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ORIGINAL PAPER

LDPE and poly(1-butene) blends: Morphology, crystallinity and rheological properties

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ABSTRACT

In this work, the compatibility and crystallinity of low density polyethylene (LDPE) and polybutene-1 (PB-1) blends were studied. Various blends of LDPE/PB-1 containing 5, 10 and 20 wt.% PB-1 were prepared in a co-rotating twin-screw extruder and characterized by scanning electron microscopy (SEM), shear oscillation rheology and wide-angle X-ray diffractometry (WAXD). A matrix-droplet morphology was observed in SEM images, indicating incompatibility of the two polymers in the solid state. Compared to neat LDPE, the relaxation spectra of the blends were broadened, and a slight increase in their relaxation times was observed. The relaxation time of the blends was enhanced by increasing PB-1 content, which was further proved by fitting rheological data in the Carreau-Yasuda model. Deviation of Cole-Cole diagrams from circular shape means that the blend samples were not miscible and the positive-deviation behavior of the complex viscosity and storage modulus from the mixing rule revealed the formation of strong interfacial interactions. The crystallinity of both LLDPE and PB-1 was decreased as a result of blending. The peaks attributed to the form II of PB-1 crystals were eliminated and the peaks related to LDPE were obviously weakened, suggesting to prevent crystallinity of polymers which is associated with a reduction in the total crystallinity percentage for the blend. The reduction of crystallinity was more pronounced in PB-1 phase. **Polyolefins J (2021) 8: 21-30**

Keywords: Blending; morphology; LDPE; crystallinity; PB-1.

INTRODUCTION

Low density polyethylene (LDPE) is a well-known polyolefin which is widely consumed in various industrial applications, especially packaging [1,2]. Poly 1-butene (PB-1), which has received much attention in the last few decades, is a semi-crystalline linear polymer with various crystal forms that is capable of crystallization via chain-folding [3, 4] and high resistance against creep and stress cracking [5,6]. Generally, blending is a method for changing the properties of polymers and achieving new properties not seen in single materials. PB-1 is usually incompatible with other polyolefins, resulting in the formation of very weak intermolecular bonds which lead to phase separation of polymers during extrusion [7,8]. This phase separation and immiscibility are necessary factors to achieve acceptable peeling properties [9] in peelable blends or

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coatings of PB-1 with LDPE, HDPE and LLDPE [10]. In fact, poor adhesion to the PB-1 and LDPE interface causes them to separate during crystallization due to their incompatibility, and the interface can act as a breaking zone, resulting in the production of easily opened peel packaging films [11, 12]. Thin films of LDPE and isotactic PB-1 blends, usually produced by a blowing process, are often used for peelable films in the food and household industries or in the medical sector [13, 14]. A concentration of 20 wt.% PB-1 of LDPE/PB-1 blends has been reported to be the upper limit of application as a peeling system [9]. These polymers are reported to be immiscible in both macroscopic and microscopic levels [12]. Since the chemical structure of LDPE and PB-1 are so similar to each other, both are composed only of C-H and C-C bonds [7,15], it is difficult to distinguish them based on their structure. Hence, investigation of melt rheology and state of crystallization can be useful to achieve a better understanding of these systems. Both LDPE and PB-1 [16] are polymorphic polymers and the crystalline region can adopt various structures. The studies on PE/PB-1 blends have shown that each phase crystallizes independently [17-19] as a reason of thermodynamic immiscibility of the components [9]. The physical structure of the film is controlled by both chemical and thermodynamic characteristics of the blend components such as viscosity and interfacial interactions [20], as well as processing parameters including mixing and melting order, stress and flow history [20,21]. In this study, LDPE and PB-1 were melt-blended to obtain a clear insight of morphological changes, rheological properties and crystallinity of components due to blending, so that compatibility and immiscibility could be correlated to crystallization.

