Preparation, Morphology, and Properties of Multilamellar Barrier Materials Based on Blends of High-density Polyethylene and Copolyester

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ABSTRACT: The multilamellar barrier materials based on the blends of high-density polyethylene (HDPE) and copolyester (PETG) were prepared via melt extrusion, and poly(ethylene-*co*-acrylic acid) (EAA) as a compatibilizer was incorporated into the blends. A systematic investigation was carried out, with regard to morphology and properties. Scanning electron microscopy observation displayed the laminar morphology for the blends with the whole compositions, and the thinner laminas of the PETG phase formed in the HDPE matrix by incorporating EAA into the blends. In addition, the number and the size of the laminas of the dispersed phases were also dependant on the die temperature and screw speed, respectively. Evaluation of the mechanical properties demonstrated that incorporation of the EAA resulted in an improvement of the mechanical properties. These behaviors are attributed mainly to better adhesion and compatibility between HDPE and PETG, which has been confirmed by thermal analysis and the rheological properties. On the basis of these premises, it is reasonable to suggest that the improved barrier properties of the ternary blends with increasing concentration of the EAA be attributed to both the increase in the number of the laminas of the PETG and the decrease in their thickness, which prohibits the organic solvent molecules from entering into and permeating through the amorphous regions of the blends. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 3791–3799, 2006

Key words: HDPE; PETG; blends; multilamellar morphology; barrier properties

INTRODUCTION

The term "barrier materials" is applied to several groups of polymeric materials, which have very low permeability to gases and liquids. There is a great interest in the development of polymers with high barrier to O_2, CO_2, water, or organic vapors, and solvents permeation for use in the packaging industry. Because of a high degree of inertness to chemicals, high-density polyethylene (HDPE) is an appropriate packaging material for containers of household and industrial chemicals as well as automobile tanks.[1] A good barrier property is an important requirement of polymers used for liquid storage. However, for pure HDPE, the relatively poor resistance to permeation by hydrocarbons and organic solvents compromises its ability to replace metal and glass for certain container applications.[2] Some products such as agricultural chemicals, paint thinners, and gasoline can easily permeate HDPE containers; the results are pollution, safety, and health problems.[3]

Different techniques have been developed to produce high-barrier materials. These include layering of polymers with different barrier properties, by coextrusion, lamination, or coating, as well as metallization, orientation, e.g., biaxial stretching and blending.[4] However, surface treatment technology such as coating and metallization is not widely used nowadays because of concern for environmental safety and health because it requires substantial capital investment. In contrast, coextrusion blow molding is widely used in fabricating barrier containers for packaging hydrocarbon solvents, but is generally not available for products with complex shapes.[5] It is obvious that the blending is thought of as one of the most economical ways, owing to avoiding the complicated processing and the substantive investment in equipments. Unlike the above-mentioned two technologies, a multilamellar-blending-blow molding process can form a layered structure containing numerous discontinuous, overlapping platelets of barrier resins such as polyamide (PA), in a polyethylene (PE) matrix, which is one of the well-proven barrier technologies to enhance the resistance of PE containers to organic solvent permeation.[6] There have been numerous investigations focused on PE/PA or modified PA, PE/poly(vinyl

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alcohol), and PE/poly(ethylene-*co*-vinyl alcohol) multilamellar blending systems.[7–9] These heterogeneous multilamellar blends exhibit significantly higher permeation barrier properties than those of the conventional homogeneous blends associated with uniformly dispersed PA within the PE matrix. The laminar platelets are formed by melt process of PE, PA, and the other additives in a specially designed blend-blow-molding machine.[10] This process lends itself to a wide variety of container designs and to processing convenience.

There is a general agreement that the properties of polymer blends are usually controlled by the properties of the components, morphology of the blends, and interaction between components in the blends.[11–13] The morphology of polymer blends is intimately correlated with the properties of the blending system. The domain size is often used to indicate the extent of compatibility of multiphase polymer systems, i.e., the smaller the domain size, the more compatible the systems are and the better the mechanical properties.[14,15] A detailed investigation on phase morphological development, phase continuity, and phase stability of polymer blends has been undertaken. The correlation between morphology and properties has been established.[16,17] For the multilamellar blends used as high-barrier materials, the morphological control plays an important role in determining the permeability. The multilamellar blends are prepared by control of distribution of the dispersed domain, by controlling melt processing, where the dispersed domain is distributed as a very large, substantially two-dimensional, thin sheet and having controlled interfacial adhesion.[18]

In this study, we adopted a copolyester (PETG) to prepare high-barrier multilamellar materials, by melt blending with HDPE, where the copolyester forms the inner lamella in the HDPE matrix. The employed copolyester with a trademark of Eastar® PETG 6763 has very low organic vapors and solvent permeabilities. It is expectable that the combination of HDPE and PETG will result in a significant improvement in barrier properties, and thus extend the application of these materials. The principal aim of this study is to attempt to establish the correlation between the barrier properties and the laminar morphology, and optimize the blending composition and the processing condition to enhance the barrier properties of these multilamellar blends. For this purpose, the multilamellar blends with several compositions of HDPE and PETG were prepared via melt extrusion, and the effects of poly(ethylene-*co*-acrylic acid) (EAA) as a compatibilizer, die temperature, and screw rotation speed on the multilamellar morphology of these blends were investigated.

