

Effect of the Electron Beam Irradiation on the Properties of Epoxidized Natural Rubber (ENR 50) Compatibilized Linear Low-Density Polyethylene/Soya Powder Blends

S. T. Sam,¹ H. Ismail,¹ Z. Ahmad,¹ C. T. Ratnam²

¹School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Pulau Pinang, Malaysia

²Radiation Processing Technology Division, Malaysian Nuclear Agency, Bangi, Kajang, Malaysia

Received 18 October 2010; accepted 10 January 2011

DOI 10.1002/app.34136

Published online 14 December 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: Linear low-density polyethylene/soya powder blends were prepared by using an internal mixer at 150°C. The soya powder content ranged from 5 to 40 wt %. Epoxidized natural rubber with 50 mol % epoxidation (ENR 50) was added as a compatibilizer. The blends were irradiated by electron beam (EB) at a constant dose of 30 kGy. The changes in gel fraction, tensile properties, morphological and thermal properties of the samples were investigated. The gel content increased after EB irradiation. However, the increment of gel content was hindered by increasing soya powder content. The tensile strength and Young's modulus of the blends were increased by EB whereas the elongation at break decreased. The tensile frac-

ture surface also support the reduction of elongation at break by EB irradiation. Further analysis on the irradiated blends using Fourier transform infrared spectra indicated an increase of oxygenated product after undergoing EB irradiation. The differential scanning calorimetry result indicated that the melting temperature of the blends decreased after EB irradiation whereas the crystallinity increased. EB irradiation also enhanced the thermal stability of the blends as indicated by thermogravimetric analysis. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 5220–5228, 2012

Key words: linear low-density polyethylene; soya powder; epoxidized natural rubber; electron beam irradiation

INTRODUCTION

Petroleum-based plastics have been widely used in packaging industry. Today, the position of the plastics is irreplaceable.¹ However, the litter from the plastics waste is creating serious environmental problem. Polyethylene (PE) is the biggest contribution in plastic waste. Thus, the development of degradable plastics that could be degraded by oxygen, heat, and microorganisms in exposed environment or soil is essential. Griffin introduced an idea of incorporating biodegradable starch into nondegradable plastics such as PE, polypropylene (PP), poly(ethylene terephthalate) (PET), and polystyrene (PS).² In recent years, several efforts have been made to incorporate starch into thermoplastic.^{3–7} Starch is polysaccharide-based materials, which is renewable and biodegradable. Those studies indicated that the starch-based polymeric materials have met some extent of processing and application requirement. Apart from using poly-

saccharide-based materials, studies have also been conducted on addition or blends of protein-based materials into polymers.^{8–10} However, nondegradable plastic such as PE is naturally hydrophobic because of its hydrocarbon structure. Consequently, it is incompatible with the hydrophilic soya powder. As such, various compatibilizers 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),^{11,12} ethylene acrylic acid copolymer,^{13–15} and ethylene-vinyl acetate copolymer.¹⁶

Apart from using compatibilizer to enhance the interfacial adhesion between polyolefin and protein-based natural polymer, radiation technology has commonly been used to enhance the physical and mechanical properties of plastic materials due to the chemical reaction between polymer molecules under irradiation. In this study, EB irradiation is used to irradiate the blends because EB irradiation has following advantages: (a) very high-dose rate achievable, (b) reliable sterilization, (c) safe and easy to operate, and (d) radiation dose and rate are easy to be controlled.¹⁷ EB irradiation has been used in polymer technology to improve the compatibility between polymer blends, for example, PP/epoxidized natural rubber blends,¹⁸ starch-modified PP blends,¹⁹ and low-density polyethylene (LDPE)/plasticized starch

Correspondence to: H. Ismail (hanafi@eng.usm.my).

Contract grant sponsor: RU Grant; contract grant number: 1001/PBAHAN/814008

Contract grant sponsors: USM-RU-PRGS Grant (Universiti Sains Malaysia), USM Fellowship.

blends.²⁰ However, not much work is reported on PE/protein-based polymer blends.

In 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. Epoxidized natural rubber with 50 mol % epoxidation (ENR 50) has been used as a compatibilizer. Based on our previous study,²¹ the mechanical and thermal properties of LLDPE/soya powder is improved with the addition of ENR 50. In this article, the compatibilized LLDPE/soya powder blends was irradiated under EB at 30 kGy. The dosage was selected based on previous research as optimum radiation dosage to achieve optimum mechanical and thermal properties ranged from 20 to 50 kGy.^{20,22,23} Gel content, tensile, morphological and thermal properties measurements were carried out to compare the properties of nonirradiated and irradiated LLDPE/soya powder blends.

EXPERIMENTAL

Materials

The soya powder was supplied by Hasrat Bestari Sdn. Bhd, Malaysia. The moisture content was 3.12% before drying and the average granular size was 12 μm . LLDPE granulates with density 0.921 g/cm^3 , and melt flow index 0.90 $\text{g}/10$ min was obtained from Polyethylene Malaysia Sdn. Bhd, Malaysia. ENR 50 was supplied by Rubber Research Institute of Malaysia (Kuala Lumpur, Malaysia).

Preparation of LLDPE/soya powder blends

The blends were prepared by melt blending in a Haake internal mixer at temperature of 150°C and rotor speed of 50 rpm. The LLDPE was first melted for 2 min before the ENR 50 was added. At 4th 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 Kao Tieh GoTech compression molding machine at 150°C under a pressure of 10 MPa for 10 min. Table I shows the blends with the various composition of LLDPE/soya powder used in this study.