EXPERIMENTAL

Materials

LDPE (LF 0200) (density 0.920 gr/cm^3) used in this study was supplied by Bandar Imam Petrochemical Company, Iran. Homopolymer PB-1 (PB 0110M) (density 0.914 gr/cm^3) was provided by Lyondell Basell Polyolefin, Germany. The melt flow rate (MFR) of LDPE and PB-1 were 2.0 and 0.4 g/10min (190°C/2.16kg), respectively.

Sample preparation

Neat LDPE and PB-1 samples as well as various blends containing 5, 10 and 20 wt.% PB-1 were prepared via passing through a co-rotating twin-screw lab extruder (L/D 40) Brabender Company, Germany. The mixing was carried out at 30 rpm and a barrel temperature profile of 130-180°C from the hopper to the die, in the presence of 1000 ppm Irganox 1010 (BASF, Germany) antioxidant to prevent thermo-oxidative degradation of the polymers. The extrudates were immediately quenched in a water bath at the die exit and then chopped as pellets. The prepared samples are listed in Table 1.

Characterization

Morphology

The morphology of the blends was studied by scanning electron microscopy (SEM; VEGA\\ESCAN, Czech Republic). The samples were first formed by hot pressing at 190°C into thin sheets 1 mm thick, followed by broken down into liquid nitrogen and finally kept immersed in cyclohexane solvent for 30 min at 70°C to extract PB-1 from the fractured surface. According to the known melting temperature of neat polymers (approximately 110°C for LDPE and 115°C for PB-1), elevated temperatures were avoided and a temperature of 70°C for etching was selected to preserve the morphology of polymers [22]. The specimens were then washed with fresh cyclohexane, dried, and gold-sputtered prior to the SEM testing.

Rheological measurements

Rheological measurements were performed by a rheometric mechanical spectrometer type MCR 300 made by Physica Anton Paar Company, Austria, in oscillation mode, on 20 mm diameter discs obtained from

Table 1. Composition and coding of the samples.

Sample Code	Composition	
	LDPE (wt.%)	PB-1 (wt.%)
LDPE	100	0
PB-1	0	100
LP5	95	5
LP10	90	10
LP20	80	20

hot pressing of samples at 190°C for 5 min. Strain sweep test was carried out to characterize the viscoelastic range of deformations for further investigations and a strain amplitude of 10% was obtained for the viscoelastic range of LDPE/PB-1 blend. Rheological measurements were performed in the linear viscoelastic region. All samples were evaluated in an angular frequency range of 0.03-600 rad/s under nitrogen atmosphere.

Wide angle X-ray diffraction (WAXD)

Crystallinity percentage (X_C) and crystal structure were investigated by a D5000 type XRD instrument by SIEMENS Company, Germany. Sheets 2 mm thick were prepared by hot pressing followed by annealing at 70°C and then subjected to measurements at ambient temperature, relative humidity of 30% and in the range of 5-70° for 2Θ. A cobalt anode was employed in this test.

RESULTS AND DISCUSSION

Morphological analysis

SEM micrographs of the prepared blends are shown in Figure 1. A droplet-matrix morphology was observed which indicates the incompatible nature of the blends in the solid state. The wide particle size distribution implies the tendency of PB-1 chains to agglomerate rather than disperse in the polyethylene matrix [23]. Image J analysis software was employed to identify the number average radius ($R_n = \frac{\sum_i n_i R_i}{\sum_i n_i}$) and volume average radius ($R_v = \frac{\sum_i n_i R_i^4}{\sum_i n_i R_i^3}$) of the particles [24, 25], where R_i is the radius of each droplet and n_i is the number of particles with the radius R_i, and the results are tabulated in Table 2.

