

The Effect of Epoxidized Natural Rubber (ENR 50) as a Compatibilizer on Spear Grass Filled Low Density Polyethylene/Soya Spent Powder

(Kesan Getah Asli Terepoksida (ENR 50) sebagai Penyerasi Terhadap Lalang Diisi dengan Polietilena Berketumpatan Rendah/Serbuk Soya)

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ABSTRACT

Composite based on spear grass with low density polyethylene (LDPE)/soya spent powder (SSP) were prepared by using twin screw extruder at 150°C. The spear grass (SG) loading was ranged from 0 to 15%. A compatibilizer, epoxidized natural rubber (ENR 50) with 50 mol % epoxidation was added. The effect of filler content and compatibilizer on the properties of LDPE/SSP composite was studied. The results indicated that the increase of SG loading leads to the reduction of tensile strength and elongation at break (E_b), whereas the Young's modulus has increased. Differential scanning calorimeter (DSC), indicated that the melting temperature and crystallinity of the composites decreased with the increase of SG loading. After the addition of ENR 50, the melting temperature increases from 98.33 to 98.63°C for 95% LDPE, 5% SSP and 5% SG, whereas the crystallinity of the same ratio decreased from 31.92 to 28.13%.

Keywords: Differential scanning calorimeter; low density polyethylene; soya spent powder; spear grass; tensile properties

ABSTRAK

Komposit yang berasaskan lalang dan polietilena berketumpatan rendah (LDPE)/serbuk soya terpakai (SSP) telah disediakan menggunakan skru penyempitan berkembar pada suhu 150°C. Pemuatan rumput serampang (SG) ialah pada kadar 0 dan 15%. Penyerasi iaitu getah asli terepoksida (ENR 50) telah ditambah dengan 50 mol % pengepoksidaan. Kesan kandungan pengisi dan penyerasi terhadap sifat LDPE/SSP telah dikaji. Hasil kajian menunjukkan pertambahan pemuatan SG telah meningkatkan kekuatan tegangan dan pemanjangan takat putus (E_b) manakala nilai modulus telah menunjukkan pengurangan. Keputusan kolorimeter pengimbas pembezaan (DSC) telah menunjukkan suhu lebur dan suhu penghabluran komposit telah berkurang dengan pertambahan SG. Selepas pertambahan ENR 50, suhu lebur telah meningkat daripada 98.33 kepada 98.63°C untuk 95% LDPE, 5% SSP dan 5% SG. Manakala suhu penghabluran bagi komposit yang sama telah berkurang daripada 31.92 kepada 28.13%.

Kata kunci: Kalorimeter pengimbas pembezaan; polietilena berketumpatan rendah; rumput serampang; serbuk soya terpakai; sifat keregan

INTRODUCTION

As we concern of waste disposal for environmental and pollution issues, new biodegradable polymers offer a good substitute for non biodegradable materials based plastics. An approach is made to produce biodegradable plastics from agricultural product. This includes starch, sugar molasses, protein and cellulose (Swain et al. 2004). For the production of biodegradable plastics, a consideration was given to some of the protein plant. These proteins are corn protein, peanut protein and wheat gluten. Soya beans composed of proteins level 40-55%. The high amount of protein shows that the soya beans must be properly plasticized when being formed into plastics materials and films (Berkesch 2005).

The degradability of low density polyethylene (LDPE) can be fasten by adding the starch and carbohydrate into synthetic polymer. At high carbohydrate content, the incompatibility of LDPE and carbohydrate affect the

mechanical properties due to different polar character (Zuchowska et al. 2000). The mechanical properties of the blends can be improved by using compatibilizers. Ramos and Costa (2008) found that the compatibilized blends have slightly higher biodegradation rates compared with uncompatibilized blends. Its relatively low density arises from the presence of a small amount branching in the chain (2% of the carbons atom) that gives more open structure. The study was carried out by incorporating SG fiber into LDPE/SSP composites. The effect filler loading was investigated by using tensile and DSC test.