Irradiation process

The EB irradiation was carried out in Malaysia Nuclear Agency by using an electron beam accelera-

TABLE I
Composition of LLDPE/Soya Powder Blends
Compatibilized with ENR 50

Materials	LLDPE (wt %)	Soya powder (wt %)
LLDPE/5% soya powder/ENR 50	95	5
LLDPE/10% soya powder/ENR 50	90	10
LLDPE/15% soya powder/ENR 50	85	15
LLDPE/20% soya powder/ENR 50	80	20
LLDPE/30% soya powder/ENR 50	70	30
LLDPE/40% soya powder/ENR 50	60	40

50 wt % of ENR 50 based on soya powder content was added in the blends.

tor (model NHV EPS 3000). The thin sheet of the LLDPE/soya powder blends was irradiated at ambient temperature using parameters below:

- Accelerating voltage: 1.5MeV
- Beam current: 10mA
- Dose per pass: 15 kGy.

FTIR measurement

Fourier transform infrared (FTIR) spectra were acquired using a Perkin–Elmer model Series 2 FTIR spectrometer. The spectrometer was operated with a resolution of 4 cm^{-1} and scanning ranges from 4000 to 400 cm^{-1} . Samples were measured by attenuated reflection method in the form of thin sheet of thickness about 1 mm.

Insoluble gel content

Insoluble gel content was determined by using Soxhlet extraction technique. The samples were first extracted using distilled water to remove the soya powder for 24 h. The remains were then extracted again with xylene during secondary extraction to remove the LLDPE for 24 h. Then, the samples were air dried and kept in vacuum oven at 60°C until constant weight was obtained. The gel content was calculated using the following equation:

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

The increment of the gel content after irradiation was calculated using the following equation:

$$\text{Increment of gel content (\%)} = \frac{\text{Irradiated blends gel content} - \text{nonirradiated blends gel content}}{\text{nonirradiated blends gel content}} \times 100\%$$

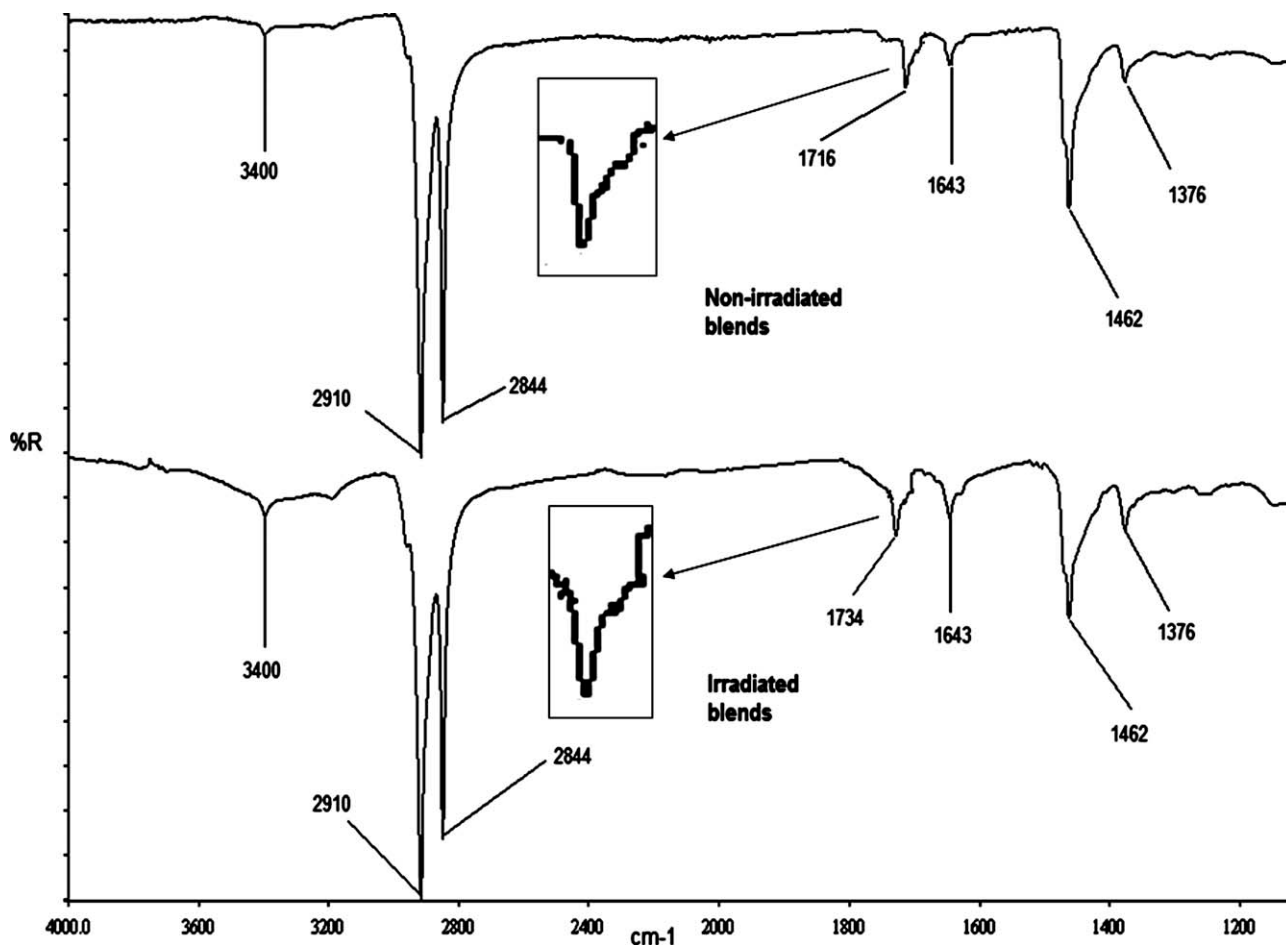


Figure 1 FTIR spectra of nonirradiated and irradiated LLDPE/soya powder blends with the soya powder content of 40 wt % and a zoom of C=O band.

