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Degradation of epoxidized natural rubber compatibilized linear low density polyethylene/soya powder blends: the effect of natural weathering

Abstract: In the present study, linear low density polyethylene (LLDPE)/soya powder blends were compatibilized with epoxidized natural rubber (ENR 50) and exposed to natural weathering. The exposure period for the blends was 1 year. It was found that the degradability of the compatibilized blends was higher than that of uncompatibilized blends. Fourier transform infrared (FTIR) spectra, the tensile test, scanning electron microscopy (SEM), and differential scanning calorimetry (DSC) were applied to analyze the degradability of the blends. IR spectra showed that the carbonyl index (CI) of the blends increased as a function of exposure period and soya powder content. The compatibilized blends gave higher carbonyl indices. The retention tensile strength and elongation at break (E_b) of the compatibilized blends after weathering was generally lower than for the uncompatibilized blends. The increase of crystallinity also indicated a reduction of the amorphous portion after degradation. The higher crystallinity in compatibilized blends further confirms the higher degradability of ENR 50 compatibilized blends. The weight loss and molecular weight change indicated that the incorporation of ENR 50 into LLDPE/soya powder blends can enhance the degradability of the blends upon outdoor exposure.

Keywords: ENR 50; LLDPE; natural weathering; soya powder.

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1 Introduction

Polyethylene is highly resistant to chemical attack, environmental weathering, and biotic consumption. This stable polymer contributes a major plastic waste in the world, particularly in packaging applications. Therefore, a more environmentally friendly polymer needs to be produced to resolve the plastic waste problem. In industry, cost is a challenge in inventing or producing a more environmentally friendly polymer. In the market, there are many commercial biopolymers including poly (lactic acid) (PLA), polycaprolactone (PCL), polyhydroxybutyrate (PHB), and polyvinyl alcohol (PVA). Nevertheless, their prices are very high in packaging applications, even though some of the mechanical, physical, and thermal properties are comparable to polyolefin.

A cheaper alternative to produce a degradable polymer is to blend with natural polymers, such as starches. There has been much research in blending the non-degradable polyolefin and starches, including potato starch [1], tapioca starch [2, 3], rice starch [4], and corn starch [5]. In addition to the polysaccharide-based starches, a protein-based natural polymer, soya powder, is a potential natural polymer to be incorporated into the non-degradable polyolefin. According to Pavlath and Robertson [6], the protein-based natural polymer is more easily biodegraded compared to polysaccharide one.

Both protein and polysaccharides are hydrophilic and not compatible with hydrophobic polyolefin. Therefore, a compatibilizer is needed to improve the interfacial adhesion of different polar polymers. In this work, linear low density polyethylene (LLDPE), as a non-degradable polyolefin, was blended with a protein-based natural polymer, soya powder. In our previous investigation [7], epoxidized natural rubber with 50% mol epoxidation (ENR 50) was used for compatibilization of the blends. The tensile and thermal properties were improved with the addition of ENR 50. In the current investigation, the compatibilized blends were exposed to natural weathering for 1 year. Natural weathering was chosen as that is the condition closest to the real environment of disposed plastic. Analysis of the degradation which included Fourier transform infrared (FTIR) spectrometry, tensile properties, thermal properties, and molecular weight change, was carried out to investigate the compatibilized LLDPE/soya powder blends after outdoor exposure.

2 Experimental

2.1 Materials and sample preparation

The materials used in this study included LLDPE (ETILINAS LL0209SA) supplied by Polyethylene Malaysia Sdn Bhd, Terengganu, Malaysia. The melt flow index was 0.90 g/10 min and the density was 0.921 g/cm^3. Soya powder with a melt flow index of 1.0 g/10 min was supplied by Hasrat Bestari (M) Sdn Bhd, Penang, Malaysia. The average granular size was 12 µm and the protein content was 44.2%.

The mixing process was carried out using the melt blending method in a Haake Reodrive 5000 internal mixer. LLDPE was charged to the chamber for 2 min and followed by the addition of ENR 50. The soya powder was added gradually from 4 to 6 min and mixing was continued until the 10th min. The operating temperature of the internal mixer was maintained at 150°C with a rotor speed of 50 rpm. Table 1 shows the composition of the blends. Then, LLDPE/soya powder blends were molded into 1 mm thin sheets in a hot press at 150°C. The samples were cut into dumbbell shapes according to ISO 527, before being exposed to the natural environment.

2.2 Natural weathering

The natural weathering test was carried out at Universiti Sains Malaysia, Penang for a period of 1 year, from June 2011 to May 2012. The meteorology data, such as average temperature, rainfall, and relative humidity, were obtained from the nearest meteorology station in Butterworth (latitude 5°28'N, longitude 100°23'E). Table 2 shows the data obtained from the meteorology station; an average of the data for each month was taken. The samples were collected after 3 months, 6 months and 1 year. The test was carried out according to ISO 877.2. The samples in a dumbbell shape were placed on an aluminum alloy exposure rack facing south and at an inclination angle of 45°. The weathered samples were washed with distilled water, dried, and weighed until a constant weight in an air-drying oven at 70°C.

Table 1 Composition of linear low density polyethylene (LLDPE)/soya powder blends.

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

50% of ENR 50 based on soya powder content was used as compattibilizer.

Table 2 Meteorology data collected from Butterworth, Malaysia meteorology station.

