

Thermoplastic Elastomers Based on Epoxidized Natural Rubber and High-Density Polyethylene Blends: Effect of Blend Compatibilizers on the Mechanical and Morphological Properties

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ABSTRACT: Epoxidized natural rubber (ENR) with a level of epoxide groups of 20 mol % was prepared via the performic epoxidation method. It was then used to blend with high-density polyethylene (HDPE) at various blend ratios. Three types of blend compatibilizers were prepared. These included a graft copolymer of HDPE and maleic anhydride (MA; i.e., HDPE-g-MA) and two types of phenolic modified HDPEs using phenolic resins SP-1045 and HRJ-10518 (i.e., PhSP-PE and PhHRJ-PE), respectively. We found that the blend with compatibilizer exhibited superior tensile strength, hardness, and set properties to that of the blend without compatibilizer. The ENR and HDPE interaction via the link of compatibilizer molecules was the polar func-

tional groups of the compatibilizer with the oxirane groups in the ENR molecules. Also, another end of the compatibilizer molecules (i.e., HDPE segments) was compatibilizing with the HDPE molecules in the blend components. The blend with compatibilizer also showed smaller phase morphology than the blend without compatibilizer. Among the three types of the blend compatibilizer, HDPE-g-MA provided the blend with the greatest strength and hardness properties but the lowest set properties. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 109: 2694–2702, 2008

Key words: blends; compatibilization; morphology; polyethylene (PE); rubber

INTRODUCTION

Thermoplastic elastomers (TPEs) based on rubber-thermoplastic blends are classified into two distinct classes. One class consists of a simple blend and is commonly called a TPE polyolefin or a thermoplastic polyolefin. The rubber phase of the thermoplastic polyolefin is unvulcanized material. In the other class, the rubber phase is dynamically vulcanized, which gives rise to thermoplastic vulcanizates, dynamic vulcanizates, or elastomeric alloys.¹ TPEs prepared from natural rubber (NR) and thermoplastic blends are known as thermoplastic natural rubbers (TPNRs). Various types of thermoplastics are used to prepare TPNRs. These include polyolefins, such as polypropylene,^{2–7} low-density polyethylene (PE),^{8,9} ultra-low-density PE,¹⁰ linear low-density PE,¹¹ and chlorinated PE.^{12,13} Some other types of

thermoplastic are also used to blend with NR: polystyrene,¹⁴ polyamide 6,¹⁵ ethylene-vinyl acetate copolymer,¹⁶ and poly(methyl methacrylate).¹⁷ High-density polyethylene (HDPE) is another type of polyolefin used to prepare TPNRs with improved tensile properties and hardnesses from the addition of liquid NR as a compatibilizer.¹⁸ However, our previous study indicated that liquid NR compatibilizer leads to inferior physical and rheological properties in NR/HDPE blends. It was found that the phenolic-modified HDPE provided a positive compatibilizing effect on this type of blend.¹⁹ Epoxidized natural rubber (ENR) is one of the modified NRs currently used to prepare TPNRs. Various types of thermoplastics were used as blend components: poly(vinyl chloride),²⁰ poly[ethylene-co-(acrylic acid)],²¹ and poly(methyl methacrylate).²² Graft copolymers of NR with poly(methyl methacrylate) are another modified NR that has also been used to prepare TPNRs.^{23,24}

In this study, an attempt was made to prepare TPNRs based on the blending of ENR with level of epoxide groups of 20 mol % (i.e., ENR-20) with HDPE via a simple blend process (i.e., a blend without curatives). Various types of compatibilizer, including a graft copolymer of HDPE and maleic

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anhydride (MA; HDPE-*g*-MA) and a phenolic-modified HDPE, were used with various blend ratios of ENR-20/HDPE. Furthermore, the mechanical and morphological properties of the blends were investigated.