MATERIALS AND METHODS

MATERIALS

The LDPE matrix used in this study was provided by Titan Petchem (M) Sdn Bhd in solid pellets form with the density

of 0.922 g/cm³. The SSP in this study was provided by Euro Chemo Sdn Bhd which consists of 2% cellulose, 17% pectin, 14% protein and 53% insoluble polysaccharide. SG was harvested from Bintong, Perlis, Malaysia.

RAW MATERIAL PREPERATION

The SG was dried at 100°C in the oven for 3 h. Then, it was grinded by using a grinder. After that, it was dried again for 3 h at 100°C to remove the moisture content.

MIXING

Soya spent powder (SSP) and LDPE were mixed in twin screw extruder. The temperature was set at 150°C and the speed was set at 50 rpm. The composites were cool down by using water. Then, the composite was pelletized. The speed was controlled at 8 rpm to get the same size of the pellet.

The SG fiber filled LDPE/SSP are summarized in Table 1. The SSP content was fixed at 5 wt. % based on the optimum properties of our previous research (Sam et al. 2012, 2011). For compatibilized blends, 0.5% of ENR 50 based on SG was added.

TABLE 1. Formulation of SG fiber filled LDPE/ SSP

LDPE (wt. %)	SSP (wt. %)	Spear Grass (wt. %)
95 (control)	5	0
95	5	1
95	5	2
95	5	5
95	5	10
95	5	15

DRYING THE PELLETT

The pellet was then dried by using vacuum oven to remove the moisture content. The temperature was set at 70°C for 3 h.

COMPRESSION MOLDING

After that, the pellet was compressed by using a hot press. The hot press temperature was maintained at 150°C while

the mould's thickness is 1 mm. The preheating time for the sample was 7 min and the pressing time was 2 min. After that, the samples were cooled for 3 min.

TENSILE TEST

Tensile tests were performed at room temperature according to ASTM D638 by using a universal testing machine (UTM) at a crosshead speed of 50 mm/min. Five specimens of each formulation were tested. Three parameters were measured which are tensile strength, elongation at break (E_b) and Young's modulus.

DIFFERENTIAL SCANNING CALORIMETER (DSC)

DSC was used to measure the heat changes that occur in materials biomolecules during controlled environment in specific range of temperature. The tests were performed by using Pelkin Elmer DSC-7 differential scanning calorimeter. The samples analyzed were sealed in aluminium pans. The samples of 10-15 mg were heated and cooled and the changes in its heat capacity were tracked as changes in the heat flow. It measured the enthalpy (ΔH) of unfolding to heat denaturation. The percentage of crystallinity was calculated by using (1) as follows:

$$\% \text{ crystallinity} = \frac{\Delta H_f^*}{\Delta H_f^0} \times 100\% \quad (1)$$

where ΔH_f^* is the heat of fusion for semicrystalline LDPE and ΔH_f^0 is the heat of fusion for 100% crystalline (276 J/g) (Aalaie et al. 2007).

RESULTS AND DISCUSSION

TENSILE PROPERTIES

Both tensile strength and elongation at break (E_b) of uncompatibilized and compatibilized SG filled LDPE/SSP blends were shown in Figures 1 and 2, respectively. Figure 2 shows that the E_b decreased with the addition of SG into the composites. The reduction of tensile strength and E_b was due to the effect of biodegradable material which is

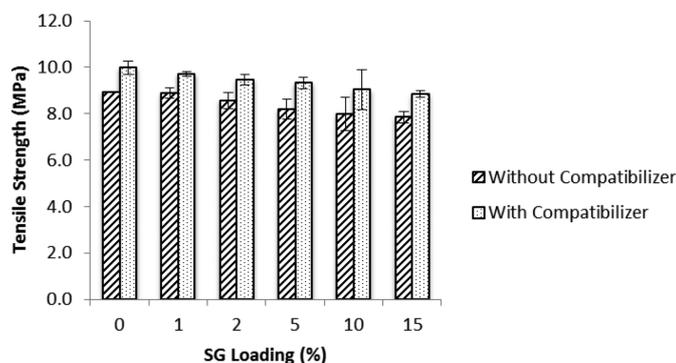


FIGURE 1. Comparison of tensile strength for uncompatibilized and ENR 50 compatibilized SG filled LDPE/SSP composites