Months	Mean max. temperature (°C)	Mean min. temperature (°C)	Humidity (%)	Rainfall (mm)
June 2011	32.6	24.6	75.9	3.0
July 2011	31.8	24.1	77.3	9.5
Aug 2011	30.7	24.0	84.8	20.3
Sept 2011	31.1	24.5	82.3	8.2
Oct 2011	30.6	24.3	83.0	6.4
Nov 2011	30.5	24	81.0	8
Dec 2011	31.6	23.9	78.0	2.74
Jan 2012	32.1	23.8	77.5	2.09
Feb 2012	33.2	24.9	74.9	4.04
Mar 2012	33.5	25.3	76.4	3.25
Apr 2012	30.6	24.3	80.6	7.1
May 2012	30.5	24.0	78.9	2.59

2.3 Investigation method of degradation

2.3.1 FTIR analysis

The FTIR analysis was carried out using an FTIR spectrometer (Perkin-Elmer model Series 2). For each spectrum, 32 consecutive scans with 4 cm^{-1} resolution were applied. The scanning range was 4000–400 cm^{-1}. Thin sample sheets with a 1 mm thickness were tested according to the attenuated reflection method. The carbonyl index (CI) was used as a parameter to observe the degree of degradation of the LLDPE/soya powder blends. CI was calculated according to the baseline method, i.e., the ratio of absorption bands at 1710–1740 cm^{-1} and 2844 cm^{-1}.

2.3.2 Tensile properties

The measurement of tensile properties, such as tensile strength, elongation at break (E_b) and Young's modulus were performed in an Instron Universal Testing Machine (Instron 3366) according to ASTM D638. Dumbbell shape samples were cut from each blends sheet. Five samples of each composition were strained at a rate of 50 mm min^{-1} at room temperature, and the average value of measurement was taken. The retention of these properties was calculated using Eq. (1):

$$\text{retention } (\%) = \frac{\text{Value after Degradation}}{\text{Value before Degradation}} \times 100\% \quad (1)$$

2.3.3 Surface morphology

The weathered samples were imaged on a scanning electron microscope (VPFESEM) model SUPRA 35VP with a voltage of 10 kV. The samples were conductively coated with gold, to prevent the accumulation of static electric charge during scanning. Images used to assess weathered surface were acquired at a magnification of 500×.

2.3.4 Differential scanning calorimetry (DSC)

Thermal analysis of weathered LLDPE/soya powder blends was carried out using a Perkin-Elmer DSC 7 thermal analyzer equipped with a liquid nitrogen cooling system. The preparation and parameters of the DSC tests were based on ASTM D3418-03 under nitrogen atmosphere. Samples (5–10 mg) were encapsulated in aluminum pans and subjected to thermal cycles. The samples were first heated to 175°C to remove the heat history. They were cooled to room temperature at a constant cooling rate of 10°C/min to favor crystallization. Then, the second heating was run at 10°C/min in a temperature range of 30–175°C. The heat of fusion was calculated by integrating the areas under the endothermic curves. The percentage of crystallinity of the LLDPE phase was calculated using Eq. (2):

$$\%\text{crystallinity} = \frac{\Delta H_f^*}{\Delta H_f^o} \times 100\% \quad (2)$$

where ΔH_f^o is the heat of fusion for 100% crystalline polyethylene and ΔH_f^* is the heat of fusion for semicrystalline LLDPE.

2.3.5 Weight loss

The weathered samples were rinsed with distilled water and dried to a constant temperature at 70°C. The weight loss percentage was calculated with the following equation:

$$\%\text{Weight Loss} = \frac{(W_i - W_f)}{W_i} \times 100 \quad (3)$$

where W_i and W_f denote the initial weight and final weight of the samples, respectively.

2.3.6 Molecular weight changes

The molecular weight of the sample was determined using gel permeation chromatography (GPC).The GPC was performed at 140°C using an Agilent 1200 GPC system connected to a Shodex K-806 and K-802 column. Chloroform was used as the solvent, with a flow rate of 0.80 ml/min. The system was calibrated using a polystyrene standard with an average molecular weight ranging from 1000 to 5,000,000. The blends were dissolved in chloroform at a temperature of 40°C for 1 week. Subsequently, 50 µl samples were filtered through a 0.45 µm polytetrafluoroethylene filter to remove contaminants and solid particles. The number average molecular weight (Mn) and weight average molecular weight (Mw) were measured.

3 Results and discussion

3.1 Chemical structure changes and CIs

FTIR spectroscopy analysis is a good indicator for the chemical changes in polymer molecules after natural weathering. Figure 1 presents the FTIR spectra of ENR 50 compatibilized blends after 6 months and 1 year natural weathering. The changes of peak can be seen at the broad peak of 1740 cm^{-1}, indicating the presence of a carbonyl group. Khabbaz et al. [8] studied the environmental degradation of polyethylene film and found that many carbonyl compounds, such as esters and carboxylic acids, existed after exposing to the outdoor environment. The carbonyl groups, such as carbon monoxide and methyl vinyl ketone, were generated through the Norrish type I and II mechanism (Figure 2). Therefore, the broad peak at 1740 cm^{-1} indicated the overlapping between the ester bonds during compatibilization [7] and carbonyl products generated after the weathering test. Table 3 summarizes the comparison of CIs between uncompatibilized and ENR 50 compatibilized blends. The chemical changes for uncompatibilized blends after outdoor exposure has been discussed in our previous study [9]. The results (Table 3) show that the CIs of LLDPE/soya powder blends increased after 6 months and 1 year of natural weathering. It was confirmed that the carbonyl product increased depending on the level of degradation. The CI for compatibilized blends was higher than that for uncompatibilized blends. Thus, ENR 50 was not only acting as a compatibilizer, but accelerating the abiotic degradation. This might be due to the elastomer, i.e., ENR 50, which generates free radicals in the presence of sunlight during weathering.

