

Modification of Low-Density Polyethylene by Graft Copolymerization with Maleic Anhydride and Blends with Polyamide 6

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ABSTRACT: Modification of low-density polyethylene (LDPE) hyperbranched grafting with a maleic anhydride (MAH) was carried out using corotating twin screw extruder in the presence of benzoyl peroxide. The LDPE/polyamide 6 (PA6) and LDPE-g-MAH/PA6 blends were obtained with a corotating twin screw extruder. The melt viscosity of the grafted LDPE was measured by a capillary rheometer. The grafted copolymer was characterized by Fourier transform infrared spectroscopy and scanning electron microscopy. The effects of variations in temperature, PA6 loading, and benzoyl peroxide and MAH concentration were investigated. The results show that most MAH monomers were grafted onto the LDPE at a lower MAH concentration. With the proper selection of the

reaction parameters, we obtained a grafting degree higher than 4.9%. Mechanical test results indicate that the blends had good interfacial adhesion and good stability of the phase structure during heating, which was reflected in the mechanical properties. Furthermore, the results reveal that the tensile strength of the blends increased continuously with increasing PA6 content. Moreover, the home-synthesized maleated LDPE could be used for the compatibilization of LDPE/PA 6 blends. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 116: 1558–1565, 2010

Key words: graft copolymers; infrared spectroscopy; morphology; polyethylene (PE); viscosity

INTRODUCTION

Manufacturing high-performance polymers is a main objective for polymer scientists and engineers. The manufacturing of multiphase plastics generally offers a way to combine several properties of individual polymer components. It can be achieved by mixing two or more polymers, which leads to the formation of polymer blends or an alloy. The possibility of compounding polyolefins [low-density polyethylenes (LDPEs)] with polar polymers such as polyamide has resulted in the development of available LDPE/polar polymer blends.^{1–4} However, LDPE and polyamide are thermodynamically immiscible and phase-separated upon blending because of poor adhesion. Improvement of the adhesion between the phases is possible by chemical compatibilization of the blend, that is, by the introduction of a third component into the two-polymer blend.^{5–11} Most commonly, compatibility can be promoted

through block or graft copolymers containing groups capable of specific interaction with the blend component. These copolymers are synthesized separately and can react with the polyamide functional group during blending.¹² Generally, the functionalization of LDPE has been successfully obtained with solutions,^{13,14} melts,^{15,16} and even a solid-state route.¹⁷ Special attention should be focused on reactive extrusion.¹⁸ For the peroxide-initiated functionalization of polyethylene (PE) in the melt state, which is often called the *reactive extrusion method*, it is generally observed that crosslinking and/or chain scission may occur simultaneously with the reaction. For PE, the dominant side reaction is crosslinking; for polypropylene, the prominent side reaction is chain scission.^{19–22} In the case of the ethylene-propylene rubber, both crosslinking and chain scission may be important and may lead to a very wide distribution of chain structure from highly degraded short chains to partially crosslinked chains and a plateau value of gel content ($\cong 50\%$).²¹ As interfacial properties are expectedly improved with increasing polar moieties, side reactions may certainly alter the rheological nature and processing characteristics along with the mechanical properties of the functionalized polymer.^{23–25} Therefore, the functionalization of LDPE via the grafting of unsaturated monomers such as maleic anhydride (MAH) and acrylic acid has

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received a great deal of attention during the last several decades.^{26–29} The modification of PE with MAH through a free-radical reaction in both solution and melt processes has been widely investigated.³⁰ Recently, Heinen et al.³¹ synthesized [2,3-¹³C₂] PE-g-MAH in both solution and melt systems, including high-density polyethylene (HDPE) and LDPE. From noise-decoupled and one-dimensional inadequate ¹³C-NMR spectroscopy, they found that the MAH groups grafted onto HDPE and LDPE were in the form of single succinic anhydrides and short oligomeric succinic anhydrides; the average chain length was about 1–2 when the grafting was administered with an MAH amount of 10 wt % PE. Covas et al.³² studied the free-radical melting grafting of MAH onto PE with an emphasis on the physicochemical phenomena developing along the screw axis of a twin-screw extruder for different PEs.

In this study, the corotating twin-screw extruder was used to study the grafting of MAH onto an LDPE chain. A Fourier transform infrared (FTIR) technique was used to evaluate the grafting degree, and another objective was to investigate the combined effects of MAH, benzoyl peroxide (BPO), and the temperature on the grafting degree, melt viscosity, and tensile strength of the reaction product.