Tensile properties

Measurements of the tensile properties were performed using a tensometer named Instron (model 3366) according to ASTM D638 at crosshead speed of 50 mm/min. Five specimens of each blending ratio were tested. The parameters were measured: tensile strength, elongation at break, and Young's modulus.

Scanning electron microscope (SEM)

The tensile fracture surface of the blends was analyzed using scanning electron microscope (VPFE-SEM) model SUPRA 35VP. The surface was coated with gold to avoid electrostatic charging.

Differential scanning calorimetry (DSC)

Thermal behavior of the nonirradiated and irradiated blends, which includes melting temperature (T_m) and enthalpy of melting (ΔH_m), crystallization temperature, and crystallize enthalpy (ΔH_c) were determined by using Perkin-Elmer DSC 7 based on ASTM D3418-03. The crystallinity of the nonirradi-

ated and irradiated blends was calculated using the following equation:

$$\% \text{ crystallinity} = \frac{\Delta H_f^*}{\Delta H_f^o} \times 100\%$$

where ΔH_f^o is heat of fusion for 100% crystalline PE (276 J/g) and ΔH_f^* is heat of fusion for the semicrystalline LLDPE.²⁴ Each 4-mg-average sample was encapsulated in a hermetically sealed aluminum pan. The sample was first heated up from -135 to 200°C at $10^\circ\text{C}/\text{min}$ and this temperature was remained constant for 5 min to remove the thermal history. Subsequently, the sample was cooled to room temperature at the same rate and then reheated to 200°C at $10^\circ\text{C}/\text{min}$. Four measurements have been taken for each sample.

Thermal gravimetric analysis (TGA)

Perkin-Elmer Pyris 6 TGA analyzer was used for measurement of thermal stability of the blends. In the current study, nitrogen flow was kept at constant rate of about 50 mL/min. About 10 mg of samples

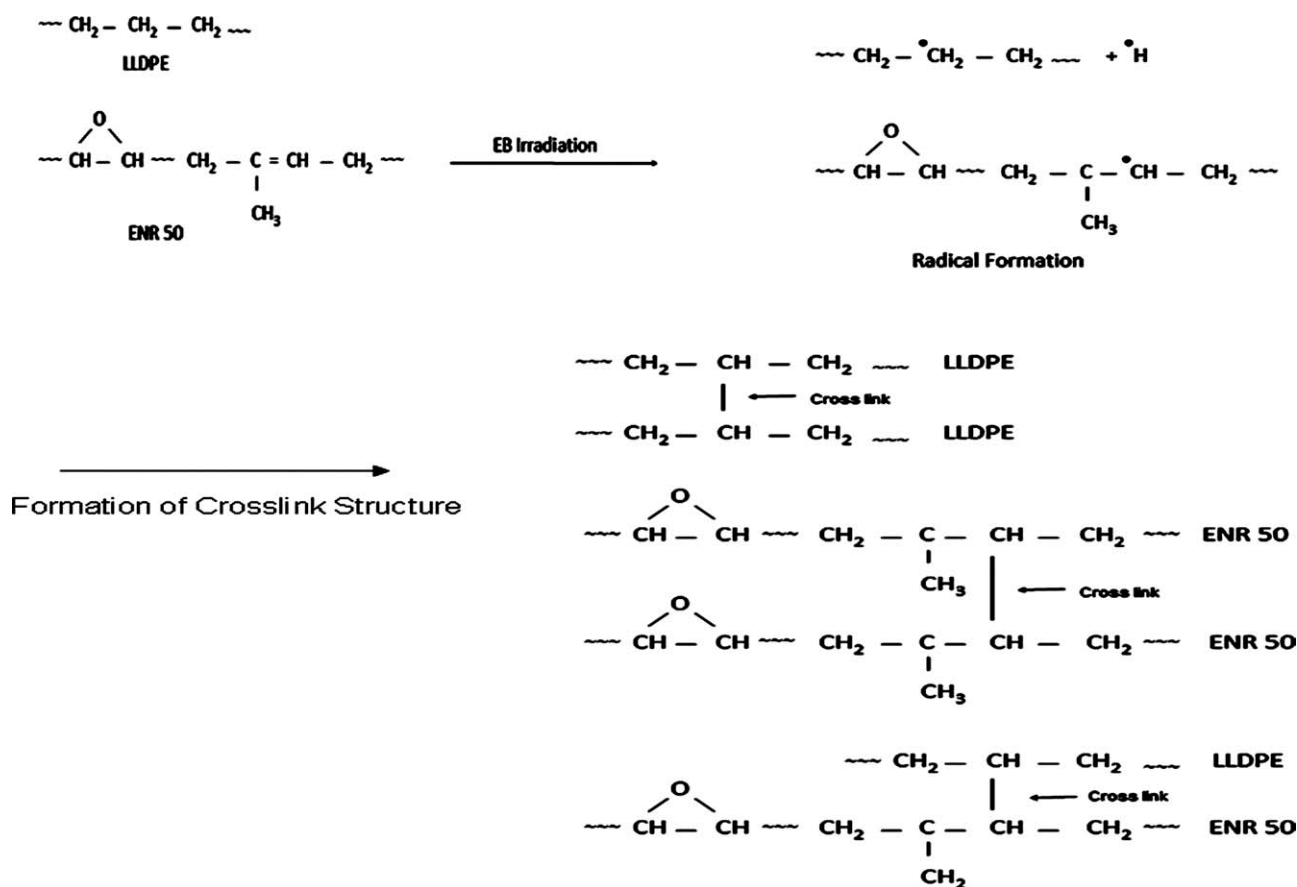


Figure 2 Schematic diagram of crosslinking reaction of LLDPE and ENR 50 under EB irradiation.