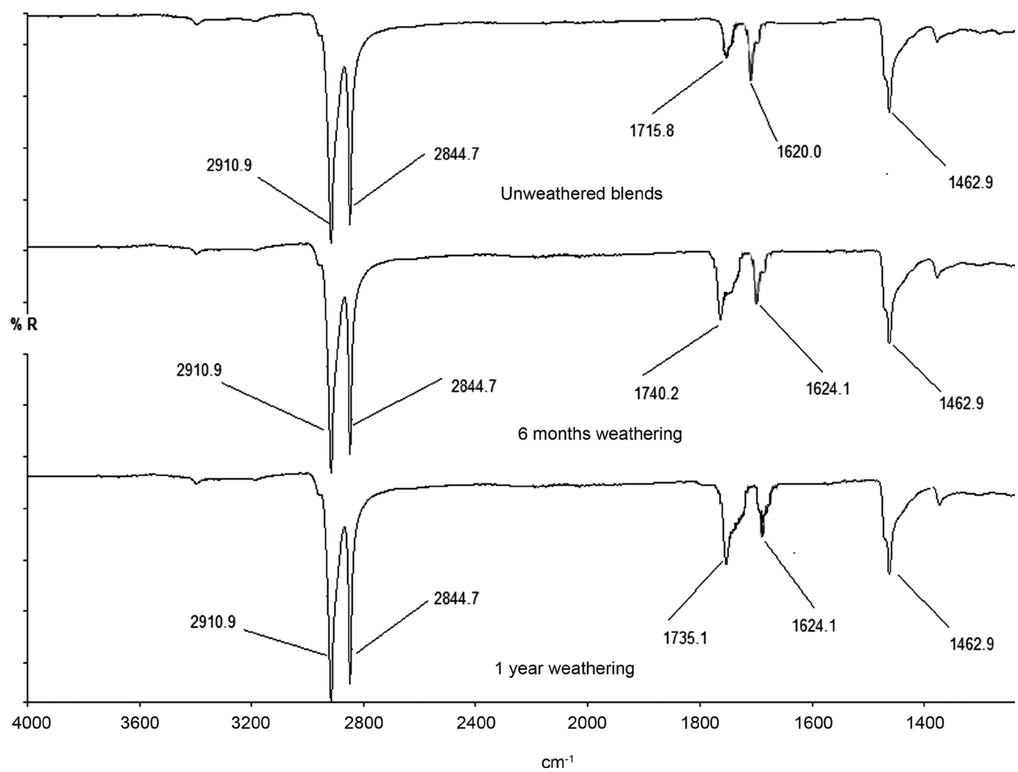

Figure 1 Fourier transform infrared (FTIR) spectra of unweathered and weathered sample for ENR 50 compatibilized linear low density polyethylene (LLDPE)/soya powder blends.

It subsequently initiated oxidation of LLDPE according to the typical oxidation scheme for polyolefins [10]. Furthermore, the elastomeric phase is the most oxidizable component, as it contains unsaturated bonds. The higher degradation of ENR 50 compatibilized blends can be further proven in tensile properties.

3.2 Tensile properties

Figure 3 shows the tensile strength of ENR 50 compatibilized LLDPE/soya powder blends over 1 year of natural weathering. It can be observed that the tensile strength reduced with increasing soya powder content, indicating

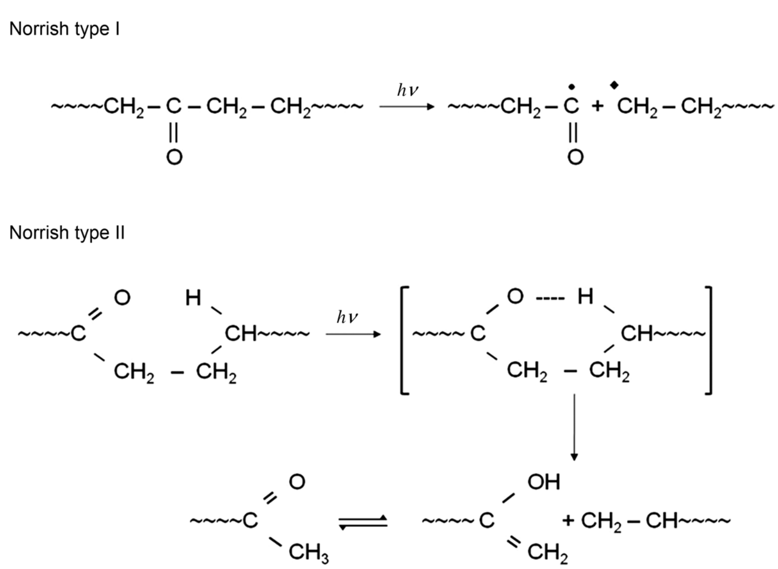

Figure 2 Schematic diagram for the formation of carbonyl groups through the Norrish Type 1 and Norrish Type II mechanisms.

Table 3 Carbonyl index of uncompatibilized and compatibilized blends for different natural weathering periods.