EXPERIMENTAL

Materials

High-ammonia concentrated NR latex used as a raw material for preparation of ENR was manufactured by Yala Latex Co., Ltd. (Yala, Thailand). The non-ionic surfactant used to stabilize the latex during epoxidation was Teric N10 (nonylphenol ethoxylates), which was manufactured by Huntsman Corp. Australia Pty, Ltd. (Ascot Vale Vic, Australia). The formic acid used as a reactant for the preparation of ENRs was manufactured by Fluka Chemie (Buchs, Switzerland). The hydrogen peroxide used as a co-reactant for the preparation of the ENRs was manufactured by Riedel De Haën (Seelze, Germany). The HDPE a blend component was manufactured by the Thai Polyethylene Co., Ltd. (Rayong, Thailand). It was injection-molding grade (H6007J) with a melt flow index of $7.5 \text{ g}/10^{-1} \text{ min}$ (2.16-kg loads at 190°C) and a density of $0.97 \text{ g}/\text{cm}^3$.

The MA used to prepare HDPE-*g*-MA was manufactured by Riedel-de Haën. Two types of phenolic resin, dimethylol phenolic resin or octylphenol-formaldehyde resin (SP-1045) and phenolic resin with active hydroxymethyl (methylol) groups (HRJ-10518; made from the reaction of octylphenol and formaldehyde), were used as reactants for the preparation of the blend compatibilizers and were manufactured by Schenectady International, Inc. (Newport, CT). The stannous chloride or tin(II) chloride hydrate ($\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$) used as a catalyst for the preparation of phenolic-modified HDPE was manufactured by Ajax Finechem, Ltd. (New South Wales, Australia).

Preparation of the ENR

The ENR was prepared with high-ammonia NR latex with a dry rubber content of approximately of 60%. Details of the preparation process of the ENR were described in our previous article.²⁵ However, in this study, the nonionic surfactant Teric N10 was used to stabilize the latex instead of Teric N30. This was because Teric N10 had a lower cloud point, in the range $65\text{--}69^\circ\text{C}$. Therefore, the ENR latex stabilized by Teric N10 was able to coagulate if heated to a temperature above 70°C . After the coagulation, the ENR was washed thoroughly with distilled water and dried in a vacuum oven at 40°C for 24 h. Infrared and $^1\text{H-NMR}$ spectroscopies were used to analyze the molecular structure of the ENR. The reaction time of the epoxidation was set according to the

required level of epoxide groups in the ENR product, as described elsewhere.⁶ The level of the ENR products was later confirmed by infrared spectroscopy via a calibration curve. In this study, ENR with a level of epoxide groups at approximately of 20 mol % (ENR-20) was used.

Preparation of the blend compatibilizers

Two types of blend compatibilizers were prepared in this study. These included the phenolic-modified HDPE and HDPE-*g*-MA with the following procedures:

1. Phenolic-modified HDPE was prepared by the melt-mixing of HDPE and phenolic resins, as described elsewhere.¹⁹ The HDPE (100 parts) was first blended with dimethylol phenolic resin (SP-1045, 4 parts) and stannous chloride (0.8 parts) at 180°C . The product was called Phenolic SP-1045 modified HDPE, PhSP-PE. The other type of phenolic-modified HDPE was prepared with the same procedure used for PhSP-PE but with SP-1045 replaced by HRJ-10518. The latter product was called phenolic HRJ-10518 modified HDPE, PhHRJ-PE. Fourier transform infrared (FTIR) spectroscopy was used to characterize the phenolic-modified products by fabrication into a thin film by compression molding. The thin film was then extracted with acetone to eliminate unreacted substance before characterization with FTIR.
2. HDPE-*g*-MA was also prepared by a melt-mixing process of HDPE and MA. Before mixing, HDPE (100 parts by weight) was dried at 50°C for 24 h to eliminate moisture. MA (5 parts by weight) and dicumyl peroxide (DCP) (2 parts by weight) were dissolved in acetone and then mixed with the HDPE granules. After the acetone was volatilized, MA and DCP adhered to the granules homogeneously. They were then incorporated into the mixing chamber of an internal mixer, a Brabender plasticorder model PLE331 (Brabender OHG, Duisburg, Germany) with a mixing chamber with a capacity of 80 cm^3 . The mixing was then performed at a screw speed of 60 rpm at 180°C and a fill factor of 0.8 for 10 min. A thin film was later prepared by the compression-molding technique. The film was then extracted by acetone to remove unreacted chemicals before characterization with FTIR.