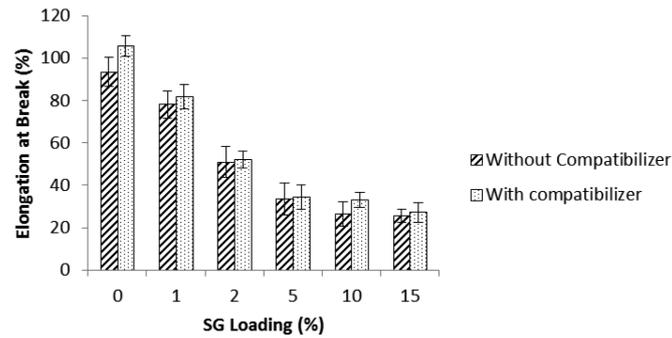


FIGURE 2. Comparison of E_b for uncompatibilized and ENR 50 compatibilized SG filled LDPE/SSP composites

SG and SSP similar to the finding of Mishra and Mahanwar (2002) that reported the effect of potato starch blends in urea with LLDPE. The decreasing also could be due to the lack compatibility between SG and LDPE. Consequently, the stress transfer of SG to the matrix was reduced.

In comparison, the composites with compatibilizer had higher tensile strength and E_b . It is because the interfacial adhesion between SG and LDPE/SSP was improved by the incorporation of ENR 50. The improvement of interfacial adhesion is important in stress transfer (Sam et al. 2011). Tensile fracture surface of LDPE/ SSP, LDPE/SSP filled SG and LDPE/SSP filled SG with the presence of ENR 50 were

shown in Figure 3. From Figure 3, it can be seen that with the increasing of SG cause more agglomeration and rougher tensile surfaces in the blends. After the addition of ENR 50, the agglomeration of SG on the tensile fracture surface was less compared to uncompatibilized blends shows in Figure 4. This structure has fewer voids introduced by fiber pull out (Lee et al. 2013).

The effect of different SG content in LDPE/SSP composites on Young's modulus is shown in Figure 5. The results showed that the Young's Modulus increased with increasing SG loading. This is because increasing of filler loading in LDPE matrix had increased the interaction. The