Blends composition	Uncompatibilized blends (carbonyl index%)		Compatibilized blends (carbonyl index%)	
	6 months	1 year	6 months	1 year
LLDPE 95/soya powder 5	13.55	20.34	15.66	22.1
LLDPE 80/soya powder 20	23.11	31.55	29.44	33.8
LLDPE 60/soya powder 40	30.87	40.58	36.21	42.9

LLDPE, linear low density polyethylene.

an increase in degradability. After 3 months of weathering, the tensile strength was generally reduced as a function of soya powder content. However, a fragmentation process occurred on the blends with 40% soya powder content. The samples were fragmented and unable to be subjected to tensile test. Based on our previous study [9], soya powder is a highly hydrophilic natural polymer and it can be easily leached out during outdoor exposure. Therefore, the remaining LLDPE can be broken down to smaller segments, by an external force like raining during the weathering test. After 1 year exposure time, the degradation effect was more pronounced as the blends with 20–40 wt% soya powder contents were fragmented. Table 4 illustrates the retention of tensile properties of uncompatibilized and compatibilized blends after weathering. The retention value can further explain the degradation effect as a function of soya powder content and exposure duration. The reduction of tensile strength retention with increasing soya powder content has confirmed that the addition of soya powder can accelerate the degradation process.

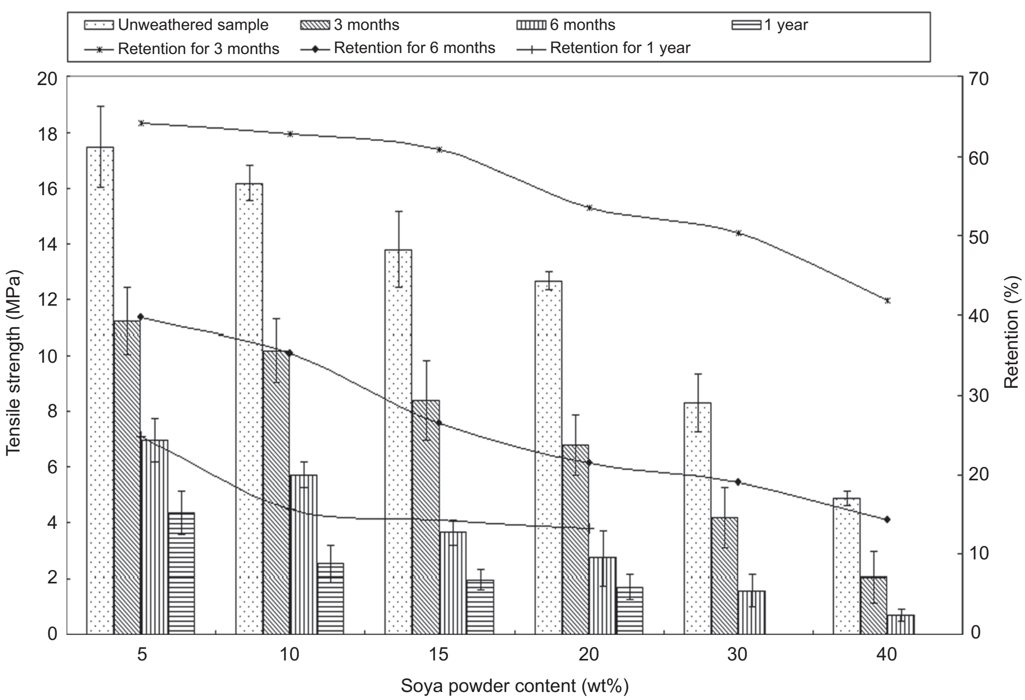

Figure 3 Tensile strength and retention of compatibilized blends after different periods of weathering.

Table 4 Retention of tensile properties for non-irradiated and irradiated blends after 1 year outdoor exposure.

Sample	Retention of uncompatibilized blends (%)			Retention compatibilized blends (%)		
	Tensile strength	Elongation at break	Young's modulus	Tensile strength	Elongation at break	Young's modulus
LLDPE/5 soya powder	24.82	7.14	134.81	32.04	9.70	128.08
LLDPE/20 soya powder	13.31	1.61	166.49	14.63	2.28	176.13
LLDPE/40 soya powder	Fragmented	Fragmented	Fragmented	Fragmented	Fragmented	Fragmented

LLDPE, linear low density polyethylene.

The E_b of blends during 1 year of natural weathering is shown in Figure 4. The trend of E_b is similar to that of the tensile strength (Figure 3). The reduction of the E_b is due to the abiotic and biotic effect from the environment. The soya powder leached out and left some pores, consequently created a bigger surface area for further degradation. The mechanism of the degradation of the blends can be explained by a morphological study. Figures 5 A–C and 6 A–C show the morphology of the surface of weathered blends after 6 months and 1 year of natural weathering, respectively. Increasing the soya powder content in the blends created more pores on the weathered surface. The size of the pores is even bigger at high soya powder content. This allows microorganisms, such as fungus, to occupy and consume the polymer. The degradation was more critical after 1 year of natural weathering (see Figure 6). As the exposure period increased, more fungus colonized on the surface of the samples and larger pores were observed. In uncompatibilized blends [9], there were fewer pores and the pore size was smaller compared to ENR 50 compatibilized blends. This can be further confirmed by the E_b retention (Table 4). The retention for compatibilized blends was also lower than for uncompatibilized blends. Apart from the fungus colonization, the pores also underwent photooxidation and thermal oxidation. The creation of carbonyl products, as discussed earlier, has indicated these degradation processes.

