF. The values of melting temperatures (T_m) of each sample were recorded in Table 4. The result in Table 4 reveals an onset T_m for each sample with no significant change with the increasing of OPMF contents. HDPE is a dominant phase in this blend and the content of OPMF was too small to influence the melting temperature of the matrix of HDPE [11, 12].

3.5 Scanning electron microscopy (SEM)

Figure 5 shows the micrographs of all samples at 500× magnifications. Sample S0 exhibited smooth and

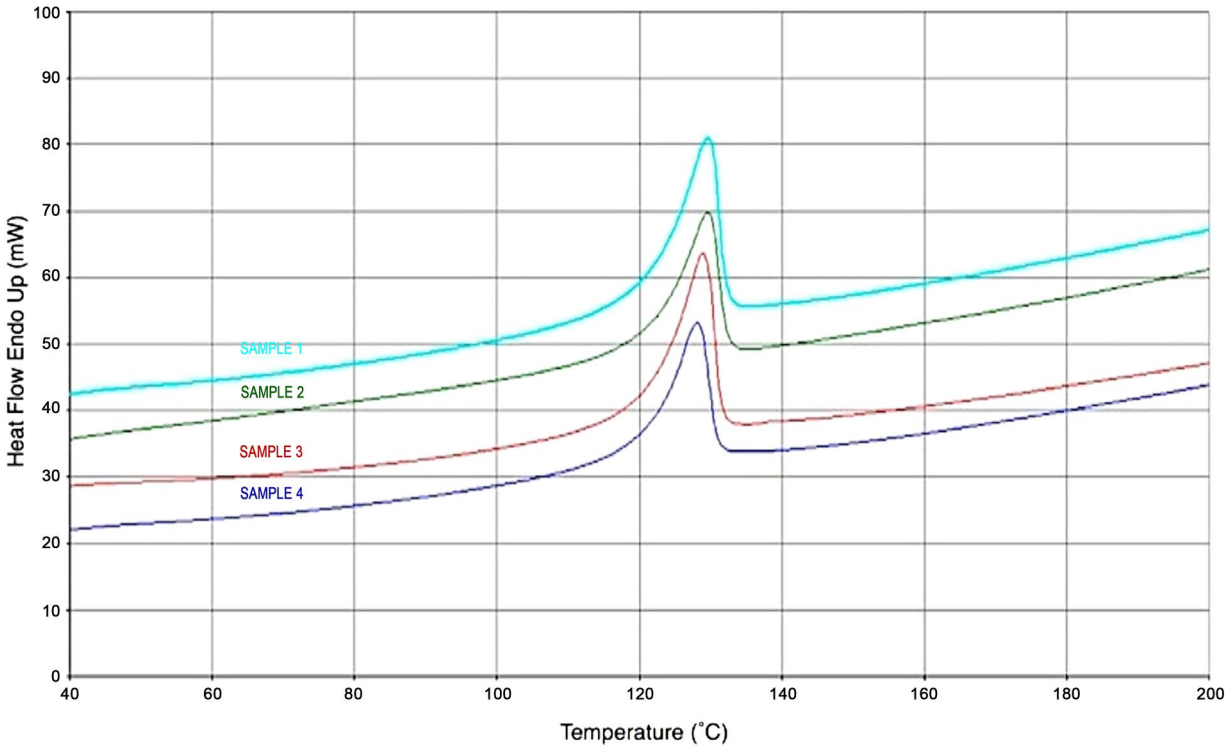

Fig. 4 DSC thermogram of S1, S2, S3 and S4 at different loading of OPMF

Table 4 Melting point (T_m) of the biofilm samples

Sample	T_m (°C)
S0	131.00
S1	129.67
S2	129.67
S3	129.00
S4	128.17

continuous HDPE matrix. However, with the addition of OPMF, the texture became rougher and the continuity of HDPE matrix was disrupted. Small holes or voids were scattered throughout the sample as observed in image of S1. The voids became larger and OPMF protruded from the crack in images of S2, and S3. This effect was very obvious in Fig. 5e due to isolated discontinuous phase of HDPE matrix which led to weakening the sample strength and lowering the flexibility [19]. These observations are in agreement with the findings from tensile test in Sect. 3.2.

3.6 Biodegradability test

Table 5 shows the weight loss of the sample over 12 days. There was drastically weight drop due to increasing of OPMF content. Therefore, HDPE/OPMF undergoes biodegradation at a faster rate compared to HDPE film without OPMF loading. The fibre seems to initiate and promote biodegradation of HDPE/OPMF, which effortlessly attacked by *Aspergillus niger* [20, 21].