Preparation of the TPEs based on ENR-20/HDPE blends

ENR and HDPE blends were prepared at various blend ratios of ENR-20/HDPE, 25/75, 40/60, 50/50,

60/40, and 75/25, with three types of blend compatibilizers, PhHRJ-PE, PhSP-PE, and HDPE-g-MA, at the same loading level of 3 wt % HDPE. Blending was carried out via a melt-mixing process with an internal mixer, the Brabender plasticorder model PLE331. The thermoplastic component, HDPE, was first dried in a hot air oven at 50°C for at least 10 h. The HDPE was introduced into the mixing chamber and left without rotation for 6 min to warm the material. Mixing was then performed for 2 min at a rotor speed of 60 rpm at 180°C. The compatibilizer was then incorporated into the mixing chamber, and mixing was continued for another 1 min. The ENR-20 was added and mixed for 2 min. The blending products were later cut into small pieces with a Bosco plastic grinder (Bosco Engineering, Samutparakarn, Thailand). The mechanical and morphological properties were later investigated.

Mechanical testing

Tensile testing of the samples was performed at $25 \pm 2^\circ\text{C}$ at a crosshead speed of 500 mm/min according to ISO 37. The instrument used was a Hounsfield tensometer model H 10 KS manufactured by the Hounsfield Test Equipment Co., Ltd. (Surrey, UK). Dumbbell-shaped specimens, 2 mm thick, were prepared by a thermoplastic injection-molding machine with a capacity of 90 tons (the clamping force, Welltec Machinery, Ltd., Hong Kong). Temperature profiles were set as 170, 175, 180, 180, and 180°C for barrel zones 1 to 4 and an injection nozzle, respectively. The mold temperature at 25°C was controlled by a water-circulating system. Tension set at 100% elongation was also determined at room temperature ($25 \pm 2^\circ\text{C}$) according to ISO 2285. The samples were kept under tension for a fixed elongation and time interval, then released from the clamp, and kept aside for another fixed time interval, and the changes in the sample dimensions were determined. The hardness of the samples was also measured with Shore A indentation according to ISO 7619.

Morphological studies

Morphological studies were carried out with a Leo scanning electron microscope model VP 1450 manufactured by Leo Co., Ltd. (Cambridge, United Kingdom). Molded samples of the TPE were cryogenically cracked in liquid nitrogen to prevent any possibility of phase deformation. The ENR phase was extracted by the dissolution of the fractured surface in chloroform. The samples were then dried in a vacuum oven at 40°C for 3 h. The dried surfaces were later gold-coated and examined by scanning electron microscopy.

RESULTS AND DISCUSSION

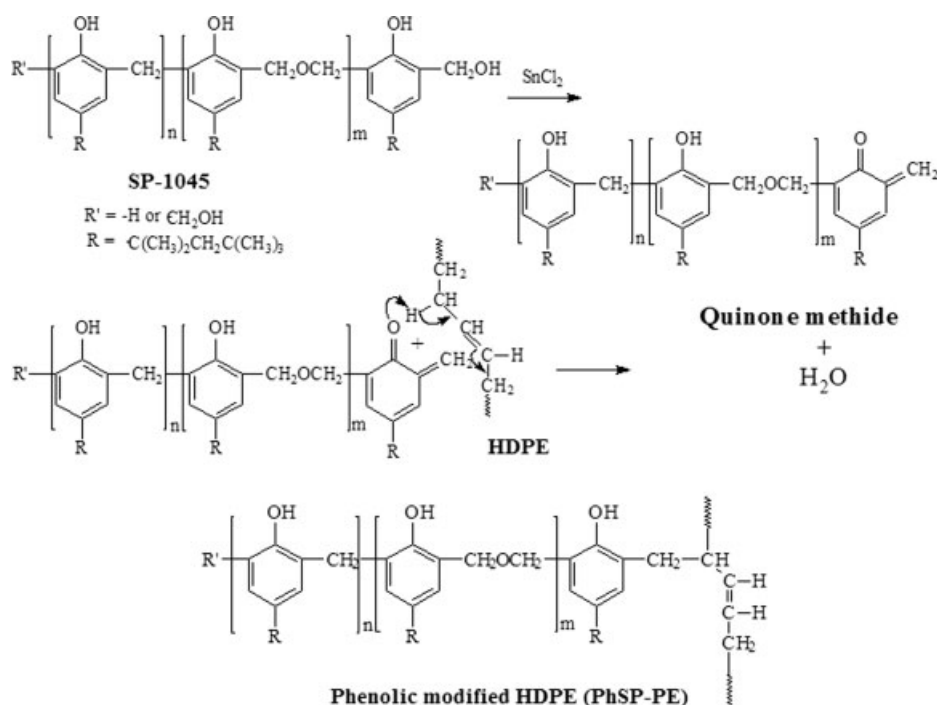
Mechanism for the synthesis of the compatibilizers