was used in the study. The heating rate of the test was $10^\circ\text{C}/\text{min}$. The degradation temperature at 5% weight loss ($T_{-5\%}$) and degradation temperature at 30% weight loss ($T_{-30\%}$) were measured through TGA thermogram.

RESULTS AND DISCUSSION

FTIR studies

FTIR spectroscopy in Figure 1 illustrates the structure of LDPE/soya powder blends in the presence of ENR 50 as a compatibilizer and the effect of EB irradiation dose at 30 kGy. The tiny peak at 1716 cm^{-1} indicates the formation of ester group due to the compatibilization reaction between ENR 50 and soya powder. The epoxy group of ENR 50 underwent epoxy ring opening during the high-temperature mixing in the internal mixer. Consequently, the ring opening reacted with the C—OH-group (carboxylic acid) of soya powder and formed an ester group. The chemical reaction formed between ENR 50 and soya powder has been reported in our previous investigation.²¹ FTIR can also be used to illustrate the chemical changes that occurred during EB irradiation. The formation of oxidative products

such as C=O and O—H can also be observed. The normalized peak of the O—H group (3400 cm^{-1}) referred to the C—H₂ band (2844 cm^{-1}) of LLDPE increased and broad C=O existed ($1785\text{--}1700\text{ cm}^{-1}$) indicating the effect of EB irradiation on LLDPE/soya powder blends. This result is in agreement with Senna and Abdel-Monem²⁰ in their investigation on LDPE/plasticized starch blends. It can be observed that the intensity of the ester group (1716 cm^{-1}) also increased after irradiation. Based on the finding of Ratnam et al.,²⁵ the irradiation can cause ring opening of ENR 50 and leads to the crosslinking between the polymer chains. Therefore, the increase in ester group intensity is believed to be associated with the increase of reaction between ring opening of ENR 50 and carboxylic group in soya powder upon irradiation. The proposed crosslinking reaction of LLDPE and ENR 50 is shown in Figure 2. The crosslinking process of the blends will be discussed in the following section.

Gel content analysis

Gel content data are used to estimate the crosslink density of irradiated polymeric materials.²⁶ The gel content of the compatibilized LLDPE/soya powder

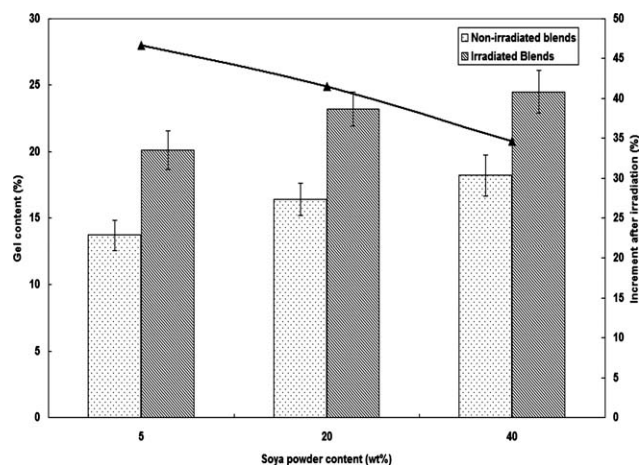


Figure 3 Effect of soya powder content and EB irradiation on gel content of LLDPE/soya powder blends.

blends with 30 kGy irradiation is shown in Figure 3. The gel content in nonirradiated blends was contributed by formation of crosslinking between ENR 50 and soya powder as a result of ester bond formed. The compatibilization effect between LLDPE and soya powder with the presence of ENR 50 has been reported in our previous study.²¹ Generally, the gel content of the irradiated blends was higher than the nonirradiated blends. This indicates that crosslinking occurred during irradiation process. Figure 4 demonstrates the proposed crosslink network in the compatibilized LLDPE/soya powder blends. It shows that the crosslink only occur within LLDPE, ENR 50, and grafting of ENR 50 on LLDPE. However, the crosslinking process did not occur within soya powder or between soya powder and other polymer matrices. This could be due to the fact polypeptide in soya powder is a radiation degradation polymer. The result is in agreement with Zhai et al.²⁷ in their investigation on the effect of irradiation on polyvinyl

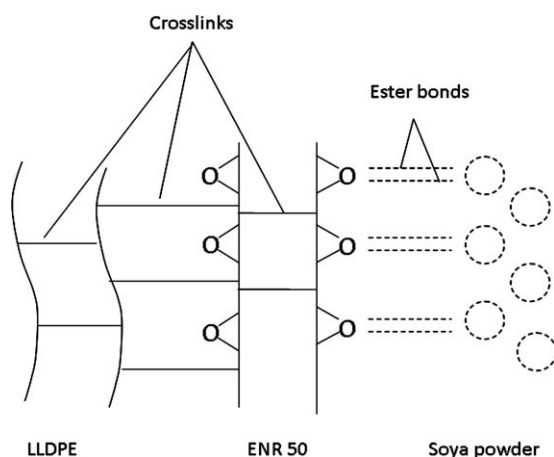


Figure 4 Schematic diagram of crosslink structure of LLDPE/ENR/soya powder after EB irradiation.