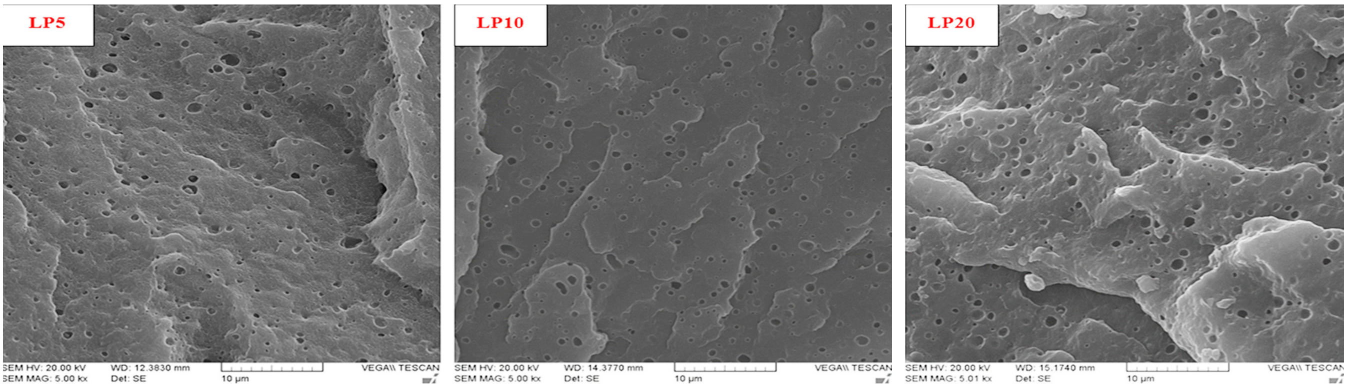
Table 2. (R_n) and (R_V) values for the prepared blends according to SEM images.

Sample	R_n (μm) ± 0.001	R_V (μm) ± 0.001	R_V/R_n
LP1	0.299	0.571	1.91
LP2	0.294	0.514	1.75
LP3	0.310	0.579	1.87

The ratio R_V/R_n stands for polydispersity and is a criterion for fine dispersion of the particles. The closer this ratio is to unity, the finer the morphology [23]. According to the calculations, the size of particles has been observed to increase with the PB-1 content. No significant difference was observed between the particle size of LP10 and LP5 samples. The increment observed in particle size of the sample LP20 in comparison with the sample LP10 is attributed to the demixing of PB-1 phase at higher concentrations, resulting in formation of larger particles. Indeed, with increasing concentration of PB-1 (minor phase), the particle-particle coalescence phenomenon becomes stronger than the interaction between PB-1 dispersed particles and the LDPE matrix [26].

Rheological behavior

It is well-known that the rheological behavior of polymer blends is affected by their morphology, which is specifically dependent on the blend composition. The complex viscosity of neat LDPE, neat PB-1 and their blends has been shown in Figure 2. The deviation of the neat LDPE sample from the shear-thinning rheo-



(a) (b) (c)

Figure 1. Morphology of the prepared blend samples.

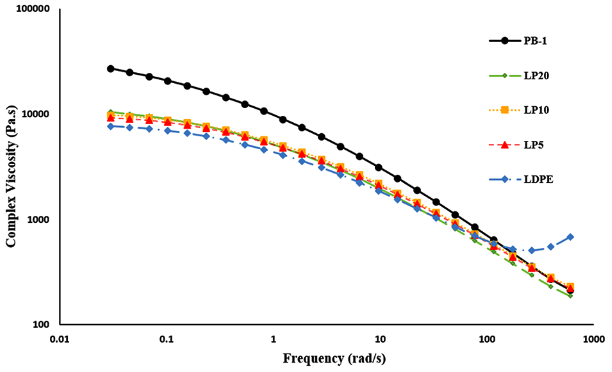


Figure 2. Complex viscosity of neat polymers and their blends versus frequency at 190°C.