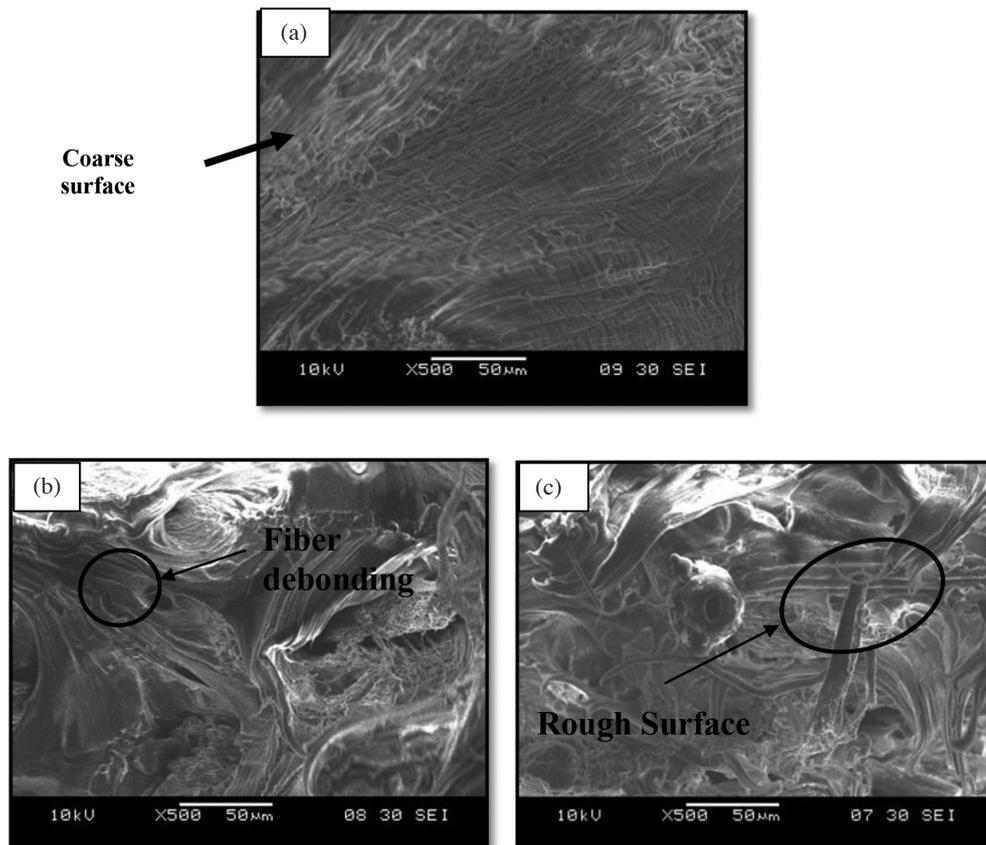


FIGURE 3. Tensile fracture surface of LDPE/SSP with a) 0% SG loading, b) 5% SG loading and c) 15% SG loading

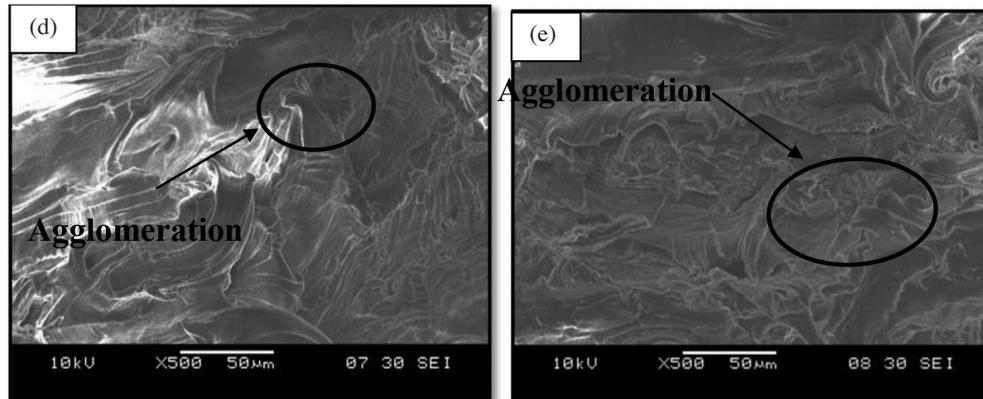


FIGURE 4. Tensile fracture surface of LDPE/SSP with d) 5% SG and 0.5% ENR 50 and e) 15% SG and 0.5% ENR 50

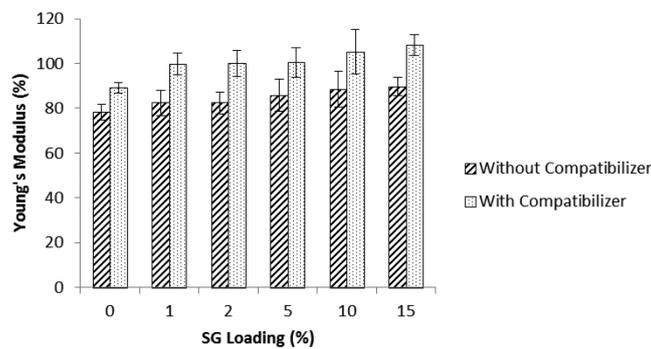


FIGURE 5. Comparison of Young's modulus for uncompatibilized and ENR 50 compatibilized SG filled LDPE/SSP composites

fiber-to-fiber interaction of the SG helps to increase the Young's modulus. During the process, the LDPE were melt, but the filler granules did not melt and retained their shape as rigid filler. The filler are stiffer than LDPE matrix which cause the dispersion during the process (Wan Aizan et al. 2009). Young's modulus for the compatibilized composites was higher than uncompatibilized blends due to better interaction between filler with LDPE/SSP upon the addition of ENR 50 as compatibilizer.

DIFFERENTIAL SCANNING CALORIMETER (DSC)

The heating and cooling thermographs are shown in Figures 6 and Figure 7, respectively. The melting

temperatures, T_m and crystallization temperatures, T_c is summarized in Table 2.