EXPERIMENTAL

Materials

High-density polyethylene (HDPE; 2200J, extrusion grade), with a melt flow index (MFI) of 5.5 g/10 min and a density of 0.64 g/cm^3, was purchased from Beijing Yanshan Petrochemical, China. PETG (Eastar 6763) with an intrinsic viscosity of 0.73 dL/g and a density of 1.27 g/cm^3 was supplied by Eastman Chemical, TN. It is a kind of clear and amorphous thermoplastic specially designed for use as packaging barrier materials with a regular transmittance of 89% (ASTM D-1003), a carbon dioxide permeability of 49 cm^3 mm/(m^2 24 h atm) (ASTM D-1434), and an oxygen permeability of 10 cm^3 mm/(m^2 24 h atm) (ASTM D-3985), respectively. Poly(ethylene-*co*-acrylic acid) (EAA; Nucrel 925) was obtained commercially from DuPont Chemical, Newtown, CT, which contained acrylic acid of 9.5 wt % and had a density of 0.92 g/cm^3 and an MFI of 500 g/10 min.

Preparation of blends

The pellets of all materials were dried in a vacuum oven at 80°C overnight before using. The components of the blends with various weight ratios were blended using a corotating and intermeshing twin-screw extruder (ZSK-25 mm, L/D = 32, Werner and Pfleidere, Germany) with a screw configuration adapted to HDPE. All the ingredients were tumble-mixed and fed through the throat of the extruder. The temperatures along the barrel were set from 180 to 230°C, and the rotation speed of the screw was 120 rpm. The samples were passed through a cooling water bath, and the pelletizing was subsequently carried out. The extrusion parameters were changed very little from one composition to another. The blends were further dried at 90°C before sheeting extrusion and blow molding the test specimens for determination of morphology and barrier properties.

Observation of morphology

A Plasti-Corder PLE-330 Brabender single screw extruder was employed to prepare sheet specimens via a sheeting extrusion molding. The die temperatures were set to 210, 220, 230, and 240°C, respectively. The rotation speed of the screw was also set at 20, 30, 40, and 60 rpm, respectively. The sheet specimens were immersed in liquid nitrogen for 5 min and were then fractured to obtain cross sections parallel to flow direction. The fractured surfaces were etched with boiling toluene for 10 min, and were then coated with a thin layer (10–20 nm) of gold–palladium. The coating was carried out by placing the specimen in a high vacuum evaporator and vaporizing the metal held in a heated tungsten basket. The fractured cross sections were observed and photographed using a scanning electron microscope (Cambridge S250). Scanning electron microscopy (SEM) measurements were performed only on the fractured surface of all the plates from the blended samples.

Thermal analysis

Thermal analysis was carried out using a Perkin–Elmer Pyris-1 differential scanning calorimetry (DSC) apparatus. All measurements were made under a nitrogen gas atmosphere at a heating rate of 10°C/min, on samples weighing about 10 mg. The first heating scan was carried out from room temperature to 240°C, and the sample was held at this temperature for 3 min to diminish the thermal and processing history effects before formal measurement. The melt temperature (T_m) and the heat of fusion (ΔH_m) of the samples were calculated from the maximum and the area of the endothermic peak, respectively.

Measurements of rheological properties

Apparent melt viscosities at various shear rates were determined using a Capirograph-1B capillary rheometer (Toyoseiki, Japan) with a capillary diameter of 1.262 mm and an L/D ratio of 10.51. The measurements were carried out at 230°C under the shear rates ranging from 1 to 10^4 s^{-1}.

Measurements of barrier properties

Thin-wall bottles were blow-molded using an SLG-350B extrusion-blow-molding machine, and the die temperatures and the screw speed were set to 40°C and 260 rpm, respectively. The thin-wall bottle with a wall thickness of 0.8 mm has a capacity of 200 mL and a weight of about 19 g. All bottle specimens were tested for barrier properties by filling them with permeant liquids, storing under ambient temperature conditions in an air-circulating chamber, and determining the permeabilities by weight losses. The permeant liquids used in this study are xylene, gasoline fuel, *n*-hexane, and ethyl acetate. The bottles were initially filled with 100 g of the mentioned solvents, respectively. The weight losses were measured every 5 days, and the overall storage time was 70 days.