logical behavior at high frequencies is due to its higher response time compared to the test observation time after sample stimulating, which is caused by strong entanglements between LDPE chains, resulting in a deviation from shear-thinning rheological behavior at high frequencies (short observation times). As can be seen, the complex viscosity curves of the blends are between those of virgin components. All blend compositions (samples LP5, LP10 and LP20) have shown Newtonian plateau at low frequencies which is transformed to shear-thinning behavior at high frequencies. In addition, it can be seen that at low frequencies, the complex viscosity is enhanced by increasing the content of PB-1 in the blends. The overlap observed at high frequencies in the complex viscosity curves of the samples can be the result of equivalent segmental relaxation behavior [27] as well as the stronger shear thinning behavior of PB-1. Several authors [28-30] in their studies on polymeric blends have shown that the deviation of complex viscosity from the "mixture rule" [31] (Equation 1) at low frequencies can be attributed to the compatibility of the blend.

$$\log(\eta_{blend}) = X_A \log(\eta_A) + X_B \log(\eta_B) \tag{1}$$

In which, X_A and X_B are the weight fractions of polymers A and B in the blend, respectively; and η_A and η_B are the complex viscosities of polymers A and B. Also, the complex viscosity of the blend is termed as η_{blend}.

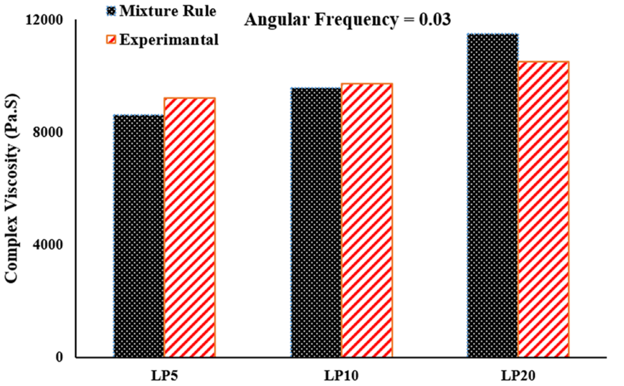


Figure 3. A comparison between theoretical (mixture rule) and experimental complex viscosity of the prepared blend samples (at 190°C and ω=0.03).

In Figure 3, the complex viscosity of the blend samples has been compared with that of the mixture rule. A positive-deviation behavior from the mixture rule has been observed in the complex viscosity of samples LP5 and LP10, confirming the compatible nature of the blends. However, the negative deviation of the sample LP20 from the mixture rule may be related to the larger droplets formed via demixing of the PB-1 phase, which is related to the results of polydispersity given in Table 2. By increasing the PB-1 content in the sample LP20, particle-particle coalescence of PB-1 dispersed particles results in the formation of larger particles. As a result, the interface and interactions between the LDPE and PB-1 phases are reduced, and the viscosity of the blend becomes less than those is expected by the mixture rule. It can be said that the compatibility of the blend is decreased by increasing the PB-1 content. Since the individual phases may behave differently, immiscible blends do not follow the mixture rule and their behavior may be complex [32]. The weighted relaxation spectra of the samples, H(λ), were plotted against characteristic relaxation time, λ, and illustrated in Figure 4. The relaxation spectrum was determined using Tschoegl approximation [32]. This method is more general than the Laplace method

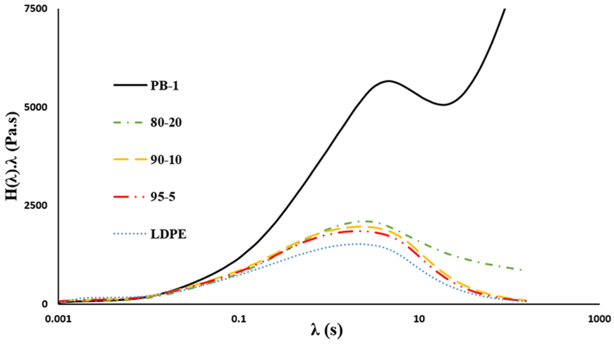


Figure 4. Weighted relaxation spectra of neat polymers and the prepared samples at 190°C.