Phenolic-modified HDPEs (i.e., PhHRJ-PE and PhSP-PE) were prepared with phenolic resins (i.e., SP-1045 and HRJ-10518). FTIR spectra were used to confirm the structures of the reaction products, as described in our previous article.¹⁹ The reaction mechanism between HDPE and the phenolic resins (i.e., SP-1045 and HRJ-10518) and the structure of the final products are postulated in reaction Schemes 1 and 2. The phenolic resin reacted with the Lewis acid catalyst ($\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$), and quinone methide was a main product, whereas water was the byproduct of this reaction. The quinone methide was capable of reacting with a trace level of unsaturation in this special type of HDPE molecule by rearrangement of the bonds. The molecules of the compatibilizers contained phenolic and HDPE moieties. The existence of double bonds in the HDPE molecules was proven in our previous study¹⁹ and are also shown in Figure 1 at a wave number of 1640 cm^{-1} .

Figure 1 shows the infrared spectrum of HDPE-g-MA compared with that of unmodified HDPE. An intense characteristic band at 1790 cm^{-1} and a weak absorption band at 1860 cm^{-1} were observed. These bands were assigned to grafted anhydride due to the symmetric (strong) and asymmetric (weak) C=O stretching vibrations of succinic anhydride rings, respectively. Also, a low content of C=O stretching vibration of succinic acid at 1710 cm^{-1} was observed. This peak was assigned to grafted maleic acid, which was a product of the ring opening of succinic anhydride because of moisture. This proved the presence of grafted anhydride on the HDPE molecules. The possible reaction mechanism is proposed as shown in Scheme 3. The radicals from the dissociation of DCP reacted randomly with the hydrogen atom on the HDPE molecular chains, which later generated a radical on the HDPE molecule. This was a reactive site to the π electron on the $-\text{C}=\text{C}-$ bond of MA. The grafted product (HDPE-g-MA) was eventually formed. In the case of the HDPE chain containing trace amounts of side groups, such as methyl, ethyl, or even larger groups, the tertiary carbon atom radicals produced by peroxide initiation could have possibly reacted with MA and formed the another type of grafted product, as shown in Scheme 4.²⁷

Mechanical properties

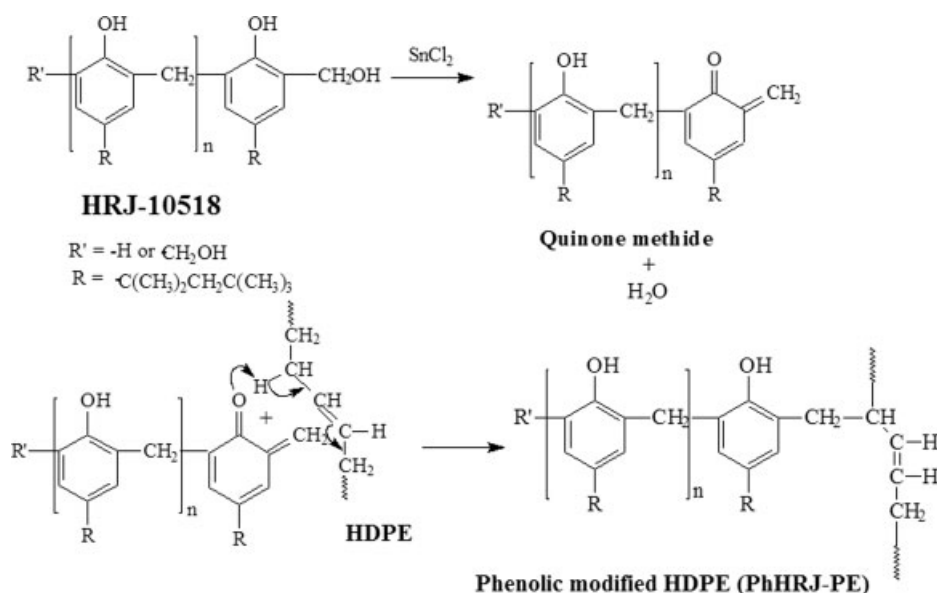
Figure 2 shows the stress-strain curves of ENR-20/HDPE blends without compatibilizer at various blend ratios. In the blends with a rich HDPE phase (i.e., ENR-20/HDPE = 25/75 and 40/60), a pronounced hump in the curve became apparent, the