EXPERIMENTAL

Materials

LDPE (1F7B, granules) with a density of 0.92 g/cm³, a melting point of 105°C, and a melt flow index of 7.0 g/10 min (at 190°C and under a load of 2.16 kg) was supplied by SINOPEC Petrochemical Co, Ltd. (China). Polyamide 6 was purchased from UBE Industry, Ltd. (Japan) with a density of 1.12 g/cm³, and a melting point of 215°C, and a melt flow index of 2.0 g/10 min (at 220°C and under a load of 2.16 kg).

BPO (assay 98.0%) was obtained from Tianjian Fine Chemical Co. of Shanghai and was used as received. MAH was obtained from the Tianlin Yong Da Chemical Reagent Development Center (China); it was reagent grade and was used without further purification. All other chemicals, including xylene and acetone, were reagent grade and were used as received.

Grafting procedure

The grafting reaction was performed in a corotating twin-screw extruder (Hebei Light Industry, Ltd. China, screw diameter = 30 mm, length = 706 mm). BPO and MAH were dissolved in acetone and mixed with LDPE granules. After the acetone was vaporized, MAH and BPO adhered onto the granules

homogeneously. The temperature profile of the extruder from the feed zone to the die included 140, 150, 160, and 150°C, and the rotation speed of the screws was constant at 60 rpm. The extrudate was obtained in the form of a strip, which was quenched in cooling water and palletized.

Blending: Preparation of the polyamide/grafted LDPE blends

Both the LDPE and polyamide 6 (PA6) raw materials were in the form of pellets. The PA6 pellets were dried at 110°C *in vacuo* for 24 h.

Blends of LDPE/PA6 and LDPE-g-MAH/PA6 with a composition ratio of 70 : 30 w/w were prepared by means of a corotating twin-screw extruder. The temperature profile of the extruder included 150, 215, 220, and 215°C, and the rotation speed of the screws was constant at 60 rpm. Pure PA6 was passed into the extruder to compare materials with the same thermomechanical history. The strip from the extruder was quenched in cooling water and separated into granules about 4 mm long.

FTIR analysis

The grafted samples were purified by dissolving in boiling xylene followed by the precipitation of the soluble part in acetone. The grafted polymers were separated by filtration, washed several times with acetone, and then dried at 60°C *in vacuo* until they reached a constant weight. The LDPE-g-MAH/PA6 blend samples were purified with formic acid and hot xylene as selective solvents. The blend samples were treated first with formic acid, and the insoluble fraction was separated by centrifugation of the suspensions, washed with the pure solvent and pure methanol, dried, and weighed. The dried sample was extracted further by repeated treatments with hot xylene to obtain xylene-soluble and xylene-insoluble fractions. The insoluble fractions were washed several times with acetone and then dried to a constant weight *in vacuo* at 60°C.

The purified products of the functionalized LDPE blends obtained were compression-molded into thin films with a thickness of about 0.05 mm for FTIR characterization with a Bruker Vector-22 spectrophotometer (Germany). The grafting degree of MAH onto LDPE was determined in comparison with the peak responsible for the asymmetric and symmetric stretching vibrations of a C=O in MAH at wave numbers of 1783 and 1735 cm⁻¹ in comparison with the -CH₂ rocking vibrations in LDPE at 720 cm⁻¹, which was used as the internal reference for the normalization of the spectra.^{16,33–35}

Morphological analysis

The surface morphology of the copolymers was investigated by scanning electron microscopy (SEM; JEOL/EO and model JSM-6380, Japan) at an accelerating voltage of 10 kV. The samples were fractured in liquid nitrogen, and the fracture surfaces were coated with a thin layer of gold before SEM examination.

Rheological characterization

Rheological measurements of the grafted copolymers were made with an RH 2000 capillary viscometer (Bohlin Instruments) with a die with a 1 mm diameter and a length/diameter ratio of 40 at 180°C.

Thermal analysis

Differential scanning calorimetry (DSC) analysis was performed under nitrogen flow at a scanning rate of 10°C/min with a Setaram DSC-141 instrument calibrated with indium and tin standards.

To prepare samples for the mechanical characterization, the blends were both molded by injection molding (JDH50 P series model).

Mechanical properties

The strength of the samples made of these blends was measured by with a computer-operated materials testing machine (Shenzhen Sans model CMT4503, China) with a dumbbell specimen according to ASTM D 415-80 at $20 \pm 3^\circ\text{C}$ with a crosshead speed of 50 mm/min. The data presented are the average of five tests for each case.