and approximates the spectral distribution functions by logarithmic differentiation of the step response, resulting in a bell-shaped relaxation spectrum [34]. According to Figure 4, neat PB-1 contains a component with a long relaxation time that cannot be completely detected in the frequency range of the experiments. On the other hand, the relaxation spectra of the blend samples are broader than that of neat LDPE, indicating an increment in the molecular weight distribution due to the blending. All blend samples showed a relaxation time and their corresponding relaxation time was slightly enhanced in comparison with LDPE due to the presence of PB-1 chains in the system. In addition, with increasing PB-1 content in blend samples, the relaxation spectrum is somewhat broadened. The distinct tail appeared on the right-hand side of the spectrum in sample LP20 could be due to the shape relaxation of the dispersed phase droplet by the interfacial tension of heterogeneous structure [35], and also may represent the presence of a different phase morphology with different characteristic length scale and relaxation time being onset [36]. Moreover, this tail has only been observed in sample LP20 due to its higher PB-1 content compared to other samples. Since PB-1 contains a component with a long relaxation time, the relaxation of that component has appeared by increasing the PB-1 content to 20% in sample LP20 which may be related to its percolation threshold, while it was not detected in other blend samples possessing lesser PB-1 content.

The values of the zero shear viscosity (η_0), relaxation time (λ) and n index for the samples were obtained via fitting the rheological data in the Carreau-Yasuda model, and the results are tabulated in Table 3. It is worth mentioning that the η_0 value for each sample was obtained through extrapolation of shear viscosity values using the Carreau- Yasuda model (Equation 2) [37]:

$$\eta^*(\omega) = \frac{\eta_0}{(1+\lambda\omega^a)^{(n-1)/a}} \tag{2}$$

Table 3. Carreau-Yasuda model parameters for neat polymers and their blends.

Sample	η_0 (Pa. s) ± 100	n index ± 0.01	Relaxation time (s) ± 0.1
LDPE	9800	0.61	2.2
PB-1	41000	0.26	4.7
LP5	10500	0.39	2.4
LP10	11100	0.40	2.6
LP20	12600	0.36	2.7

According to the data given in Table 3, the relaxation time of the blends has enhanced by increasing the PB-1 content. It should be noted that by increasing the concentration of PB-1 to 10 wt.% (sample LP10), the shear thinning behavior was slightly raised while a less shear thinning behavior was observed by enhancement of the PB-1 content to 20 wt.% in sample LP20. The values of the relaxation time (λ) obtained by curve-fitting in the model are in good agreement with the relaxation spectra represented in Figure 4.

One effective way to investigate the compatibility and homogeneity of the polymeric blends is the Cole-Cole plot [38-40]. The presence of a single circular arc in the Cole-Cole curve is a sign of a homogeneous melt system, while the appearance of a shoulder or a second circular arc on the right-hand side of the curve denotes the existence of a second phase with longer relaxation time [41, 42]. It has been shown [43] that the shape of the Cole-Cole plots is affected by the particle size of the dispersed phase as well as the interfacial strength [41, 44, 45]. The Cole-Cole plots of neat LDPE, neat PB-1 and the prepared blends are shown in Figure 5. It has been observed that by increasing percentage or size of the dispersed phase (PB-1), the radii of the arcs in the Cole-Cole plots have enhanced and deviated from the circularity, suggesting that the systems have moved towards more incompatibility. The deviation of the sample LP20 from circularity is in good agreement with its relaxation spectrum, representing the presence of a different phase morphology [36].