Scheme 1 Mechanisms of the reactions between SP-1045 and HDPE.

apex of the hump being a yield point. However, with increasing levels of ENR, the yielding phenomena disappeared. The characteristic stress-strain behavior of the elastomers was more pronounced. That is, at lower stresses, the higher strain was observed. Young's modulus was also estimated from a slope of the linear region of the stress-strain curves. The moduli increased with increasing HDPE content in the blend. Therefore, an increase in the stiffness of the material was consequent. The influ-

ence of various types of blend compatibilizers (i.e., HDPE-*g*-MA, PhHRJ-PE, and PhSP-PE) on the stress-strain behavior of 60/40 ENR-20/HDPE blends is shown in Figure 3. The incorporation of blend compatibilizers caused an abrupt increase in the tensile stress at a given strain (compared to the blend without compatibilizer). That is, the trend of the tensile stress of the blends at the same level of strain could be ordered with the type of blend compatibilizer as follows: HDPE-*g*-MA > PhHRJ-PE



Scheme 2 Mechanisms of the reactions between HRJ-10518 and HDPE.

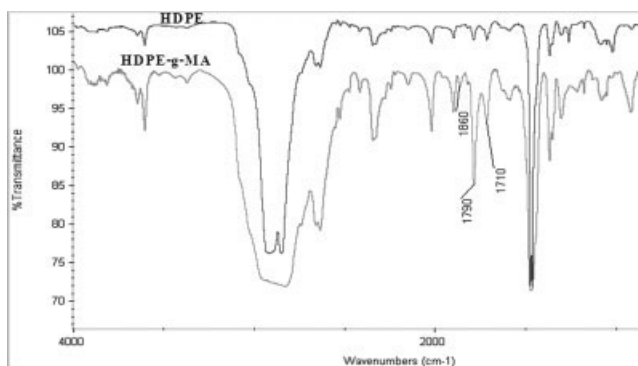
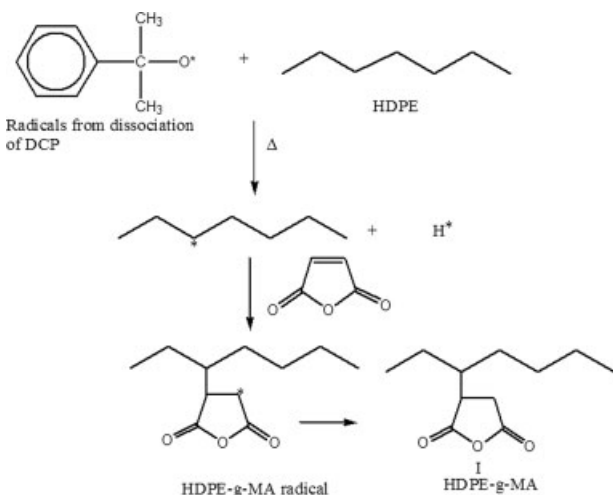


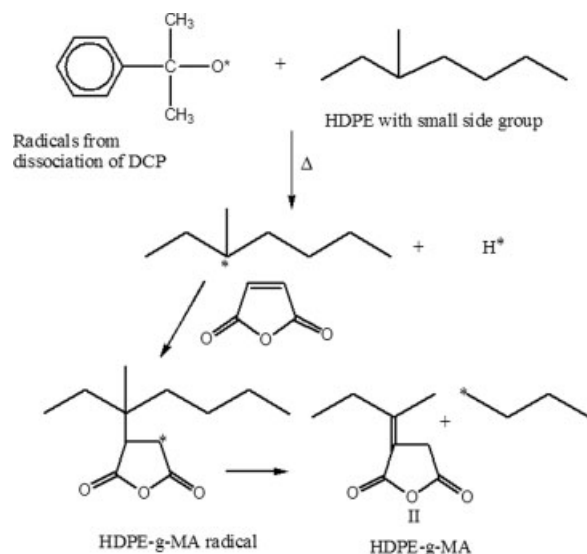
Figure 1 Infrared spectrum of HDPE-g-MA compared with that of unmodified HDPE.