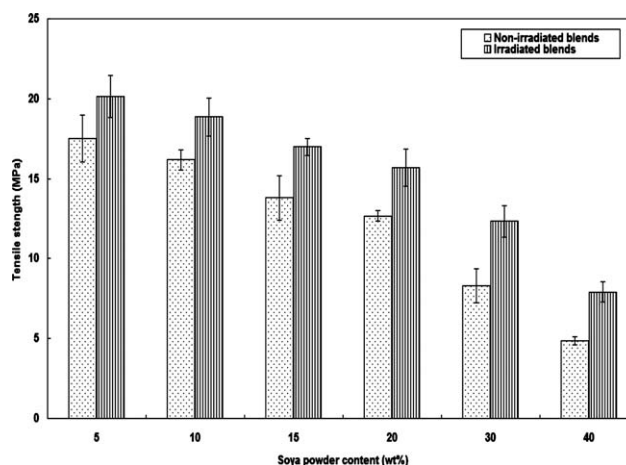


Figure 5 Comparison of tensile strength for nonirradiated and irradiated blends in various soya powder content.

alcohol (PVOH)/polysaccharide blends. They suggested that a few components in polysaccharides were susceptible to EB irradiation even in low dosage. Therefore, the soya powder that also contains polysaccharides cannot form radical site under low irradiation and might degrade at higher irradiation dosage. It can be observed from Figure 3, the gel content increment after irradiation decreased with increasing soya powder content. The result might be due to the increasing volume of soya powder had hindered the radical formed in LLDPE and ENR 50. Subsequently, the crosslink density was reduced. Zhai et al.²⁷ also reported similar observation.

Tensile properties

Tensile properties are another good indicator of crosslink density and energy dissipation.²⁸ Figure 5 shows the tensile strength of nonirradiated and irradiated LLDPE/soya powder blends. The tensile

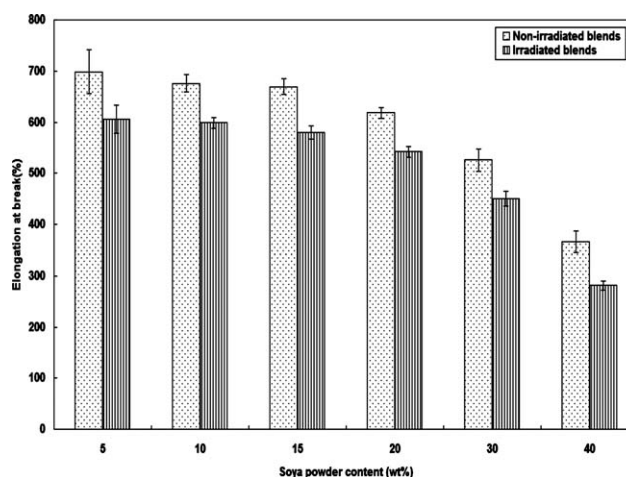


Figure 6 Comparison of elongation at break for nonirradiated and irradiated blends in various soya powder content.

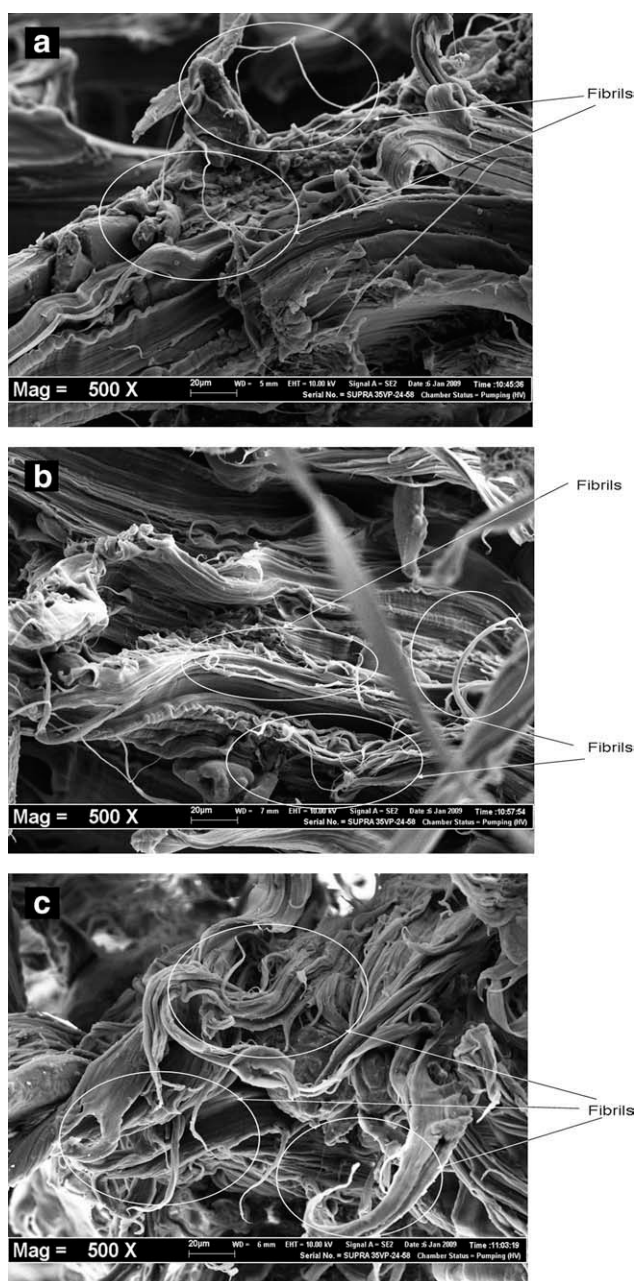


Figure 7 (a–c) Tensile fracture surface ($\times 500$ magnification) of nonirradiated LLDPE/soya powder blends with: (a) 5 wt %, (b) 20 wt %, and (c) 40 wt % soya powder content.