Table 2 summarized the melting temperature (T_m), crystallization temperature (T_c), heat of fusion for semicrystalline LDPE ΔH_f^* and degree of crystallinity. From Table 2, it indicates that the crystallinity of LDPE/SSP decreased with increasing of SG loading. The reduction in crystallinity was due to the addition of SG which hindered the crystallization of LDPE and led to the formation of more complex and less perfect of crystalline during cooling. The decreasing of T_m is due to the increasing of the crytallinity. According to the Hassan et al. (2008), the melting point could be reduced when the crosslinking and branching in the interface between the crystalline and

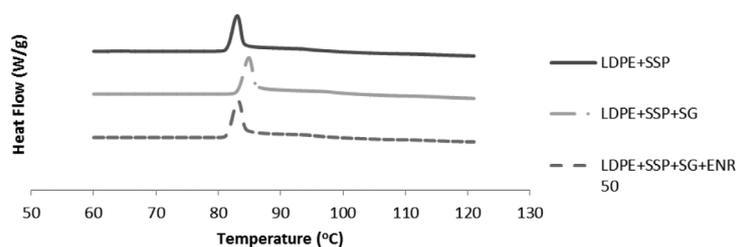


FIGURE 6. DSC melting thermogram of SG filled LDPE/SSP

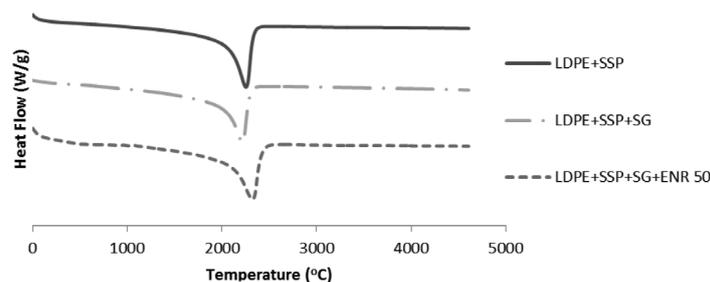


FIGURE 7. DSC cooling thermogram of SG filled LDPE/SSP

TABLE 2. Thermal properties of LDPE/ SSP filled with SG

Sample	T_m (°C)	T_c (°C)	ΔH_f^* (J/g)	Crystallinity (%)
LDPE (95%) + SSP (5%) + SG (0%)	98.75	108.34	101.1 J/g	36.63
LDPE (95%) + SSP (5%) + SG (5%)	98.33	108.05	88.09 J/g	31.92
LDPE (95%) + SSP (5%) + SG (15%)	98.12	108.02	85.73 J/g	31.06
LDPE (95%) + SSP (5%) + SG (0%) + ENR 50 (0.5%)	99.16	109.82	86.60 J/g	31.38
LDPE (95%) + SSP (5%) + SG (5%) + ENR 50 (0.5%)	98.63	108.48	77.63 J/g	28.13
LDPE (95%) + SSP (5%) + SG (15%) + ENR 50 (0.5%)	98.12	108.39	65.22 J/g	26.63

amorphous regions formed slightly impaired crystalline phase.

In ENR 50 compatibilized composites, the crystallinity was slightly decreased compared to uncompatibilized composites. This reduction was due to the interaction of the compatibilizer with SG fiber during mixing consequently reduced the interfacial tension between the SG and LDPE/ SSP composites. The nucleus was then shifted to the interface and the LDPE crystal would grow on the compatible to blends with the compatibilizer. The change to crystallization temperature, T_c is minimal but significant (Sam et al. 2012).

CONCLUSION

The E_b decreased with increasing SG loading from 0 to 15%. The compatibilized SG with LDPE/SSP had higher tensile strength and E_b compared to uncompatibilized composites. On the other hand, the Young's modulus of uncompatibilized and compatibilized SG filled LDPE/ SSP composites increased with increasing of SG loading. The ENR 50 successfully improved the tensile properties. The melting temperature (T_m), crystallization temperature (T_c) and heat of fusion decreased with the addition of SG loading ranging from 0% to 15%.

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