RESULTS AND DISCUSSION

FTIR analysis is an analytical tool frequently used to follow the maleation reaction. In Figure 1(a–c), three spectra are depicted; they are, respectively, referred to as the following: pure LDPE [Fig. 1(a)], the purified products of LDPE-g-MAH [Fig. 1(b)], and the LDPE-g-MAH/PA6 sample after a formic acid extraction procedure [Fig. 1(c)]. As shown in Figure 1(b), new absorptions appeared in comparison to Figure 1(a). These new absorptions were located at 1735 and 1783 cm^{-1} and were assigned to the asymmetric and symmetric stretching, respectively, of the carbonyl bond (C=O) of the grafted anhydride. The band observed at 720 cm^{-1} was attributed to the rocking vibrations of the $-\text{CH}_2$ bonds in LDPE. These results were consistent with the experiment of Hayes and McCarthy.³⁶ In Figure 1(c), the characteristic peaks for PA6 absorption were as follows: at 1646 cm^{-1} , C=O stretching vibrations; at 1541 cm^{-1} , N–H bending vibrations; and at 3302 cm^{-1} , N–H stretching vibrations.

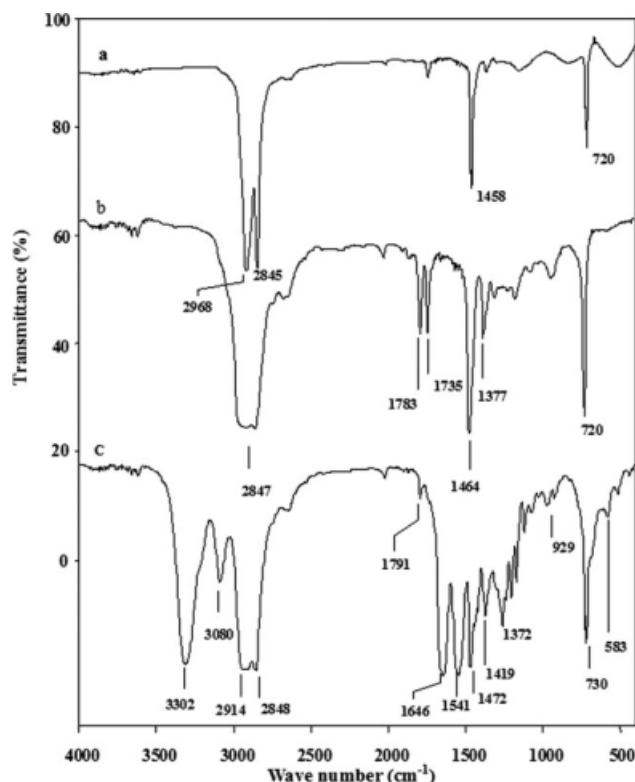


Figure 1 FTIR spectra of (a) virgin LDPE, (b) LDPE-g-MAH, and (c) LDPE-g-MAH/PA6.

Morphology

SEM micrographs of the pure LDPE, MAH-grafted LDPE, LDPE/PA6, and LDPE-g-MAH/PA6 blends are shown in Figures 2(a,b) and 3(a–f). It is clear from Figure 2(a) that the dispersed LDPE particles with an average size of about 5 μm had almost completely smooth surfaces. On the other hand, the LDPE-g-MAH surfaces are very rough, and the shape of the MAH particles is irregular; this indicated that the MAH monomers were grafted onto LDPE, as shown in Figure 2(b). The blends without added LDPE-g-MAH displayed low interfacial adhesion and poor dispersion of the minor phase droplet size. We readily observed from the micrograph shown in Figure 3(a) that the minor phase dispersion was particularly bad when blended with LDPE. The effects of the addition of an initiator are shown in Figure 3(b–f) for the 70/30 LDPE-g-MAH/PA6 blends with a 0.1, 0.2, 0.3, 0.4, and 0.5 phr BPO, respectively; when the interfacial adhesion was enhanced, the average size of the droplets was reduced considerably, and the dispersion of the minor phase was improved. Figure 3(f) clearly demonstrates the strong reduction in the dimensions of the dispersed particles in the presence of 0.5-phr BPO. Furthermore, the adhesion and the minor phase dispersion were significantly improved. An SEM microphotograph of a fracture surface of the