The Young's modulus of the blends was generally increased after 1 year of natural weathering (Figure 7). The increment of the Young's modulus might be due to the formation of radical crosslinking in the presence of sunlight. The formation of crosslinking was confirmed by the carbonyl product generated, as shown in FTIR spectra (Figure 1). The embrittlement of the sample also resulted in the increment of Young's modulus. The trend is in agreement with the reduction of E_b after weathering. The blends with a high soya powder content (20–40 wt%) were too brittle and fragmented upon outdoor exposure for 1 year. As shown in Table 4, the retention of Young's modulus of compatibilized blends was lower than that of uncompatibilized blends. This was because during the weathering test, the leach out effect of compatibilized blends was higher and the weathered samples lost rigidity. Thus, the leach out effect in compatibilized blends was more pronounced compared to uncompatibilized blends. This is in agreement with the superior degradability of ENR 50 compatibilized blends.

3.3 Crystallinity study

The crystallinity change upon environmental exposure is one of the indicators for degradation. Figures 8 and 9 show the melting and cooling thermograms of the blends after 1 year of natural weathering. The melting temperature (T_m) did not significantly change after weathering.

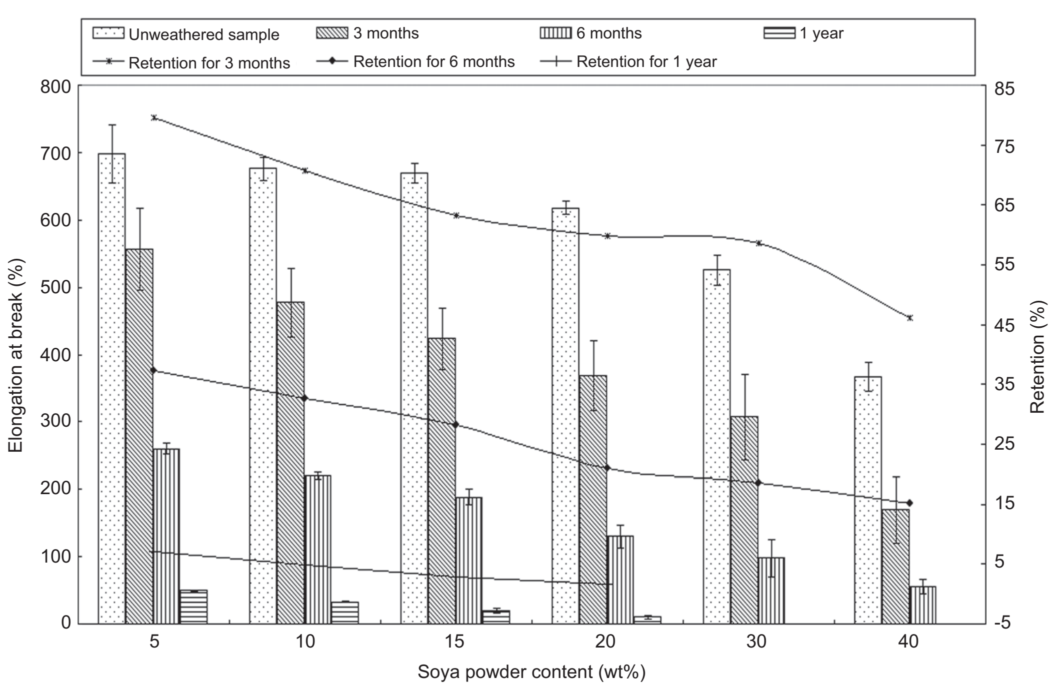

Figure 4 Elongation at break and retention of compatibilized blends after different periods of weathering.