Variations in the storage modulus (G') of all samples versus frequency at 190°C have been illustrated in

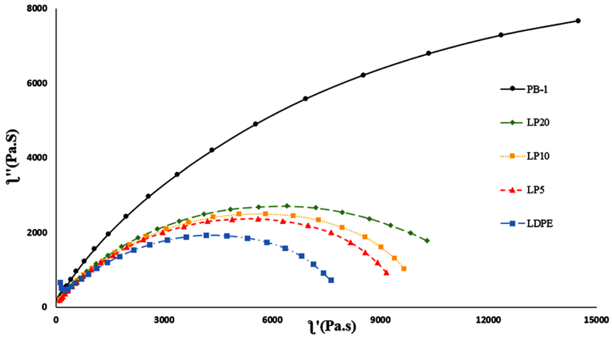


Figure 5. Cole-Cole plots of the prepared samples at 190°C.

Figure 6. The storage moduli of all blends have fallen between those of neat polymers. The theoretical G' values of the blend samples were calculated based on the mixture rule and compared with the experimental data, indicating a slight deviation from the theoretical storage modulus in the whole frequency range. This implies the formation of interactions between LDPE and PB-1 in the interface of the blend, resulting in deviation from the mixture rule [46]. Moreover, the increment in the storage modulus of the blend samples at low frequencies can be attributed to the heterogeneity of the system as well as the shape relaxation of the dispersed phase droplets by the interfacial tension [47, 48].

Crystallinity

Both LDPE and PB-1 are polymorphic polymers with various crystal cell forms that gradually transform to their stable forms [16]. Crystals of polyethylene exist in two forms of orthorhombic (stable) and hexagonal. On the other hand, crystals of PB-1 exist in at least four various forms [49] of I and I' (hexagonal), II (tetragonal) and III (orthorhombic). The most stable form for PB-1 is form I. Although the crystallization of PB-1 from the molten state results in the form II mesophase, it subsequently transforms to the stable form I [50]. The values of 2Θ (based on copper anode) and WAXD planes for various crystal forms of LDPE and PB-1 are summarized in Table 4, based on the literature [51-53].

WAXD was performed on samples at ambient temperature, 30% relative humidity and range 5 to 70° for 2Θ with an anode made of cobalt. Characteristic peaks obtained by cobalt anode differ from those given in the literature with respect to copper anodes. Hence, the results were corrected to allow comparison. Since the various blends in this study were indistinguishable from SEM micrographs and rheological measurements, and due to the close range of compositions in the prepared samples, the authors have only studied the crystallinity of the sample LP10 to monitor the effect of blending. WAXD patterns for the neat polymers and sample LP10 are demonstrated in Figure 7. As can be seen, polyethylene has three characteristic peaks at 24.9, 27.5 and 42.2° (based on cobalt anode) that are attributed to the crystalline planes of (110), (200) and (210), respectively. On the other hand, PB-1 revealed two crystal forms of I and II with the form I represent characteristic peaks at 11.3, 19.9 and 23.6° which are attributed to the planes (200), (220) and (300), respectively; and the peaks for the form II appeared at 13.5, 21.2 and 32.3° relating to the planes (110), (200) and (300), respectively. Since forms I' and III of PB-1 crystals were often expected to be obtained from solution in crystallization [54], they were not observed in the sample of this study which is formed by melt-crystallization. On the other hand, although all characteristic peaks of LDPE were appeared in sample LP10, the intensity and area below their peak clearly decreased. Furthermore, characteristic peaks attributed to form II of PB-1 have been removed from the blend, suggesting inhibition of crystallization due to the blending. The results are in good agreement with Kishore et al. who reported a sharp decrease in PB-1 crystallization in blending with LDPE while the other component was not greatly affected [17]. The crystallinity of the samples in WAXD test was calculated through the following equation after deconvolution of the patterns [55]:

Table 4. Crystal structures for LDPE and PB-1 (based on a copper anode).