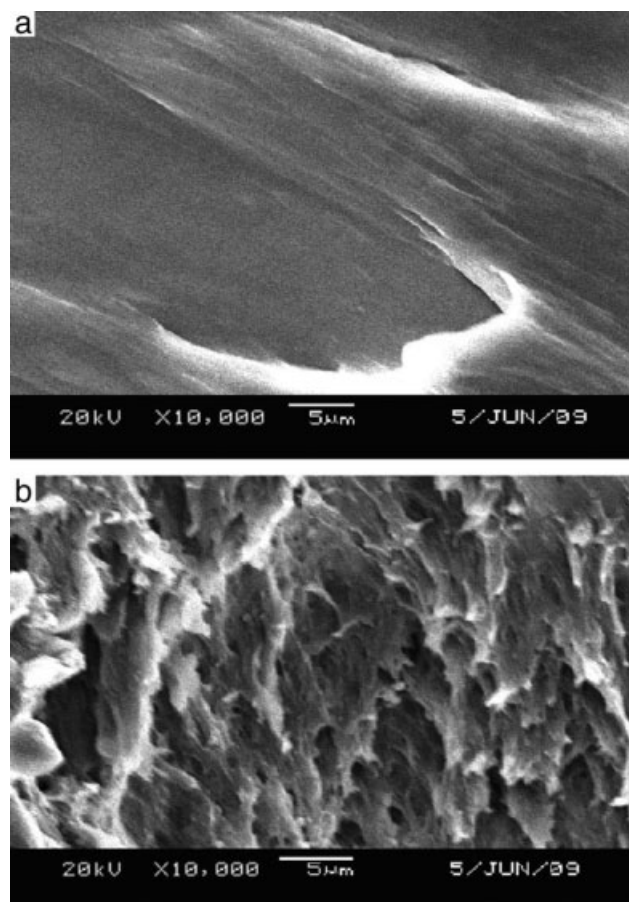


Figure 2 SEM photographs of (a) virgin LDPE and (b) LDPE-g-MAH (grafting degree = 4.9%).

ternary blends obtained from the twin-screw extruder is also shown in Figure 3(f). The influences of the simultaneous chemical and mechanical compatibilization are visible in the relatively smaller size particles of the disperse phase, and the size distribution is narrow. The better dispersion and improved interfacial adhesion were attributed to the formation of an LDPE-g-MAH/PA6 copolymer during the melt mixing.

Effect of the MAH concentration

The accurate evaluation of the form of the grafted anhydride was beyond the scope of this investigation. Indeed, even if a mathematical elaboration of the FTIR spectra could be of help in discriminating among the two cyclic forms, our spectroscopic characterizations were addressed only with the aim of determining the amount of cyclic carbonyl compound grafted to the polymer, as this method was proven to be more effective than acid group titration when the polymer was analyzed in the form of a film and properly treated for the removal of unreacted anhydride. A set of experiments was performed to investigate the influence of the initial con-

centration of MAH on the grafting process. Table I shows the variations of the grafting degree of MAH with LDPE. The grafting degree of MAH was defined as $W_{\text{MAH}}/W_{\text{total}}$, where W_{MAH} is the weight of the grafted MAH monomers and W_{total} is the total weight of the LDPE and grafted MAH monomers. Again, as shown in Table I, the grafting degree increased systematically up to about 0.1 phr but later decreased linearly with increasing MAH concentration. Similar behavior was reported by Yang et al.³⁷ In their experiment, they found that the grafting degree increased linearly with increasing MAH content and then decreased with increasing MAH content. Interestingly, the grafting degree of our laboratory-grafted LDPE is higher than that of the work described by other researchers.^{23,38} Additionally, the number-average molecular weight of the grafted PE increased markedly with increasing MAH content up to 0.1 phr, and later, the percentage of grafted MAH leveled off or decreased and, therefore, contributed to increasing the melt viscosity of the materials. The results demonstrate that the chemical reactions for the grafting almost terminated at 0.1 phr when the thermal degradation was neglected.

The influence of MAH concentration on the melt viscosity of MAH grafted LDPE is displayed in Figure 4. The melt viscosity of LDPE-g-MAH showed an increased tendency when the amount of MAH increased. According to Gaylord,³⁰ the crosslinking reaction resulted from the generation of radical sites on the polymer backbone followed by coupling. When MAH was present, the number of these sites increased, and therefore, the extent of crosslinkage increased. In addition, the interpolymer polar interaction between anhydride groups in the grafted LDPE contributed to the melt viscosity increase.