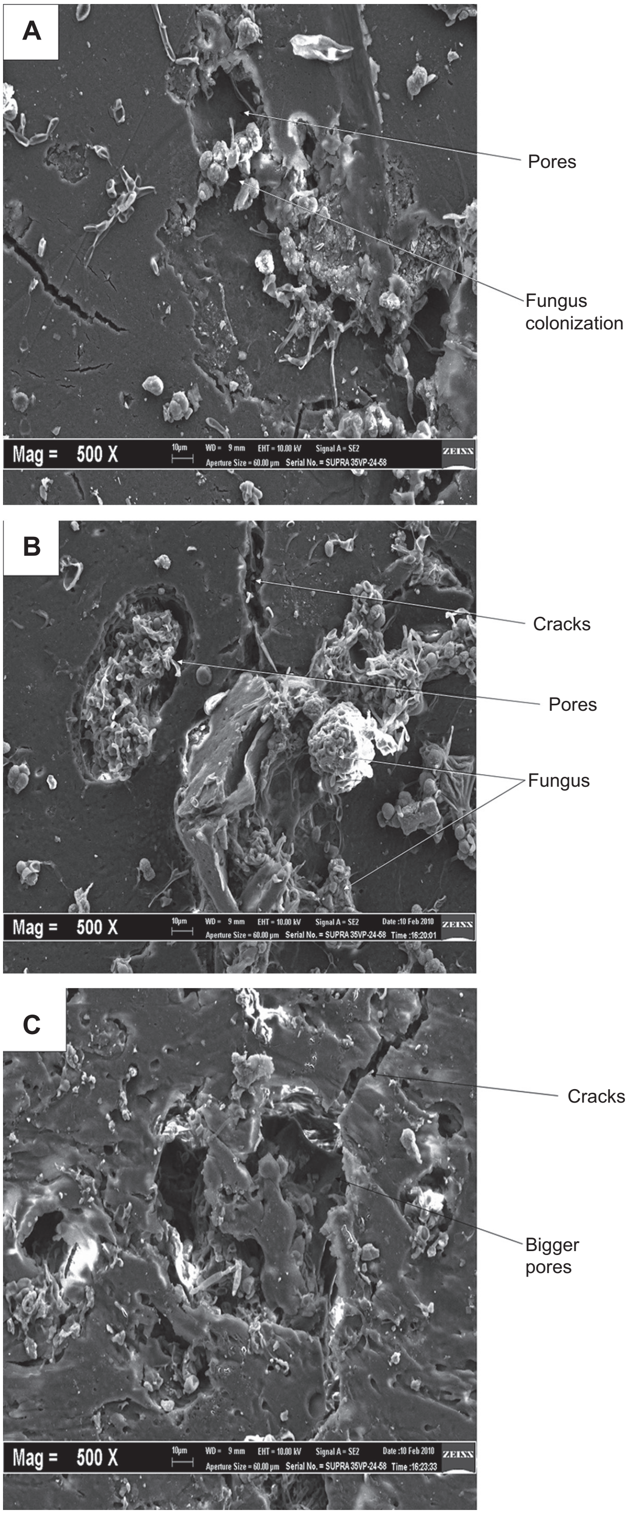

Figure 5 Weathered surface (500× magnification) of compatibilized blends with (A) 5 wt%, (B) 20 wt%, and (C) 40 wt% soya powder content after 6 months weathering.

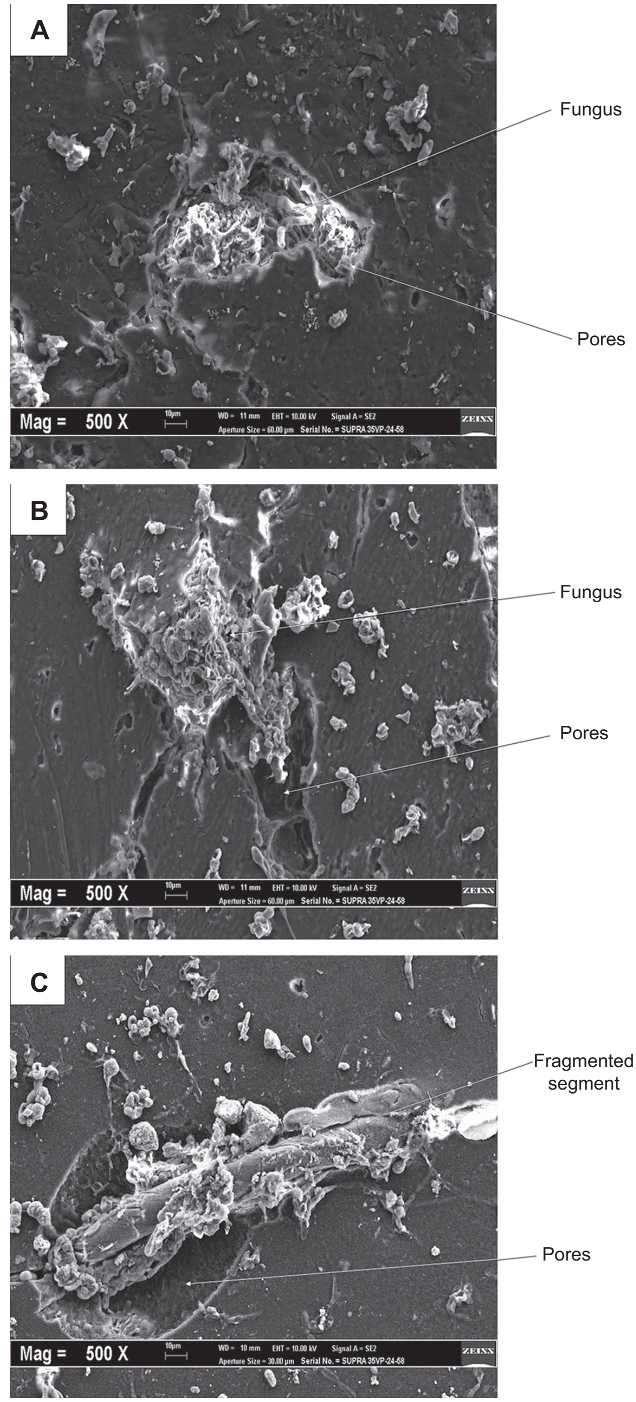

Figure 6 Weathered surface (500× magnification) of compatibilized blends with (A) 5 wt%, (B) 20 wt%, and (C) 40 wt% soya powder content after 1 year weathering.

