The Effect of Electron Beam Irradiation on PE-g-MA Compatibilised Linear Low-density Polyethylene/Soya Powder Blends

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Abstract: In current investigation, the linear low-density polyethylene (LLDPE)/soya powder blends were compatibilised by polyethylene grafted maleic anhydride (PE-g-MA). The blends were prepared using internal mixer at a rotor speed of 50 rpm and temperature 150°C. The effect of LLDPE/soya powder composition and irradiation dose on the tensile properties of the blends was investigated. The results showed that an increase in soya powder content in the blends reduced the tensile strength and elongation at break. However, the tensile strength of LLDPE/soya powder blends was enhanced with the addition of the PE-g-MA and further improved after irradiated with electron beam (EB) irradiation. The tensile strength of the blends increased with increasing irradiation dose, but the elongation at break decreased. From the morphological study done with scanning electron microscopy (SEM), the LLDPE formed continuous matrix after irradiation.

Keywords: Linear low-density polyethylene, soya powder blends, polyethylene grafted maleic anhydride, electron beam irradiation, tensile properties

1. INTRODUCTION

Polyethylene makes up the largest part of plastic waste and litters from this waste are creating a serious environmental problem. Thus, there is an urgency to develop a form of plastic that can be degraded by oxygen, heat and microorganisms when being exposed to the environment. Griffin introduced an idea of incorporating biodegradable starch into non-degradable plastics such as polyethylene (PE), polypropylene (PP), poly(ethylene terephthalate) (PET) and polystyrene (PS).¹ In recent years, several efforts have been made to incorporate

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starch into thermoplastic. Those studies indicated that starch-based polymeric materials, to some extent, meet the processing and application requirements.

Apart from using polysaccharide-based materials, studies have also been conducted on addition or blends of protein-based materials into polymers.²⁻⁴ However, non-degradable plastics such as PE is naturally hydrophobic due to its hydrocarbon structure. Therefore, it is naturally incompatible with the hydrophilic soya powder. As such, various compatibilisers have been used to improve the interfacial adhesion between PE and polysaccharides or protein based natural polymer such as polyethylene grafted maleic anhydride (PE-g-MA), ethylene acrylic acid copolymer and ethylene–vinyl acetate copolymer (EVA).

Radiation technology has been commonly used to enhance the physical and mechanical properties of plastic materials due to the chemical reaction between polymer molecules under irradiation. In this study, electron beam (EB) irradiation is used to irradiate the blends for a number of reasons. These include: its capability to achieve a high dose rate; reliable sterilisation; safe and easy to operate; and easily controlled radiation dose and rate.⁵

In the current study, soya powder has been used in blends with linear low-density polyethylene (LLDPE). The soya powder contains 60–62% protein, 25–28% polysaccharides, little percentage of fat and moisture. PE-g-MA has been used as a compatibiliser. Based on our previous study,⁶ the mechanical and thermal properties of LLDPE/soya powder is improved with the addition of PE-g-MA. In this investigation, the PE-g-MA compatibilised LLDPE/soya powder blends were irradiated under EB at 30 kGy. Based on previous researches, the optimum radiation dosage to achieve optimum mechanical and thermal properties ranged from 20–50 kGy.^{7–9} Tensile properties and morphological measurements were carried out to compare the properties of non-irradiated and irradiated LLDPE/soya powder blends.

2. EXPERIMENTAL

2.1 Materials

The soya powder was purchased from Hasrat Bestari Sdn. Bhd., Malaysia. The average granular size was 12 μ m. LLDPE granulates with density 0.921 g cm⁻³ and melt flow index 0.90 g 10 min⁻¹ were obtained from Polyethylene Malaysia, Terengganu, Malaysia. PE-g-MA with 3% grafted level was purchased from Aldrich Chemical Company, Milwaukee, United States.

2.2 Preparation of LLDPE/Soya Powder Blends

Haake internal mixer was used to mix the LLDPE and soya powder. The mixing temperature was set at 150°C with the rotor speed of 50 rpm. The LLDPE was first melted for 2 min before the addition of PE-g-MA. After 4 min, soya powder was added gradually into the mixer. The mixing process was followed to be 10 min. Then, the blended samples were compression-molded into sheets of 1-mm thickness using a compression molding machine at 150°C under a pressure of 10 MPa for 10 min. Table 1 shows the blends with the various composition of LLDPE/soya powder used in this study.

Table 1: Composition of LLDPE/soya powder blends compatibilised with PE-g-MA.	Table 1:	Composition	of LLDPE/soya	powder blends	compatibilised	with PE-g-MA.
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Materials	LLDPE (wt%)	Soya powder (wt%)
LLDPE/ 5% soya powder/ PE-g-MA	95	5
LLDPE/ 10% soya powder/ PE-g-MA	90	10
LLDPE/ 15% soya powder/ PE-g-MA	85	15
LLDPE/ 20% soya powder/ PE-g-MA	80	20
LLDPE/ 30% soya powder/ PE-g-MA	70	30
LLDPE/ 40% soya powder/ PE-g-MA	60	40

Note: 50 wt% of PE-g-MA based on soya powder content was added in the blends.

2.3 Irradiation Process

The thin LLDPE/soya powder sheet was EB-irradiated with an electron beam accelerator (model NHV EPS 3000). The irradiation process was carried out at ambient temperature using parameters below:

- (a) Accelerating voltage: 1.5 MeV
- (b) Beam current: 10 mA
- (c) Dose per pass: 15 kGy.