strength generally decreased with increasing soya powder content due to the incompatibility between hydrophobic LLDPE and hydrophilic soya powder. However, ENR 50 had successfully compatibilized LLDPE and soya powder as discussed in our previous study.²¹ The blends showed significant improvement in tensile strength after exposure to EB irradiation. The crosslinking that formed between polymer chains of LLDPE and ENR 50 had enhanced the tensile strength of the blends in a certain extent.

Nevertheless, the elongation at break of the blends decreased after EB irradiation as indicated in Figure 6. This observation again is attributed to the occurrence of irradiation-induced crosslinking in the blend. The polymer chains were locked by the crosslink after irradiation, consequently resisted the mobility of the polymer molecules. The phenomenon of reduction of elongation at break for irradiated polymer is in agreement with various EB irradiation studies.^{19,25} SEM micrographs of tensile fractured surface of the nonirradiated and irradiated blends are shown in Figures 7(a–c) and 8(a–c), respectively. For nonirradiated blends, fibrils can be found on the

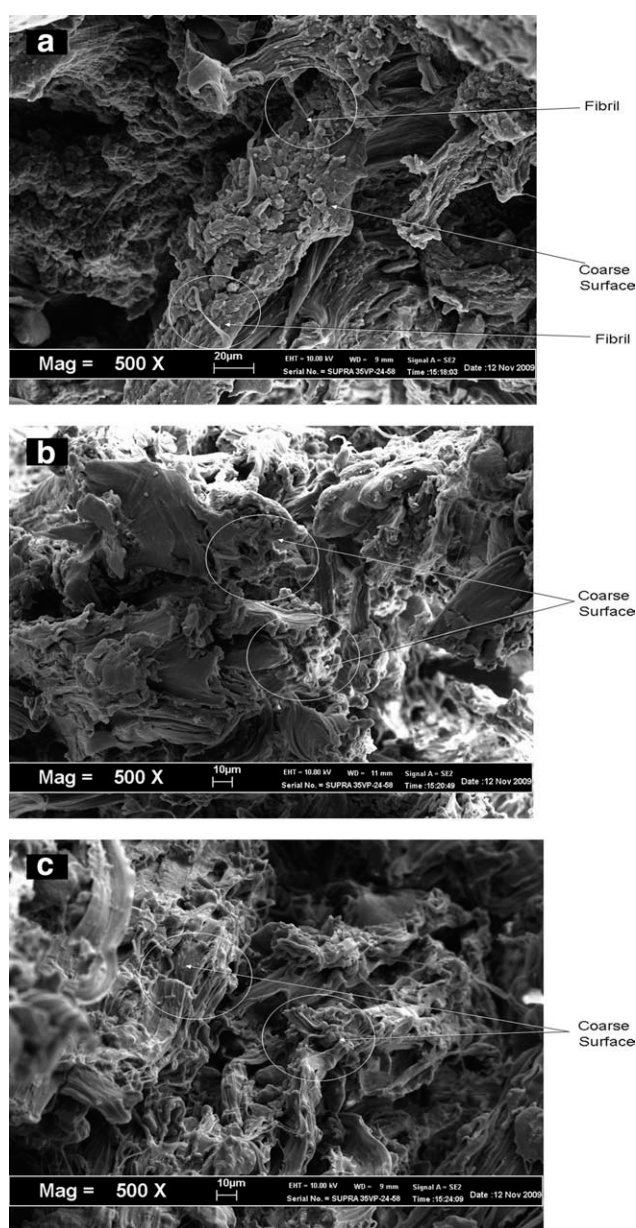


Figure 8 (a–c) Tensile fracture surface ($\times 500$ magnification) of EB-irradiated LLDPE/soya powder blends with: (a) 5 wt %, (b) 20 wt %, and (c) 40 wt % soya powder content.

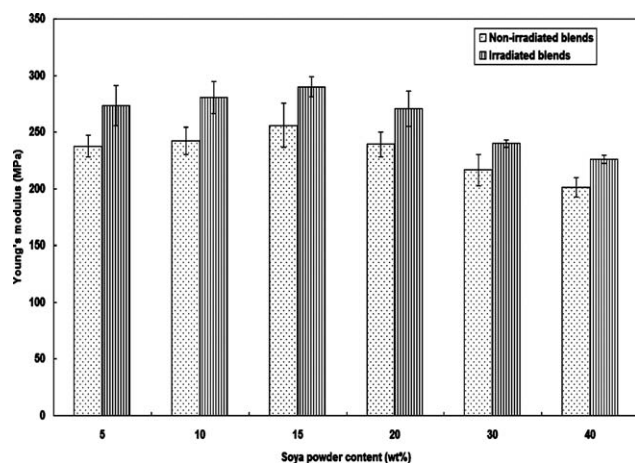


Figure 9 Comparison of Young's modulus for nonirradiated and irradiated blends in various soya powder content.

tensile fracture surface. The presence of fibril implies the high elongation at break of ENR 50 compatibilized LLDPE/soya powder blends as discussed in previous study.²¹ In irradiated blends, the decrease in fibril on the tensile fracture surface corresponded with the result of reduction in elongation at break.