Measurements of mechanical properties

The notched impact and tensile test bars were prepared via injection molding using an SZ-250 reciprocating screw injection machine; the barrel temperature was 240°C and the mold temperature was 90°C. The tensile properties were determined with an Instron-1185 universal testing machine using a 5000-N load transducer according to the standard of ASTM D-638. Notched Izod impact strength was measured with a SUMITOMO impact tester according to the standard of ASTM D-256. The thickness of Izod impact specimens was 1/8 in. and impact energy was 4 J. All the tests were done at room temperature and five tests were carried out for each data point.

RESULTS AND DISCUSSION

Effect of EAA on morphology

To examine the morphology of HDPE/PETG binary and HDPE/PETG/EAA ternary blends, all the sheet specimens were first subjected to brittle fracture parallels to the flow direction at a low temperature, and fractured cross sections were etched with boiling toluene to remove PETG domain or EAA domain. The remained holes could reflect the distribution of the dispersed domains in the HDPE matrix.[19]

In the HDPE/PETG binary blend at a weight ratio of 80/20 [Fig. 1(a)], some demarcated PETG laminas were observed distributing along with the flow direction; however, some spherical particles were also found in the HDPE matrix. It is well known that the HDPE is immiscible with PETG, owing to the significant polarity difference between these two polymers. In the absence of a compatibilizer, the interfacial adhesion between the HDPE and the PETG is not strong enough to keep the HDPE stretching the entire PETG domains into elongated laminas during the sheeting extrusion process. This resulted in a partial multilamellar morphology as shown in Figure 1(a). When EAA as a compatibilizer was incorporated into the HDPE/PETG blend, more defined laminas were found on the fractured cross sections of the HDPE/PETG/EAA ternary blends with an increase of EAA content, as shown in Figure 1(b–e). For the HDPE/PETG/EAA ternary blend at a weight ratio of 80/20/7, the micrograph shows that the spherical holes almost disappeared and only the defined laminas were observed on the fractured cross section, in Figure 1(e). These results suggest that HDPE should easily stretch the PETG into the elongated laminas, since the PETG is modified to adhere well to the HDPE matrix by the introduction of EAA and is not too hard or too soft to draw into continuous multilamellar structures during the blow-molding process. Because of similar chain sections between the EAA and the HDPE, these two polymers are miscible in all proportions. On the other hand, the acrylic acid moieties enhance the polarity of the EAA, and thus result in its compatibilization with the PETG.[20] Therefore, after incorporating the EAA into the HDPE/PETG blends, the EAA played the role of a bridge between the HDPE matrix and the PETG domains so as to improve their interfacial adhesion, and made the PETG-rich domains to be

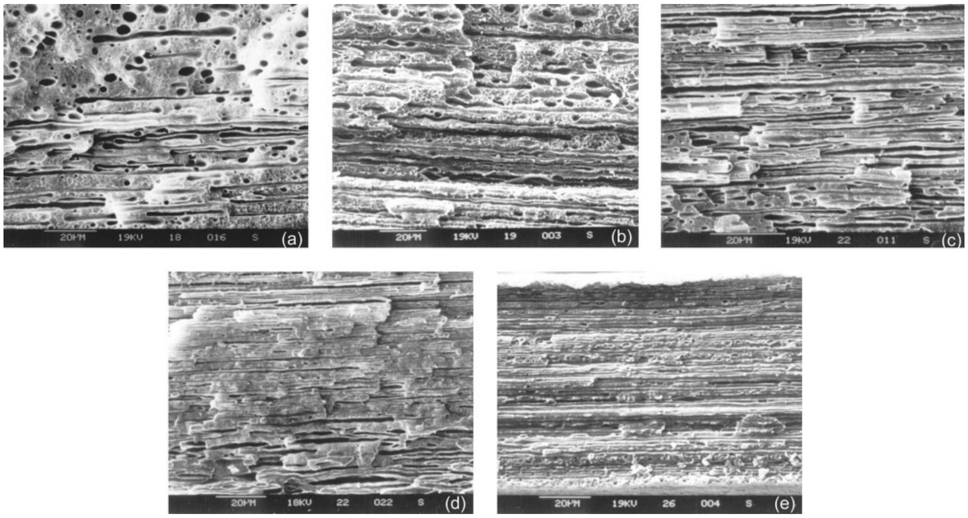

Figure 1 SEM micrographs of the fractured surfaces of the HDPE/PETG/EAA blends at a weight ratio of: (a) 80/20/0, (b) 80/20/1, (c) 80/20/3, (d) 80/20/5, and (e) 80/20/7.

drawn into continuous laminar structures by the HDPE matrix much better than that in the absence of the EAA.