This confirmed that there was no new crystalline formed during degradation. The DSC data for the uncompatibilized and compatibilized blends after outdoor exposure is summarized in Table 5. The crystalline temperature (T_c) and crystallinity increased as a function of the weathering period, corresponding to a degradation of the amorphous phase in the blends. According to Khabbaz et al. [8], the amorphous phase in the polymer was first attacked during thermal or oxidative degradation, consequently increasing

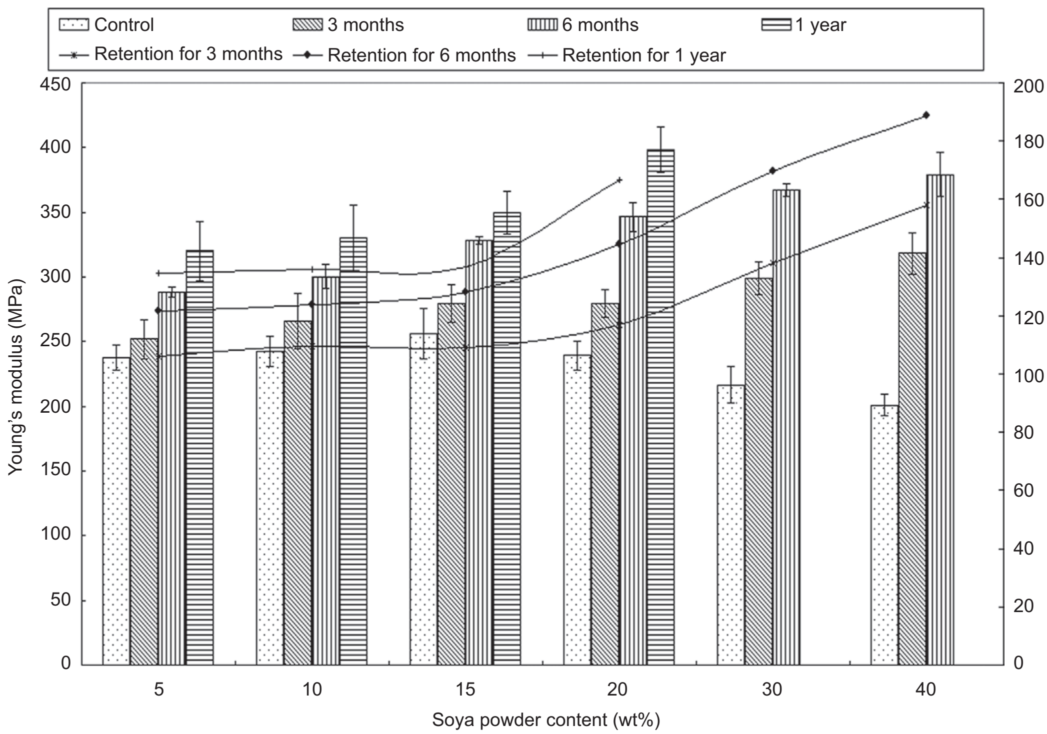

Figure 7 Young's modulus and retention of compatibilized blends after different period of weathering.

the portion of the crystalline phase in the remaining blends. Therefore, the increase in crystallinity of weathered samples was regardless of new crystalline formation and was solely contributed to by the reduction of the amorphous phase. Soya powder content in the blends played an important role in contributing to the degradation. At the same exposure period, the blends with higher soya powder content exhibited higher crystallinity. This agreed with the study by Lodha and Netravali [11], in which the crystallinity of soya protein isolate resin increased after degradation. In comparison, the crystallinity of compatibilized blends was generally lower (see Table 5). The effect of ENR 50 in enhancing the degradation through the formation of radicals was discussed in a previous section.

3.4 Weight loss

Figure 10 shows the comparison of the weight loss of uncompatibilized and compatibilized blends during

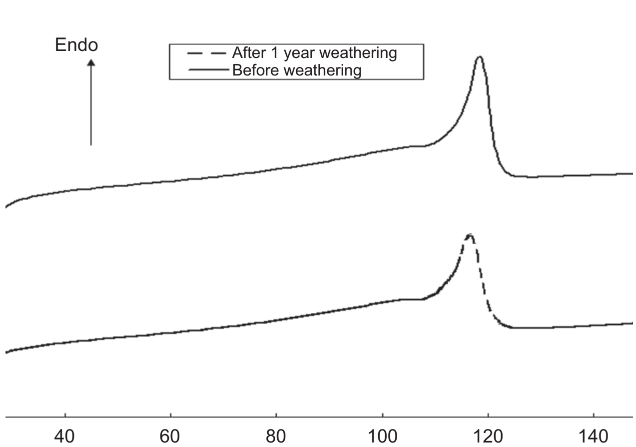

Figure 8 Melting thermogram of compatibilized blends before and after weathering.

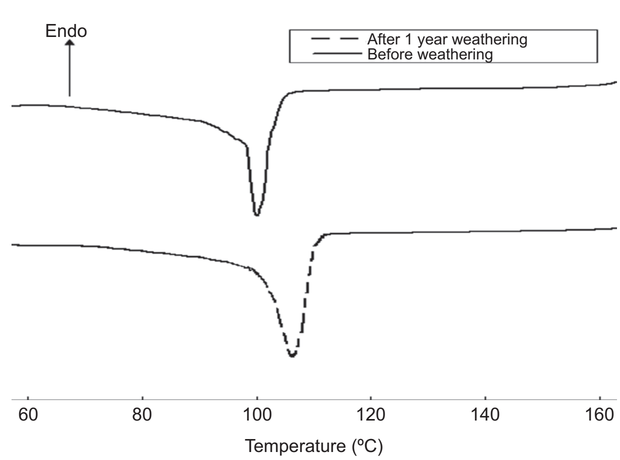

Figure 9 Cooling thermogram of compatibilized blends before and after weathering.

Table 5 Differential scanning calorimetry (DSC) results of uncompatibilized and compatibilized linear low density polyethylene (LLDPE)/soya powder blends after different period of weathering test.