In addition to that, it is observed that EB irradiation also contributed to the increase of Young's modulus of the LLDPE/soya powder blends (Fig. 9). This enhancement in Young's modulus is believed to be due to the occurrence of irradiation-induced crosslinking in the polymer matrices (LLDPE and ENR 50) as indicated by the gel content results in Figure 3. The formation of network structure in the blend upon exposure to irradiation thus increased

the stiffness. Bhowmick et al.²⁹ has concluded in their study that the increase of modulus and reduction in elongation at break are proportional to the crosslink density.

DSC analysis

DSC analysis was used to determine the variation in the thermal properties and the structural changes in compatibilized LLDPE/soya powder blends. DSC measurements include melting temperature (T_m), enthalpy of melting (ΔH_m), and crystallization temperature (T_c). The crystallinity can be calculated from ΔH_m as previously mentioned. It is observed that the T_m , T_c , and crystallinity (Table II) decreased with the addition of soya powder. Our earlier study also indicated that the soya powder content could affect the T_m of the blends for a certain extent.²¹ Decrease in crystallinity was due to the addition of the soya powder which hindered the crystallization of LLDPE and led to the formation of more complex and less perfect of crystalline during cooling. Furthermore, the migration and diffusion of molecular-chain branched PE was limited by interfacial adhesion between soya powder and LLDPE, consequently hampering the crystallization of LLDPE during cooling.

Based on the DSC measurement for T_m , ΔH_m , T_c , and crystallinity, similar trend was observed for both irradiated and nonirradiated blends for different soya powder content in the blends (Table II). The melting thermogram and cooling thermogram for nonirradiated blends and irradiated blends are shown in Figures 10 and 11, respectively. Since the

TABLE II
DSC Results of Nonirradiated and Irradiated ENR 50 Compatibilized LLDPE/Soya Powder Blends

Sample	T_m (°C)		T_c (°C)		ΔH_f^*		Crystallinity (%)	
	Value	Standard deviation	Value	Standard deviation	Value	Standard deviation	Standard deviation	Standard deviation
LLDPE/5% soya powder/ENR 50 (nonirradiated)	122.7	0.14	102.2	0.17	31.8	1.65	11.5	0.07
LLDPE/20% soya powder/ENR 50 (nonirradiated)	120.2	0.17	101.5	0.24	27.6	1.11	10.0	0.22
LLDPE/40% soya powder/ENR 50 (nonirradiated)	118.9	0.15	100.2	0.19	25.1	1.25	9.1	0.16
LLDPE/5% soya powder/ENR 50 (irradiated)	119.5	0.11	99.7	0.22	35.8	1.10	13.0	0.13
LLDPE/20% soya powder/ENR 50 (irradiated)	116.1	0.13	97.7	0.27	32.2	0.89	11.7	0.11
LLDPE/40% soya powder/ENR 50 (irradiated)	114.7	0.16	96.2	0.12	29.0	1.61	10.5	0.25

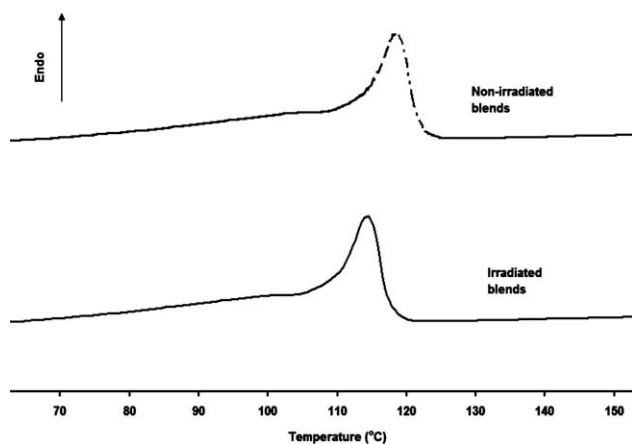


Figure 10 Melting thermogram of nonirradiated and irradiated blends.

melting and cooling trend for all soya powder content in the blends were similar, blends with 40 wt % soya powder content were used to represent the thermal behavior of the blends in Figures 10 and 11, respectively. The decrease in T_m after irradiation was attributed to the reduction of the molecules of amorphous region due to the chain scission; consequently weaken the interlamellar connection according to Zhang et al.³⁰ The decrease of T_m was concomitant to the increase of crystallinity of the blends after EB irradiation as shown in Table II. According to Hassan et al.,³¹ the crosslinking reaction can only occur in amorphous region of polymer if the irradiation temperature is lower than its melting temperature. Nevertheless, the melting temperature could be reduced when the branching and crosslinking in the interface between the crystalline and amorphous regions formed a slightly impaired crystalline phase. This observation is in agreement with the reduction of T_m and increase of crystallinity in this study.