Effect of processing condition on morphology

Many investigations on multilamellar morphology have reported that the number and size of layers are largely dependent on the concentration of dispersed phases.[5,6] However, under the constant concentration of the dispersed phases, the multilamellar morphology is influenced not only by the interfacial adhesion between the matrix and the dispersed domains, but also by their melt viscosity ratio during melt processing, in which the die temperature plays a key role in determining the final morphology of the blends. Figure 2 shows the SEM micrographs of the fractured cross sections for the HDPE/PETG/EAA ternary blends (weight ratio: 80/20/7) at the different die temperatures at a screw speed of 30 rpm during the sheeting extrusion. Notice that a small number of long and thick PETG layers appear on the cross section parallel to flow direction at the die temperature of 210°C. It is also found that the higher die temperatures result in an increasing number of the thinner demarcated layers. Effects of the viscosity ratio on the formation of multilamellar morphology have been studied by various investigators.[9,10] They observed various morphologies, depending on the viscosity ratio, and reported that a lower viscosity ratio was more favorable for the deformation of domains and thus formation of the laminar domains. At relatively lower temperature, the viscosity ratio of PETG phases to the HDPE matrix is so high that PETG domains cannot be dispersed in small domains and only form the larger size of the layers. Owing to the high sensitivity of the polar PETG to temperature, when the die temperature increases, the viscosity ratio decreases, and so this favors the formation of a well-developed laminar structure. As a result, the PETG phases can be easily dispersed in the small domains and then is drawn into stacked laminas by the HDPE matrix. However, when the die temperature rises to a much higher value, that is 240°C for this study, the viscosity ratio is close to unity. Although the PETG phases easily form the small domain, its laminas are easily broken during stretching, too. Therefore, a large number of the short laminas were found in the cross section at the die temperature of 250°C, as shown in Figure 2(d).

The shear effect is also significant to determine the multilamellar morphology, as demonstrated in Figure 3, which shows the SEM micrographs of the fractured cross sections for the HDPE/PETG/EAA (weight ratio: 80/20/7) ternary blends at the die temperature of 230°C in different screw rotation speeds during sheeting extrusion. It can be noted that the blends exhibit the long and regular laminas on the cross sections only in low screw speeds. The faster the screw speed, the shorter the laminas, and the sharp of the laminas also becomes anomalous, which may be resulted from the higher shear stress during the sheeting extrusion. It is believed that the stronger shear stress resulted by higher screw speed can break the melt PETG domains more easily, and is not a favorable condition for the formation of the laminas of the domains. Thus it can be concluded that the die temperature and screw speed have influences on the viscosity ratio of the blends and the shear stress, respectively, resulting in a significant effect on the formation of the multilamellar morphology.

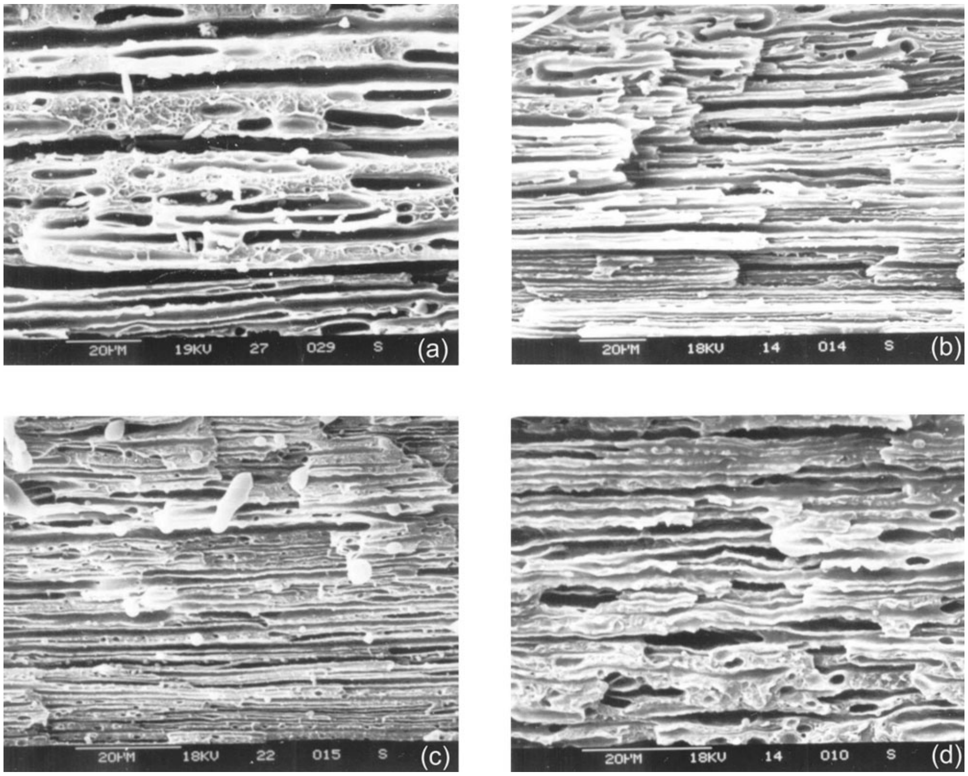

Figure 2 SEM micrographs of the fractured surfaces of the HDPE/PETG/EAA blend (weight ratio: 80/20/7) at a die temperature of (a) 210°C, (b) 220°C, (c) 230°C, and (d) 240°C.