Sample	T_m (°C)		T_c (°C)		ΔH_f*		Crystallinity (%)	
	6 months	1 year	6 months	1 year	6 months	1 year	6 months	1 year
95 LLDPE/5 soya powder	124.3	124.9	106.3	108.9	95.2	115.7	34.5	41.9
80 LLDPE/20 soya powder	123.7	123.5	107.4	109.4	108.7	121.6	39.4	44.1
70 LLDPE/40 soya powder	122.9	122.8	108.1	111.5	115.6	128.2	41.9	46.4
95 LLDPE/5 soya powder/ ENR 50	121.3	121.0	106.9	107.6	98.3	118.2	35.6	42.8
80 LLDPE/20 soya powder/ ENR 50	118.2	118.4	107.8	108.7	101.7	122.6	39.4	44.4
70 LLDPE/40 soya powder/ ENR 50	117.1	117.0	109.3	110.2	104.1	127.1	39.9	46.0

ΔH_f*, heat of fusion for semicrystalline LLDPE.

different periods of natural weathering. The weight loss of compatibilized blends was higher than that of uncompatibilized blends. It can be observed that the weight loss increased dramatically from 30 to 40 wt% soya powder, because the interaction between LLDPE and soya powder became weaker. Consequently, it was much easier for the soya powder to be leached out during the degradation. As mentioned in a previous section, the degradability of compatibilized blends was higher. Therefore, the number of fragmented segments for compatibilized blends was higher and subsequently leached out from the weathered samples. The presence of pores on compatibilized blends indicated the leached out effect. Apart from the abiotic effect, the biotic factor might result in the weight loss of the blends. The existing microorganism such as fungus in the environment can occupy the sample surface and consume the soya powder phase. The colonization of fungus on the sample surface is shown in Figures 5 and 6. The consumption of the soya powder also contributed to the sample weight loss. The results are in agreement with some weight loss studies in LDPE/starch blends [12, 13], in which the starch is a highly hydrophilic material.

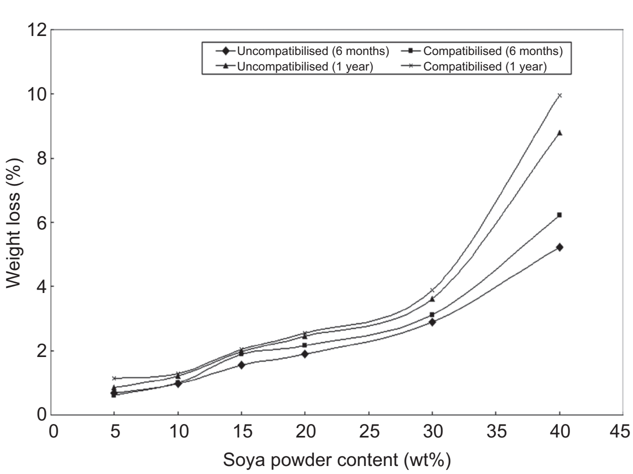

Figure 10 Comparison of weight loss for uncompatibilized and compatibilized blends after weathering.

3.5 Molecular weight change

The weight loss study was the gravimetric analysis of the weathered blends. The molecular weight change was used to analyze the degradation of sole LLDPE. It is claimed that in polyolefins/natural polymer blends, only natural polymers can be degraded or consumed by microorganisms [14]. However, the molecular weight of LLDPE/soya powder showed changes after natural weathering in the current study. Table 6 summarizes the Mn and Mw of uncompatibilized and compatibilized blends. Before weathering, the Mn and Mw of LLDPE were 27.7×10^3 Da and 97×10^3 Da, respectively. The compatibilized LLDPE phase in the blends was the same with the uncompatibilized blends, because there were no co-polymerization and chain breaks during the compatibilization process. In both the uncompatibilized and compatibilized blends, the molecular weight decreased with increasing soya powder content. The results again explained the leach out of soya which created a bigger surface area for the abiotic degradation, such as UV and thermal degradation. In ENR 50

Table 6 Comparison of molecular weight for uncompatibilized and compatibilized linear low density polyethylene (LLDPE)/soya powder blends after 1 year natural weathering.

Sample	Uncompatibilized blends		Compatibilized blends	
	Mn (10^3)	Mw (10^3)	Mn (10^3)	Mw (10^3)
LLDPE	25.1	93.1	19.2	66.4
LLDPE/5 soya powder	17.5	55.4	13.6	42.9
LLDPE/20 soya powder	13.5	47.1	10.2	34.2
LLDPE/40 soya powder	9.2	31.9	5.9	24.6

compatibilized blends, the reduction of the molecular weight was greater than in the uncompatibilized blends. This may be due to the fact that the double bond in ENR 50 was more susceptible to photo and thermal degradation. Consequently, the formation of a peroxide radical further attacks the molecular chain of LLDPE.

4 Conclusion

In this study, ENR 50 was used to compatibilize LLDPE and soya powder. Abiotic and biotic degradation during natural weathering was proved during this study. The CI increased with increase in soya powder content and exposure period. The tensile strength and Eb decreased with increasing soya powder content upon exposure. However, the Young's modulus increased. The reduction of crystallinity indicates that the reduction of the amorphous phase resulted from degradation. By contrast, the weight loss of the blends increased as a function of soya powder content and exposure time. The reduction of molecular weight upon outdoor exposure indicates the chain scission of the LLDPE phase during degradation. Generally, the addition of soya powder improved the degradability of LLDPE. In comparison, the ENR 50 compatibilized blends showed better degradability compared to uncompatibilized blends.

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