The results displayed in Figure 4 show that MAH had a pronounced effect on the tensile strength. The relative tensile strength of the LDPE-g-MAH/PA6 blends increased with increasing MAH concentration and then decreased above 0.1-phr MAH concentration. It is observed from the results that the addition of MAH substantially strengthened LDPE and PA6 interfacial adhesions. This increased interfacial adhesion for the blends is believed to have been due to the chemical reaction of the MAH group with the amine end group in the PA6 main-chain molecule; and, consequently, improve the mechanical properties of the blends. However, a further increased MAH content reduced the relative tensile strength.

Effect of the BPO concentration

Figure 5 shows the influence of the BPO concentration on the melt viscosity at a constant MAH content (0.1 phr). In the absence of BPO, no grafting occurred. As expected, the amount of grafted MAH

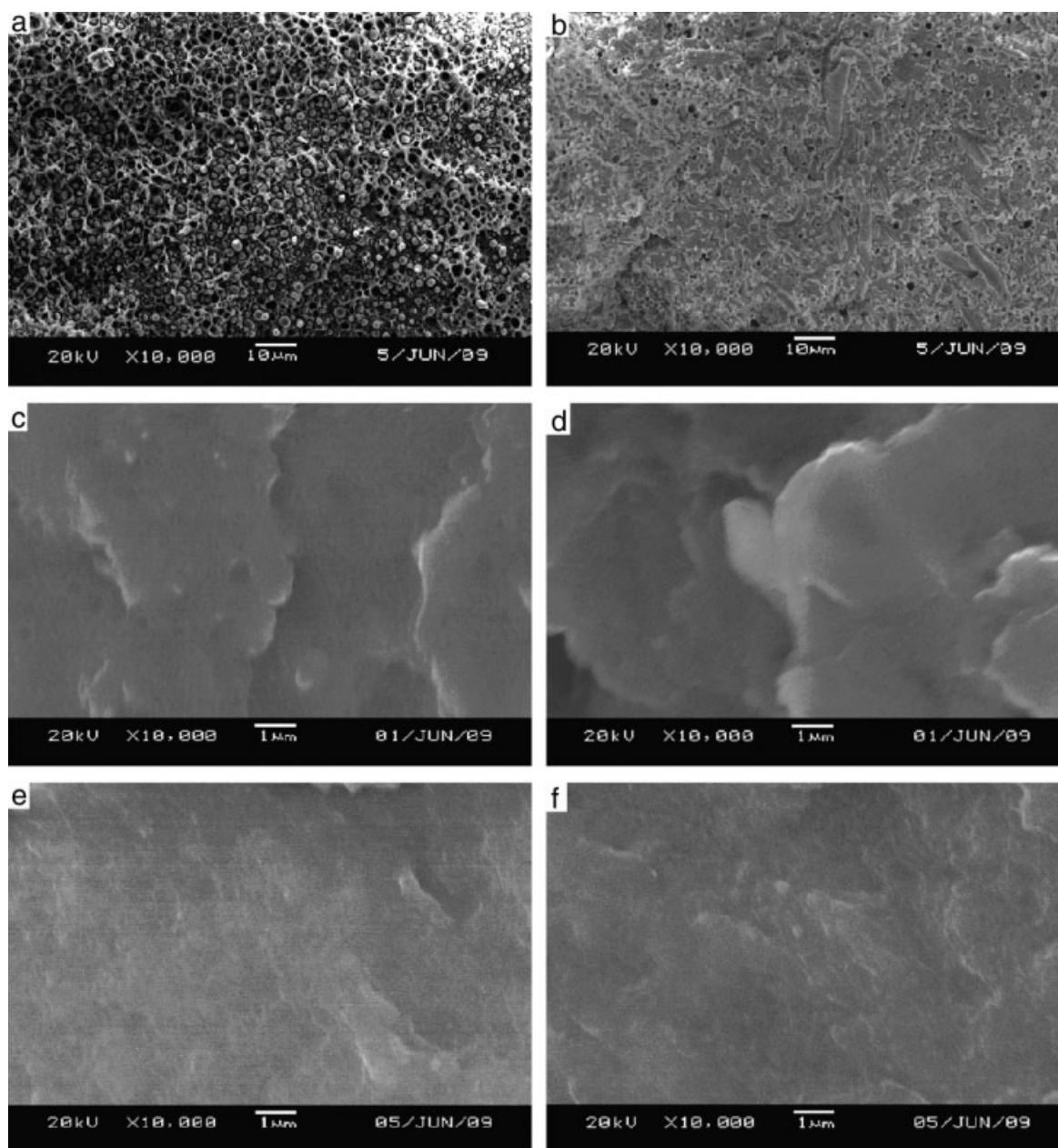


Figure 3 SEM photographs of the LDPE/PA6/BPO/MAH blends: (a) 70/30/0/0, (b) 70/30/0.1/0.1, (c) 70/30/0.2/0.1, (d) 70/30/0.3/0.1, (e) 70/30/0.4/0.1, and (f) 70/30/0.5/0.1.

increased when the initiator concentration was increased up to 0.5 phr; further increments beyond this value did not lead to any improvement in the amount of grafted MAH. However, the crosslinking side reaction increased with increasing BPO content in the MAH-grafted LDPE, the extent of which was reflected by the increase in the melt viscosity.

The influence of BPO concentration on the grafting degree is summarized in Table II.

It is observed from the results that increasing the initiator concentration at a constant MAH content of 0.1 phr resulted in an increase in the grafting degree up to about 0.5 phr, but later it dropped continu-