Polymer	Crystal form	WAXD planes	2Θ (°)
Polyethylene	Orthorhombic	(110), (200), (020)	21.4, 23.9
Polybutene-1	I (hexagonal)	(110), (300), (200)	20.5, 17.4, 10.1
	I' (hexagonal)	(200), (300), (110)	20.5, 17.4, 10.1
	II (tetragonal)	(200), (220), (213)	18.2, 16.7, 11.7

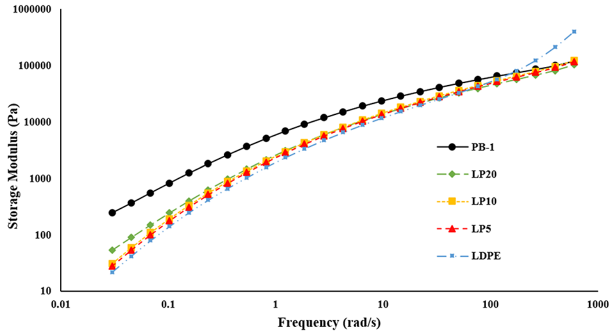


Figure 6. Storage modulus versus frequency at 190°C.

$$X_C(\%) = (\frac{A_C}{A_T}) \times 100 \tag{3}$$

In which A_C and A_T are the total area of the crystal peaks and the total area of whole peaks of patterns,

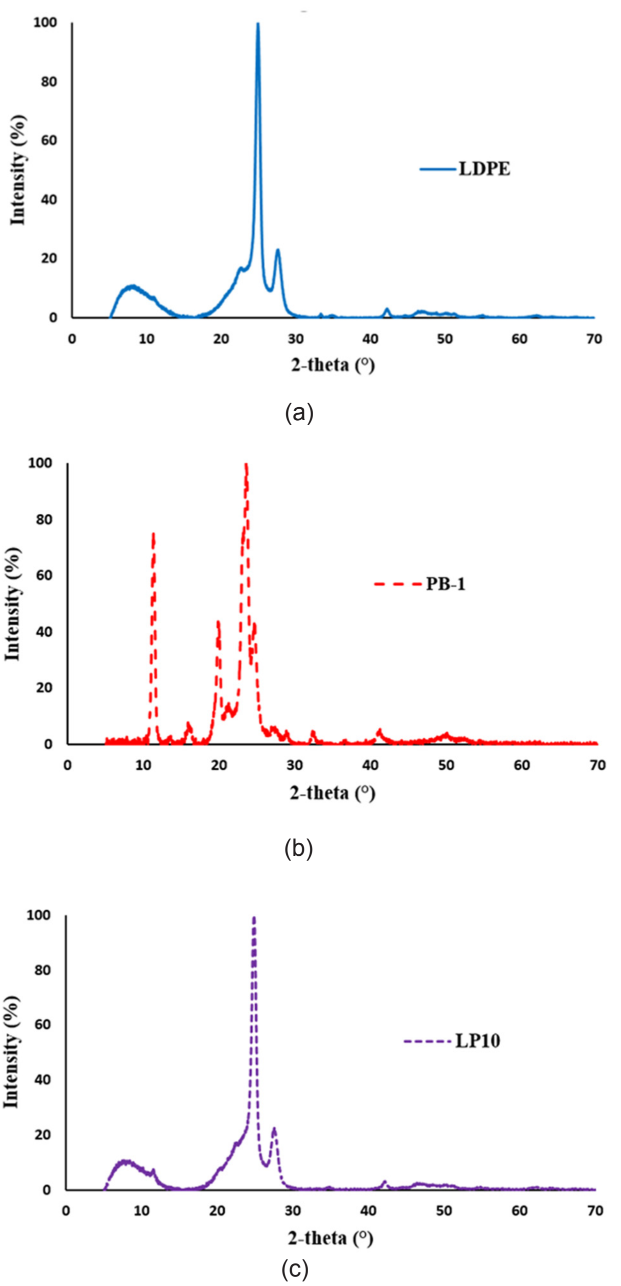


Figure 7. WAXD patterns of neat polymers and their blend (sample LP10).

respectively.