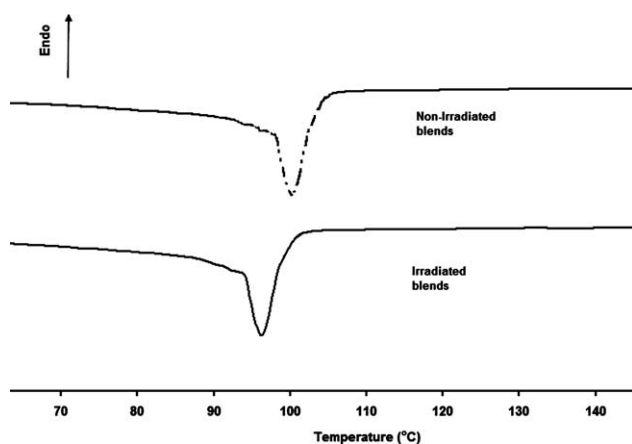


Figure 11 Cooling thermogram of nonirradiated and irradiated blends.

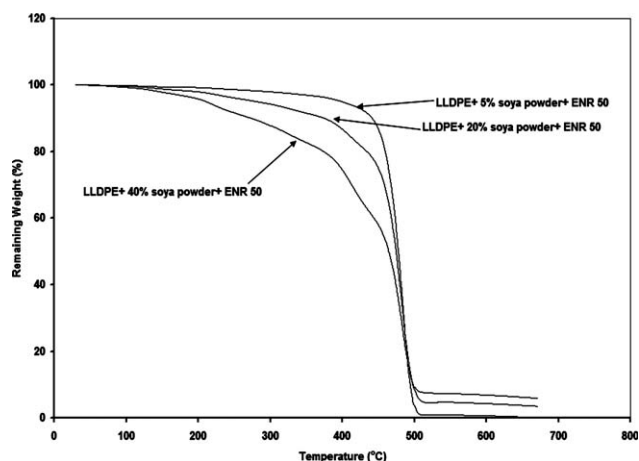


Figure 12 TGA thermogram of nonirradiated LLDPE/soya powder blends.

Thermogravimetric decomposition behaviors

TGA thermograms of nonirradiated and irradiated compatibilized LLDPE/soya powder blends were shown in Figures 12 and 13, respectively. The TGA data were listed in Table III. TGA thermograms (Figs. 12 and 13) showed that the thermal stability of the blends decreased with increasing soya powder content as the decomposition temperature started at lower temperature for higher soya powder content. This could be due to the low thermal stability of soya powder. Soya powder is a globular protein-based material,³² and its thermal resistance is relatively low compared to petroleum-based polymers. Based on our previous study,²¹ the incorporation of elastomeric compatibilizer, i.e., ENR 50 has significantly enhanced the thermal stability of the blends due to the improved interfacial adhesion between LLDPE and soya powder. However, the interfacial adhesion of compatibilized LLDPE/soya powder blends was

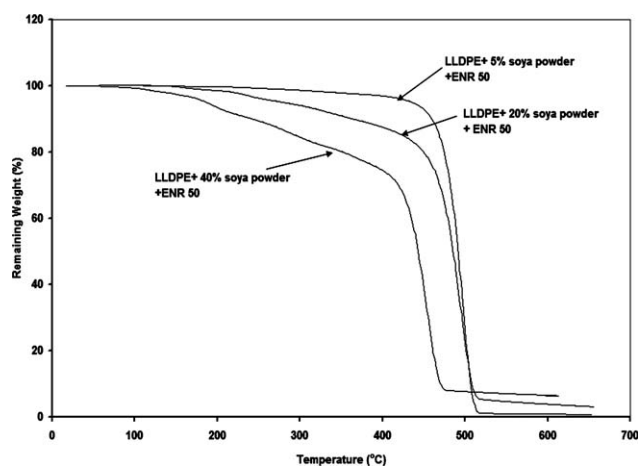


Figure 13 TGA thermogram of EB-irradiated LLDPE/soya powder blends.

TABLE III
TGA Data for Nonirradiated and Irradiated ENR 50 Compatibilized LLDPE/Soya Powder Blends

Sample	$T_{-5\%}$ (°C)		$T_{-30\%}$ (°C)	
	Nonirradiated	Irradiated	Nonirradiated	Irradiated
LLDPE/5% soya powder/ENR 50	402	410	468	482
LLDPE/20% soya powder/ENR 50	280	287	459	469
LLDPE/40% soya powder/ENR 50	211	215	413	419

reduced at the higher degradation temperature. Thus, the change in T_{-30} was not significant.

On the other hand, the T_{-5} and T_{-30} of irradiated blends was higher than the nonirradiated blends as shown in Table III. The results indicated that the thermal stability of compatibilized LLDPE/soya powder blends was enhanced by EB irradiation. Higher dissociation energy from crosslinking process might have resulted in the improvement of thermal stability. After irradiation process, more covalent bonds, i.e., C—C (348 kJ/mol), C—O (360 kJ/mol), C—H (413 kJ/mol), and O—H (366 kJ/mol), which have high dissociation energy were formed. This argument was in agreement with the FTIR result as discussed in the previous section. Hence, the irradiated LLDPE/soya powder blends with any soya powder content resulted in higher thermal stability compared to nonirradiated blends.

CONCLUSIONS

The increase in gel content upon irradiation indicated that the irradiation-induced crosslinking occurred in the blends. Results revealed that the EB irradiation of LLDPE/soya powder blends has improved the tensile strength and Young's modulus of the blend with a concomitant decline in elongation at break. The introduction of EB irradiation also reduced the T_m and T_c of the blends but increased the crystallinity of the blends. The thermal stability of the blends was also improved after EB irradiation.

The authors are deeply grateful to Malaysian Nuclear Agency for providing the electron beam irradiation facility.

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