ously with increasing BPO concentration. Moreover, the number-average molecular weight of grafted LDPE increased systematically with increasing BPO

TABLE I
Effect of the MAH Content on the Grafting Degree

Sample	MAH (phr)	BPO (phr)	Grafting degree (%)
1	0.03	0.5	0.5
2	0.05	0.5	0.9
3	0.1	0.5	4.9
4	0.2	0.5	2.2
5	0.3	0.5	0.8

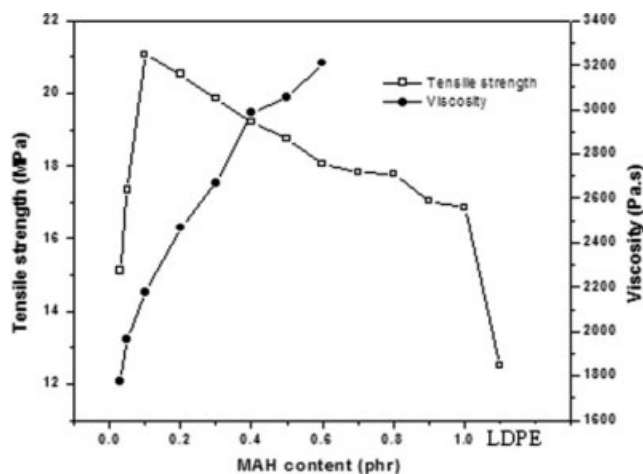


Figure 4 Effects of the MAH concentration on the melt viscosity of maleated LDPE and on the tensile strength of LDPE-g-MAH/PA6 (100-phr LDPE and 0.5-phr BPO).

concentration and, therefore, contributed to a net enhancement in the melt viscosity of the material.³⁹ It is suggested that the chemical reactions for the grafting are almost complete at 0.5 phr when the thermal degradation is ignored.

It was expected that the instantaneous mechanical properties, namely, the tensile strength, were found to depend exclusively on the BPO concentration. The results are depicted in Figure 5. As shown in Figure 5, the tensile strength significantly increased up to about 0.5 phr; then, it decreased continuously with increasing BPO concentration. The increasing tendency of the tensile strength confirmed that the interfacial adhesion was enhanced by the reaction of the polyamide with the MAH units. These results are consistent and, to some extent, better than those reported by Kelar and Jurkowski.⁴⁰ In their study, they observed that the mechanical properties of

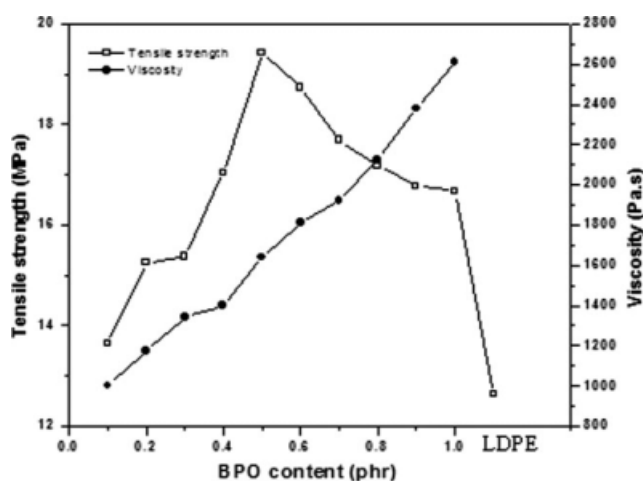


Figure 5 Effects of the BPO content on the melt viscosity of maleated LDPE and on the tensile strength of LDPE-g-MAH/PA6 (100-phr LDPE and 0.1-phr MAH).