4 Conclusion

As a conclusion of this research, OPMF has strong effect on physical and mechanical properties of HDPE film. Biodegradation characteristic of HDPE films is enhanced by the addition of OPMF. OPMF is environmentally friendly and fully degradable by microorganism, however, OPMF has a contrary effect on the mechanical properties, which decreasing the tensile strength while increasing the

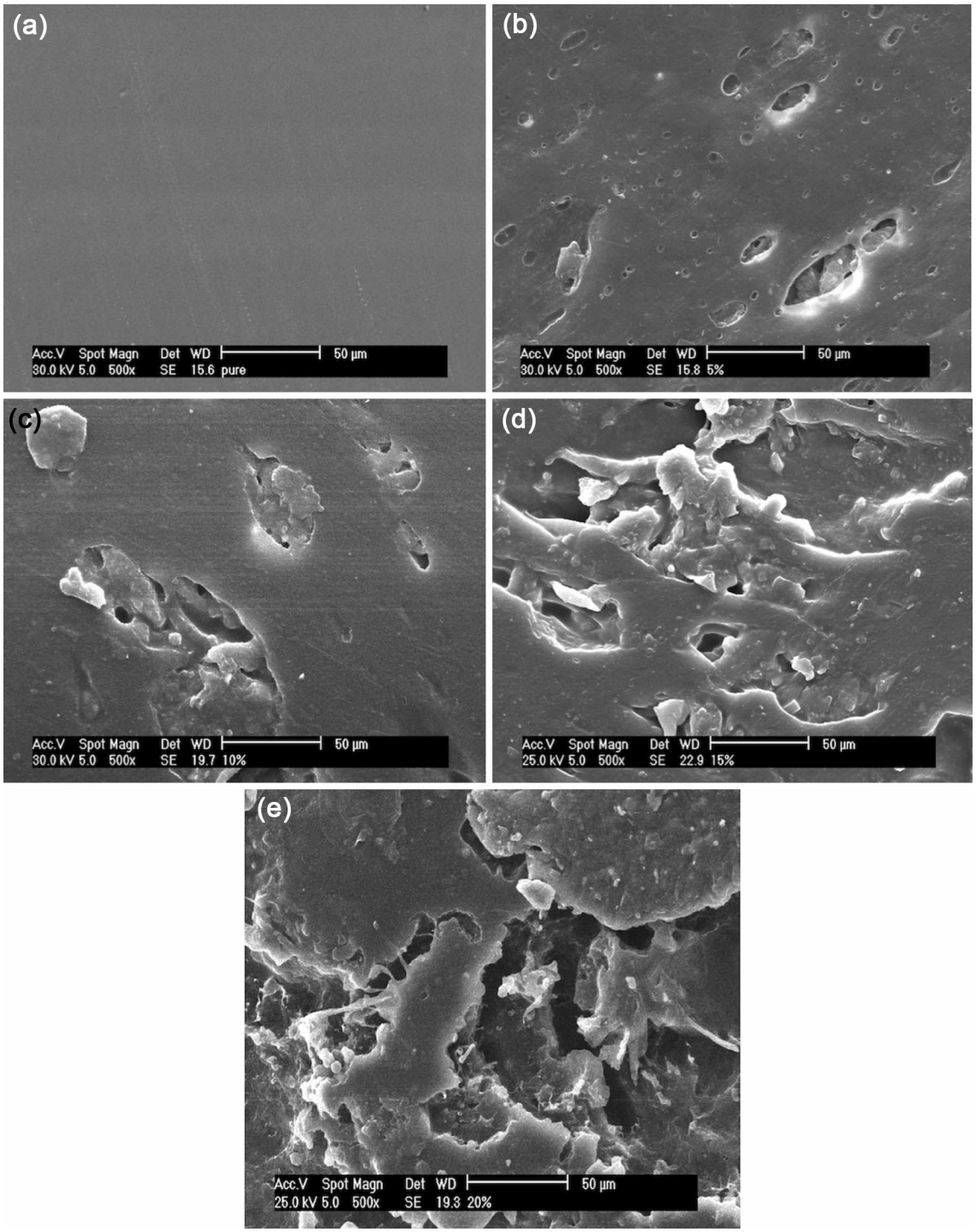

Fig. 5 Micrographs of the samples at magnifications of × 500, **a** S0, **b** S1, **c** S2, **d** S3, **e** S4

Young's modulus of HDPE films. Water absorption was increase due to the existence of hydroxyl group (OH) of OPMF, which enhance the affinity of HDPE toward water and forming hydrogen bonding with water molecules. Additional characterizations of the produced biofilm are required and will be investigated in another article.

Table 5 Percentage of weight loss of biofilm samples

Sample	Percentage of weight loss (%)
S0	0
S1	70.16
S2	80.90
S3	90.31
S4	95.86

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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