Thermal properties

DSC was performed to characterize the thermal behavior of the HDPE and its blends with PETG and EAA. Figure 4 shows the DSC thermograms of these samples, and the melt point (T_m) and the values of heat fusion (ΔH_m) obtained from DSC analysis are summarized in Table I, in which the values of ΔH_m are first normalized to the amount, of the HDPE phases under consideration, of their concentration. The thermal data confirm the high crystallinity of the pure HDPE with T_m of 131.2°C and ΔH_m of 174.9 J/g. Furthermore, no significant changes in the T_m and ΔH_m of the HDPE phases were detected in the HDPE/PETG binary blend at a weight ratio of 80/20, while no transition temperature was observed in the DSC thermogram for the PETG phase, which reveals that the PETG is an amorphous polymer. However, from the data in Table I, it is noticed that the T_m of the HDPE phases in the ternary blend decreased slightly, but the correlative ΔH_m decreased intermediately with addition of 1 wt % EAA. It is indubitable that the transition parameters corresponding to the T_m and the ΔH_m of the HDPE phases in the ternary blend are dependant strongly on the concentration of the EAA. The higher the concentration of the EAA, the greater the decrement of the T_m and ΔH_m of the HDPE phases in the ternary blends. These results are indicative of the interaction between the EAA phases and the HDPE matrix. The presence of the EAA in the blend disturbs the crystallization of the HDPE and results in a decrease in crystallinity of the HDPE phases. On the other hand, owing to the compatibility of the EAA with the PETG, the results mentioned previously also implied a significant compatibilizing effect of the EAA on the HDPE/PETG blends.

Rheological properties

Apparent melt viscosities of the HDPE, the PETG, and their blends based on various content of the EAA were measured with a capillary rheometer at 230°C and were shown in Figure 5. It is observed that the PETG has a higher viscosity than the HDPE at low shear rates, and is opposite at high shear rates. It is well known that the rheological behaviors of the polymer blends are generally a reflection of the change in molecular weight and interaction among the components. The melt viscosity of the HDPE/PETG binary blend at the ratio of 80/20 exhibits a significant collapse, which is due to the immiscibility between these two polymers. Rheological behaviors of immiscible polymer blends having minor dispersed phases are different from those of homogeneous polymers, owing to the contributions of the minor separated phases and of the interface between the two component phases. Repulsion resulting from the immiscibility between the

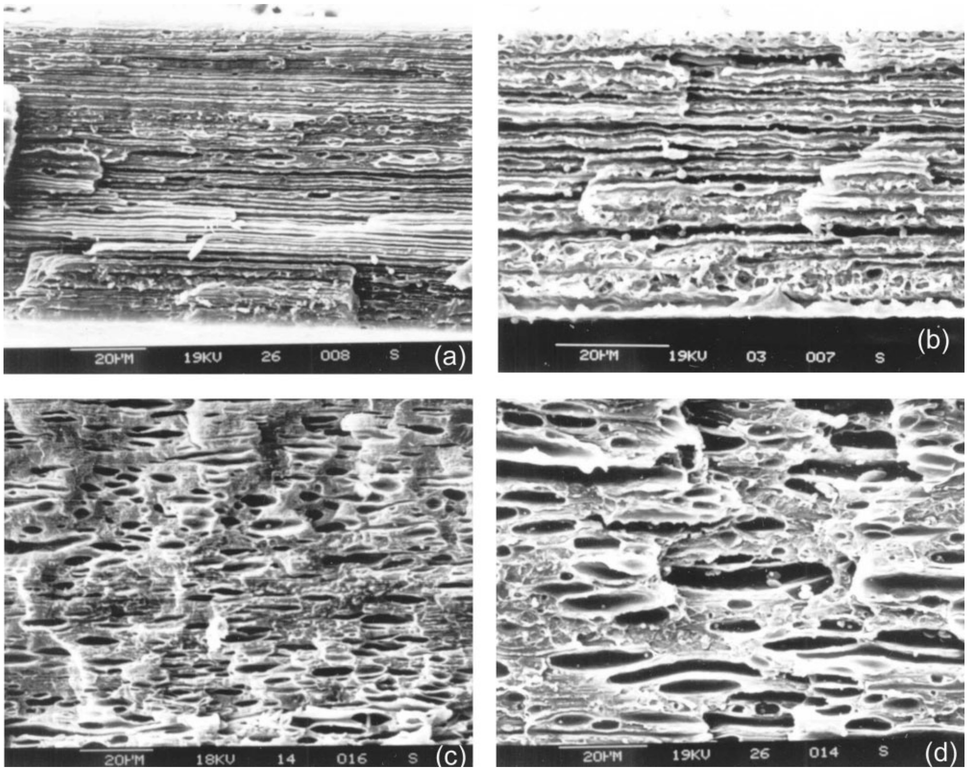

Figure 3 SEM micrographs of the fractured surfaces of the HDPE/PETG/EAA blend (weight ratio: 80/20/7) at a screw speed of (a) 20 rpm, (b) 30 rpm, (c) 40 rpm, and (d) 60 rpm.