TABLE II
Effect of the BPO Concentration on the Grafting Degree

Sample	MAH (phr)	BPO (phr)	Grafting degree (%)
1	0.1	0.3	0.7
2	0.1	0.4	1.9
3	0.1	0.5	4.9
4	0.1	0.6	2.02
5	0.1	0.7	0.75

LDPE/PA6 blends increased as the miscibility of the system became better.

Effect of temperature

The influence of the reaction temperature was also a source of investigation. In fact, quite obviously, when the temperature was increased, the rate of generation of free-radical species increased owing to the faster decomposition of the initiator. We observed that at fixed BPO and MAH ratios, the grafting level initially increased with increasing reaction temperature up to about 160°C and then decreased continuously with further increasing temperature. The results are reported in Table III. The decrease in grafting level was due to chain scission of LDPE at higher reaction temperatures.

The results of the temperature dependence on the melt viscosity are depicted in Figure 6; with an observation that the melt viscosity of the sample appears to some first increase with increasing reaction temperature and then obviously decrease above a reaction temperature of 160°C. The decrease in the melt viscosities caused by the generation of free-radical species was made faster owing to the faster decomposition of BPO, which led to the chain scission of LDPE at high temperatures. The tensile properties of the blends are summarized in Figure 6. The tensile strength increased initially with increasing reaction temperature up to about 160°C; then, it decreased linearly with increasing reaction temperatures. The results demonstrate that with the proper selection of the operative conditions, the grafting degree increased considerably with increasing reaction temperature; thereafter, the percentage of grafted leveled off or decreased with increasing reaction temperature.

TABLE III
Effect of Temperature on the Grafting Degree

Sample	Temperature (°C)	MAH/BPO (phr)	Grafting degree (%)
1	150	0.1/0.5	2.01
2	160	0.1/0.5	4.2
3	170	0.1/0.5	2.3
4	180	0.1/0.5	1.02

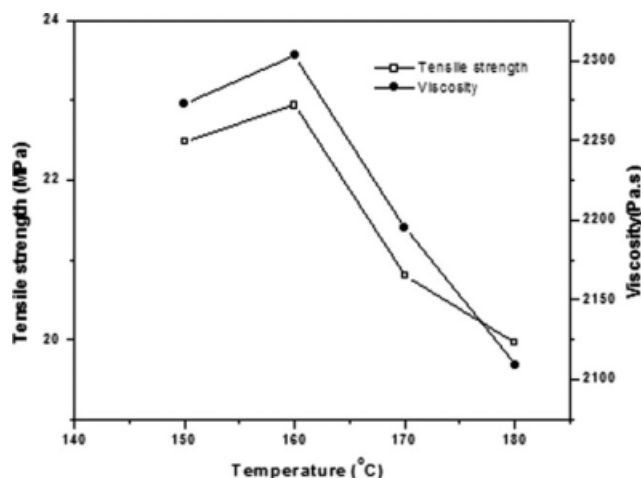


Figure 6 Effects of the temperature on the melt viscosity of maleated LDPE and on the tensile strength of LDPE-g-MAH/PA6 (100-phr LDPE, 0.1-phr MAH, and 0.5-phr BPO).

Mechanical properties

A further part of our study was oriented to evaluate the end-use properties of the grafted and ungrafted LDPE, and their blends with different content of PA6 are illustrated in Figure 7; and were found to further support the morphological observations. It is observed that LDPE-g-MAH/PA6 exhibit a higher tensile strength than LDPE/PA6. As shown in Figure 7, the blending of PA6 with LDPE-g-MAH obviously improved the mechanical properties, and the mechanical properties of blends increased as the miscibility of LDPE and PA6 improved. The tensile strength of the blended polymer was found to depend exclusively on PA6 loading. The results reveal that the tensile strength increased almost linearly with increasing PA6 contents in a gradient way.