Moreover, interplanar distance (d) and crystal size, defined as the lamellar thickness (L_{hkl}) in the vertical direction to the crystal plane (hkl), were determined through the following equations [56]:

$$d = \frac{\lambda}{2\mathrm{Sin}\,\theta} \quad (4)$$

$$L_{hkl} = \frac{k\lambda}{B\,\mathrm{Cos}\,\theta} \quad (5)$$

Table 5. Diffraction parameters of crystal planes and crystallization percentage.

Sample			LDPE	PB-1	LDPB10
Polyethylene	Plane (110)	d (nm)	0.415	---	0.416
		L (nm)	14.22	---	14.43
	Plane (200)	d (nm)	0.376	---	0.377
		L (nm)	10.50	---	10.47
	Plane (210)	d (nm)	0.248	---	0.249
		L (nm)	14.99	---	15.33
Polybutene-1 (form I)	Plane (200)	d (nm)	---	0.910	0.895
		L (nm)	---	19.30	---
	Plane (220)	d (nm)	---	0.518	---
		L (nm)	---	19.37	---
	Plane (300)	d (nm)	---	0.438	---
		L (nm)	---	10.62	---
Polybutene-1 (form II)	Plane (110)	d (nm)	---	0.762	---
		L (nm)	---	---	---
	Plane (200)	d (nm)	---	0.486	---
		L (nm)	---	83.36	---
	Plane (300)	d (nm)	---	0.321	---
		L (nm)	---	---	---
X_c(%)			48.0	44.6	41.2

In these equations, λ is the wave length of X-ray (1.789 Å), B is the full width at half maximum (FWHM), ϴ is the angle the peak appears at, and k is the Scherer constant which is assumed equal to 0.9.

Diffraction parameters of the samples are summarized in Table 5. As it was expected, the initial crystallinity of LDPE is higher than that of PB-1 due to their different structure and presence of butyl side chains in PB-1 which avoid crystallization. The crystallization of both LDPE and PB-1 decreased as a result of the blending. In other words, the simultaneous presence of polymers in the blend prevented their crystallization [17, 19]. According to Table 5, the interplanar distance and lamellar thickness of LDPE crystal planes have not varied considerably. On the contrary, these parameters could not be calculated for PB-1 based on the WAXD result, as its crystallization was severely avoided. One can conclude that although the crystallinity of both phases was affected and reduced due to blending, this reduction was rather significant in PB-1 crystallization compared to LDPE.

CONCLUSION

Blends of LDPE and PB-1 containing 5, 10 and 20 wt.% PB-1 were prepared by melt mixing in a co-rotating twin-screw extruder. SEM images revealed a matrix-droplet morphology with a wide distribution of particle size. By increasing the concentration from 10 to 20 wt.%, larger particles along with a transformation of viscosity from positive to negative-deviation in comparison with mixture rule were observed, implying a week interfacial interaction between matrix and larger PB-1 droplets formed due to the demixing phenomenon. The complex viscosity and shear thinning behavior of all prepared blends were between those of neat polymers and were higher than those of neat LDPE. Moreover, the relaxation spectra of the blends were broadened by increasing the PB-1 concentration due to the presence of a component with long relaxation time in PB-1. The relaxation times were slightly increased and the results were in good agreement with those obtained by the curve-fitting the experimental data in the Carreau-Yasuda model. The higher storage modulus of the blends in comparison with the mixture rule revealed the formation of a strong interaction between the components at the interface. Characterization of the crystallinity and crystal forms of the samples by WAXD due to their coexistence in the blend showed a decrease in the crystallization of both LDPE and PB-1. Furthermore, several peaks attributed to the form II of PB-1 crystals were eliminated in the blend, while peaks attributed to LDPE were only weakened in intensity. It was found that the effect of blending on prevention of crystallization was more pronounced on PB-1 rather than LDPE. Finally, it was concluded that the blends of LDPE and PB-1 are incompatible but homogeneous, making them suitable for peeling packaging systems as reported in the literature.

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