HDPE and the PETG caused this collapse in the melt viscosity.

It can also be found that the ternary blends showed positive deviation in melt viscosities, when the EAA was incorporated into the binary blends. The higher the concentration of the EAA, the larger the positive deviation. It is known that the positive deviation in viscosity generally occurs when strong interaction exists among components at low shear rates, or when the blend has an interlocked morphology. In this system, it is evident that there is an intermolecular entanglement and an intermolecular interlock between the HDPE and the EAA, owing to the compatibility of these two polymers. In addition, it is known that strong intermolecular interaction between the PETG and the EAA was reported in the literature.[20] These miscibility and strong intermolecular interaction may be responsible for the positive deviation in the melt viscosity of the ternary blends.

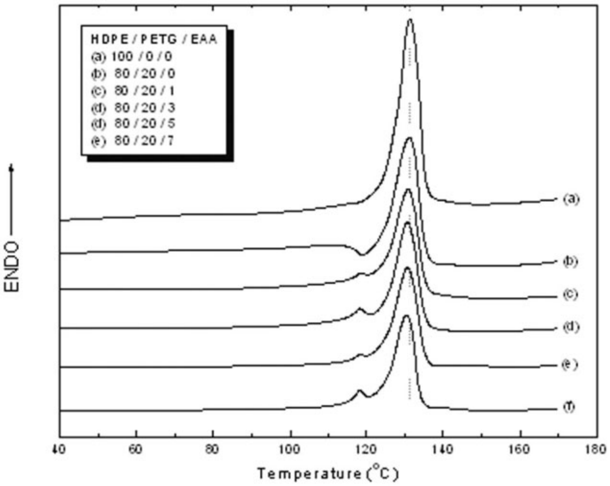

Figure 4 DSC heating traces of the HDPE and its blends with the PETG and the EAA.

TABLE I
The Thermal Properties of the HDPE and Its Blended Samples from the DSC Analysis

Sample (wt/wt/wt) HDPE/PETG/EAA	T_m (°C)	$\Delta H_m{}^a$ (J/g)
100/0/0	131.2	174.92
80/20/0	131.1	175.06
80/20/1	130.8	169.12
85/20/3	130.2	161.47
85/20/5	129.3	153.86
85/20/7	128.6	150.04

[a] Values were normalized to the amount of the HDPE phase in the blends.

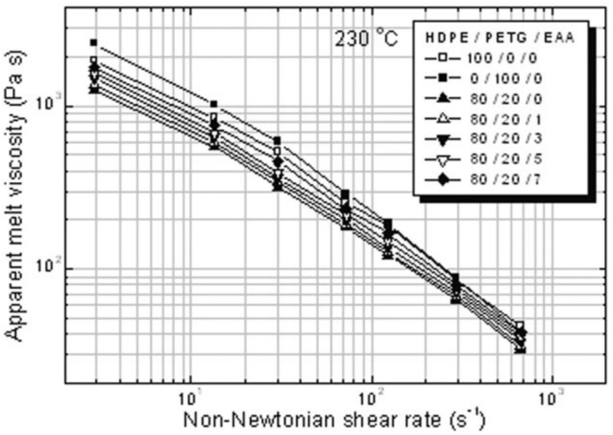

Figure 5 Plots of the apparent melt viscosity versus the non-Newtonian shear rate for the HDPE/PETG/EAA blends.

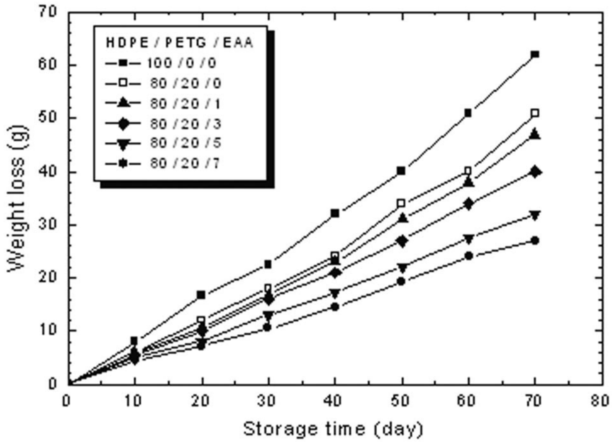

Figure 7 *n*-Hexane weight losses of the HDPE and its blends as a function of the storage time.