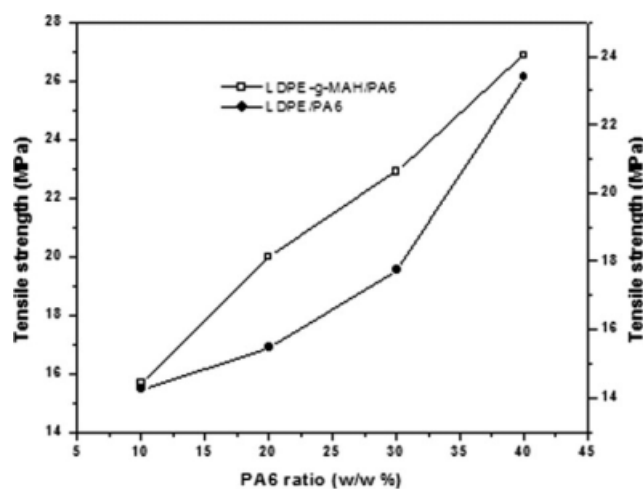


Figure 7 Effects of PA6 on the tensile strength of LDPE/PA6 and LDPE-g-MAH/PA6 (100-phr LDPE, 0.1-phr MAH, and 0.5-phr BPO).

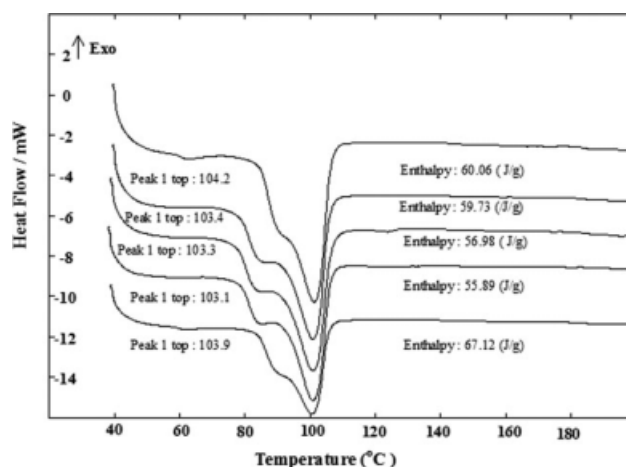


Figure 8 DSC curves for the heating of LDPE-g-MAH.

This is due to both particle size reduction and an improvement in the interfacial adhesion caused by the formation of the LDPE-g-PA6 copolymers, which appeared to lead to an improvement in the mechanical properties.

The DSC melting thermogram of LDPE-g-MAH is shown in Figure 8. The melting behavior of the extruded LDPE sample was clearly affected by the presence of the MAH functional group. With increasing grafting degree, the melting temperature shifts to a lower temperature slightly and then increase a little. This suggests that a high grafting degree may have induced a different microstructure than that of a sample with a low grafting degree. The introduction of the MAH group led to an obvious decrease in the crystallinity. These results are in agreement with those of Collar et al.,⁴¹ who studied the thermal behavior of HDPE. These authors found that if polar groups were introduced to the repetitive units on the crystalline phase, the system response was acutely effected by a need to accommodate a more voluminous amorphous phase. Therefore, the crystallinity of each grafted sample is lower than that of pure HDPE. DSC analysis of our maleated LDPE samples show multiple endotherms with a peak of about 84°C and a second endotherm at 103°C; this indicating a lower degree of crystallinity for the functionalized PE.

CONCLUSIONS

The reactive extrusion process was developed for the chemical modification of LDPE with BPO as an initiator. The experimental results evidence that the grafting degree and tensile strength increased systematically to a peak value and then dropped sharply with increasing BPO concentration. Moreover, the results show that the number-average molecular weight of the grafted PE increased linearly with

increasing BPO content up to 0.5 phr, but later, the percentage of grafted PE leveled off or decreased and, therefore, contributed to an increase in the melt viscosity of the materials. The results reveal that the tensile strength grafting degree increased appreciably with increasing MAH concentration up to about 0.1 phr and then decreased continuously with further increasing MAH content. Furthermore, the experimental results indicate that the melt viscosity increased almost linearly with increasing MAH content.

The mechanical properties of the PA6/LDPE blends increased as the miscibility of the LDPE and PA6 improved. Proper selection of the operative conditions obtained a grafting degree higher than 4.9%. The whole experimental picture suggested that the maleation of LDPE was successfully performed by reactive extrusion in the twin-screw extruder and, therefore, allowed the achievement of a high grafting level in the presence of limited molecular degradation of the polymer.

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