2.4 Insoluble Gel Content

Soxhlet extraction technique was used to determine insoluble gel content. Initially, the samples were extracted by using distilled water to remove the soya powder for 24 h. Thereafter, secondary extraction was performed to remove the LLDPE for 24 h in xylene. The samples were air-dried and kept in vacuum oven at 60° C until the weight was constant. The gel content was calculated using the following equation:

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Insoluble gel content (%) =
$$\frac{\text{Weight after extraction (g)}}{\text{Weight before extraction (g)}} \times 100\%$$

2.5 Tensile Properties

Tensile tests of the blends were performed using a tensometer named Instron (model 3366) according to ASTM D638 at crosshead speed of 50 mm min⁻¹. A minimum of five specimens for each blending ratio was tested. Tensile strength, elongation at break and Young's modulus were obtained from the tensile test.

2.6 Scanning Electron Microscope (SEM)

The tensile fracture surface of the blends was analysed using a scanning electron microscope (VPFESEM) model SUPRA 35VP. The surface was coated with gold in order to avoid electrostatic charging.

3. **RESULTS AND DISCUSSION**

3.1 Gel Content Analysis

Gel content is an estimation of the crosslink density for irradiated polymeric materials. Figure 1 shows the gel content of the PE-g-MA compatibilised LLDPE/soya powder blends with 30 kGy irradiation. The gel content in non-irradiated blends was solely contributed by the formation of crosslinking between PE-g-MA and soya powder as a result of ester bond formed. In our previous investigation,¹⁰ the PE-g-MA was proven as a good compatiliser for LLDPE and soya powder.

As can be seen in Figure 1, the gel content of the irradiated blends was generally higher than the non-irradiated blends. The trend indicates that crosslinking occurred during irradiation process. The crosslinking occured within LLDPE, PE-g-MA and grafting of PE-g-MA on LLDPE but not within soya powder. This could be due to the fact that polypeptide in soya powder is a radiation degradation polymer. The result is in agreement with Zhai et al.¹⁰ who investigated the effect of irradiation on polyvinyl alcohol (PVOH)/polysaccharide blends.

In their study, a few components in polysaccharides were found to be susceptible to EB irradiation even in low dosage. Thus, the soya powder, which also contains polysaccharides cannot form radical site under low irradiation. In Figure 1, the gel content after irradiation decreased with increasing soya powder content. It is proven that increasing the volume of soya powder had hindered the radical formed in LLDPE and ENR 50. Subsequently, the crosslink density was reduced.



Figure 1: Comparison of gen content between non-irradiated blends and irradiated blends.

3.2 Tensile Properties

Figure 2 demonstrates the tensile strength of non-irradiated and irradiated compatibilised LLDPE/soya powder blends. The tensile strength decreased with increasing soya powder content due to the incompatibility between hydrophobic LLDPE and hydrophilic soya powder.⁶ The incorporation of PE-g-MA had successfully compatibilised LLDPE and soya powder, consequently increasing the tensile strength. After EB irradiation, the compatibilised LLDPE/soya powder was further improved. This is due to the crosslinking formed between polymer chains of LLDPE and PE-g-MA had enhanced the tensile strength of the blends.

Figure 3 shows the comparison of elongation at break between nonirradiated blends and irradiated blends. Generally, the elongation at break of the blends decreased after EB irradiation. The decrement is attributed to the occurrence of irradiation-induced crosslinking in the blend. The polymer chains were crosslinked after irradiation, subsequently resisted the mobility of the polymer molecules. The trend is in agreement with various EB irradiation studies.^{7,11}



Figure 2: Tensile strength of non-irradiated blends and irradiated blends.

Figures 4 and Figures 5 show the SEM micrographs of tensile fractured surface of the non-irradiated and irradiated blends, respectively. For non-irradiated blends, fibrils can be found on the tensile fracture surface. The presence of fibril indicates the high elongation at break of PE-g-MA compatibilised blends as discussed in a previous study.⁶ However, it can be observed that the decrease in fibril on the tensile fracture surface corresponded with the result of reduction in elongation at break.



Figure 3: Tensile strength of non-irradiated blends and irradiated blends.



Figure 4: Tensile fracture surface of non-irradiated LLDPE/soya powder blends with (a) 5 wt%, (b) 20 wt% and (c) 40 wt% soya powder content.



Figure 5: Tensile fracture surface of EB irradiated LLDPE/soya powder blends with (a) 5 wt%, (b) 20 wt% and (c) 40 wt% soya powder content.

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Figure 6 demonstrates the comparison of Young's modulus for irradiated and non-irradiated blends. The Young's modulus was improved after the EB irradiation. The enhancement might be due to the occurrence of irradiation-induced crosslinking in the polymer matrix as indicated by the gel content results in Figure 1. The EB irradiation resulted in the formation of network structure in the blend and consequently increased the stiffness. According to Bhowmick et al., the increase of modulus and reduction in elongation at break are proportional to the crosslink density.¹²



Figure 6: Comparison of Young's modulus between non-irradiated blends and EBirradiated blends.

4. CONCLUSION

The EB irradiation on PE-g-MA-compatibilised blends had improved the tensile strength and Young's modulus whereas the elongation at break decreased. The gel content, which is an indicator of crosslink density, was found to increase after the irradiation process. Therefore, the formation of crosslinking is proven.

5. ACKNOWLEDGEMENT

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