Barrier properties

Barrier properties of the blow molding bottles of the HDPE and its various blends were evaluated by measuring the permeability of xylene, gasoline fuel, *n*-hexane, and ethyl acetate through the thin wall of the bottle. The weight loss data characterizing the permeability properties are displayed in Figures 6–9. As expected, the pure HDPE exhibited a poor barrier property for these four solvents. It is generally recognized that the nonpolar HDPE can allow the nonpolar organic solvent molecules to easily enter into and permeate through its amorphous regions.[21] So, the weight loss of the pure HDPE decreases according to the polarity of these four solvents in the order: *n*-hexane > gasoline fuel > xylene > ethyl acetate. The presence of the polar PETG in the HDPE can prevent the nonpolar organic solvent molecules to permeate through the blends.[22] As a consequence, the HDPE/PETG binary blend at a weight ratio of 80/20 exhibits a better barrier property than the pure HDPE. On the other hand, it is found that the HDPE/PETG/EAA ternary blends exhibit much better barrier properties than the binary blend. This significant enhancement in barrier properties is attributed to the possible formation of the multilamellar structure of the PETG during melt extrusion and blow molding, which can significantly prohibit the organic solvent molecules from entering into and permeating through the whole amorphous region of the PETG. Notice that the dependence of the permeabilities on the concentration of the EAA exhibits typical characteristics expected from the compatibility of the ternary blends. Many studies on the laminar morphology have reported that the number and size of laminas in the matrix are greatly dependent on the compatibility between the dispersed phases and the matrix.[23,24] As discussed in the previous section regarding the morphology, under the constant concentration of the PETG phase, the larger portion of the EAA results in an increasing number of the

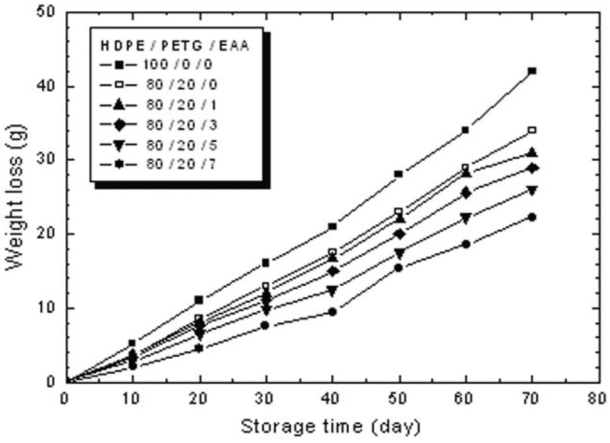

Figure 6 Gasoline fuel weight losses of the HDPE and its blends as a function of the storage time.

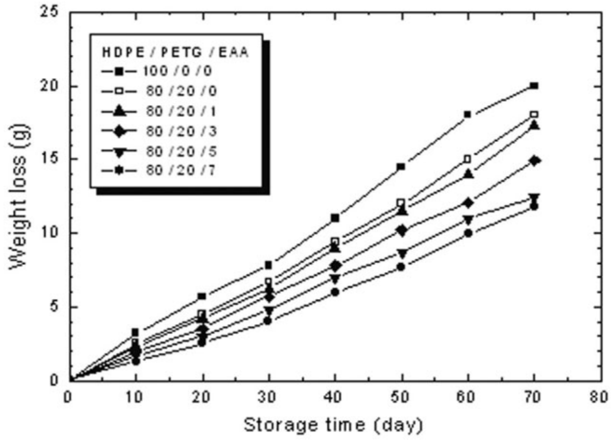

Figure 8 Xylene weight losses of the HDPE and its blends as a function of the storage time.

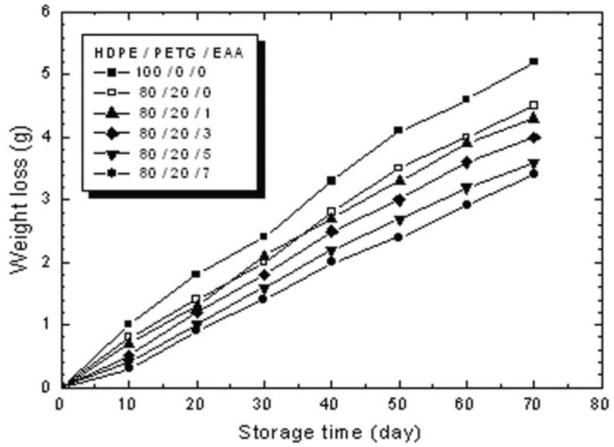

Figure 9 Ethyl acetate weight losses of the HDPE and its blends as a function of the storage time.

stacked laminas of the PETG in the HDPE matrix, and thus the barrier properties become better. In addition, the incorporation of the EAA also reduces the degree of crystallinity of the HDPE, as mentioned in the previous section regarding thermal analysis, and the increasing amorphous regions can provide much better barrier properties for the blends.[25]

Mechanical properties

Notched Izod impact strength, tensile strength, and elongation at break of the HDPE/PETG/EAA blends as a function of the EAA concentration are presented in Figures 10 and 11. The HDPE/PETG binary blend at a weight ratio of 80/20 exhibits a significant reduction with a notched Izod impact strength of 8.2 J/m by comparison of the value of 13.6 J/m for the pure HDPE as measured in this study. Furthermore, its tensile strength and elongation at break also drops sharply to 16.8 MPa and to 72.5% from the corresponding values of 25.6 MPa and 572.4% for the pure HDPE, respectively. It is understandable that the poor interfacial adhesion between the two phases resulting from the incompatibility of these two polymers deteriorates the mechanical properties of the binary blend. However, the HDPE/PETG/EAA ternary blends exhibit higher toughness accompanied by the much higher tensile properties than does the HDPE/PETG binary blend. The incorporation of the EAA into the HDPE/PETG blend results in the enhanced compatibility between the two phases. Thus, in the presence of EAA, the interfacial adhesion between the two phases increases and in turn generates better stress transfer from one phase to the other. The higher the concentration of the EAA, the larger the interfacial adhesion. As a result, the mechanical properties become better. On the other hand, EAA is also an elastomeric copolymer, and the Izod impact strength increases as the concentration of EAA increases, indicating the function of EAA as an impact modifier for HDPE. This fact can be explained in terms that the rubbery phases are highly deformed during the impact test, and thus, absorb a part of the impact energy. The rubber domains are deformed because of the shear yield, which is considered to be the main mechanism of impact toughness of the ternary blends at the service condition.

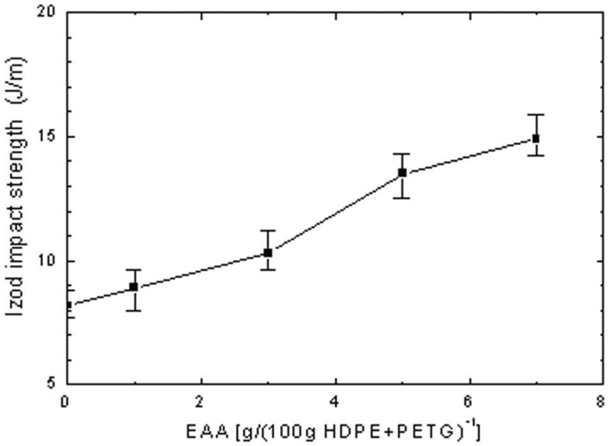

Figure 10 Notched Izod impact strength of the HDPE/PETG/EAA ternary blends as a function of the concentration of the EAA.

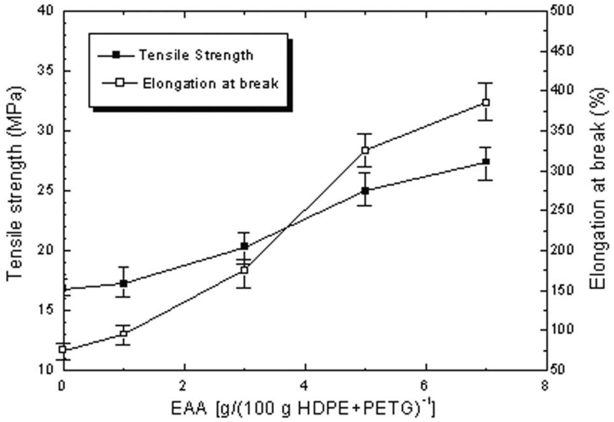

Figure 11 Tensile strength and elongation at break of the HDPE/PETG/EAA ternary blends as a function of the concentration of the EAA.

CONCLUSIONS

The multilamellar barrier materials based on the blends of the HDPE and the PETG were prepared via melt extrusion and blow molding, and their performances were evaluated in terms of the morphology, the thermal properties, the rheological properties, the barrier properties, and the mechanical properties. The laminar morphology could be observed for the blends with the whole compositions, however, more and thinner laminas of the PETG phases were formed in the HDPE matrix by incorporating the EAA into the blends, which could result in an enhancement of the compatibility between these two polymers. In addition, the number and the size of the laminas of the dispersed phases were also dependant on the die temperature and screw speed under the same composition, respectively. The incorporation of the EAA also resulted in an improvement in the mechanical properties. This is attributed mainly to better adhesion and compatibility between the HDPE and the PETG, which has been confirmed by thermal analysis and the rheological properties. On the basis of these premises, it is reasonable to suggest that the improved barrier properties of the ternary blends with increasing concentration of the EAA can be attributed to both the increase in the number of the laminas of the PETG and the decrease in their thickness, as well as the increase in the amorphous regions, which prohibits the organic solvent molecules from entering into and permeating through the amorphous regions of the blends.

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