> PhSP-PE > Without compatibilizer. The reinforcement of the blend was attributed to the chemical interaction between the HDPE and ENR-20 phases via the compatibilizer molecules. In the case of the HDPE-g-MA compatibilizer, the chemical interaction between polar groups in the ENR molecules and succinic anhydride groups of the HDPE-g-MA molecules was the reason for the higher tensile stress. The grafted MA groups on the HDPE molecules were capable of ring opening to form succinic acid, which could have possibly reacted further with oxirane groups in the ENR molecules, as proposed in reaction Scheme 5. The HDPE backbone in the HDPE-g-MA was capable of compatibilizing with the HDPE in the blend component. Consequently, the blend with this type of compatibilizer generated higher chemical interactions between the ENR molecules and the HDPE molecules.

Two types of phenolic-modified HDPE compatibilizers (i.e., PhHRJ-PE and PhSP-PE) were also capable of reacting with ENR, as proposed in the mechanism in reaction Scheme 6, where R' is a methylol group



Scheme 3 Proposed grafting mechanism of linear HDPE and MA during the melt reactive process.



Scheme 4 Proposed grafting mechanism of HDPE with side groups and MA during the melt reactive process.

(CH₂OH). These two types of compatibilizers had the same end functional groups. Therefore, they reacted with ENR in a similar way. However, the blend with PhHRJ-PE, as shown in Figure 3, exhibited a higher tensile stress than the blend with PhSP-PE. This was attributed to differences in the molecular structures of the two types of phenolic resins (i.e., SP-1045 and HRJ-10518). SP-1045 consisted of both methylene and ether-bridged monomeric units, whereas HRJ-10518 contained only the methylene bridge. This caused the compatibilizer based on SP-1045 (i.e., PhSP-PE) to exhibit a higher chain flexibility. Also, the HDPE segments in the compatibilizer molecules were possibly miscible with the HDPE in the blend component.

Figures 4 and 5 show the tensile strength and elongation at break calculated from the respective

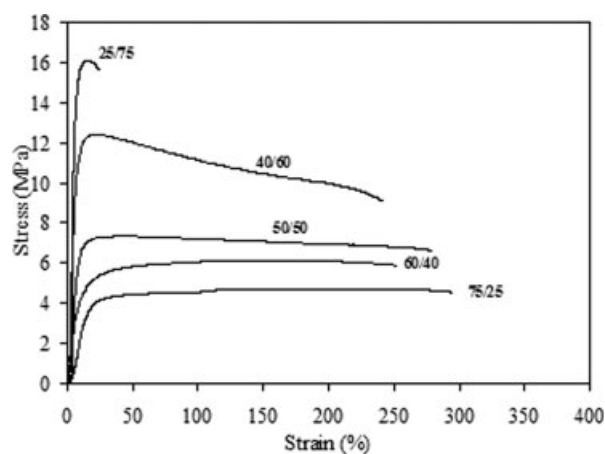


Figure 2 Stress-strain curves of TPEs based on ENR-20/HDPE blends at several blend ratios without compatibilizer.

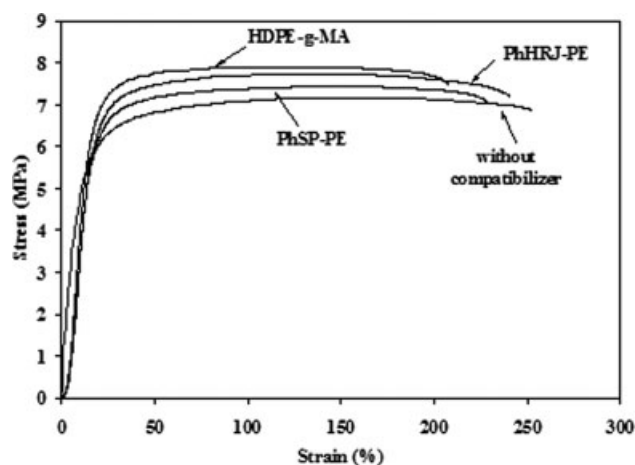
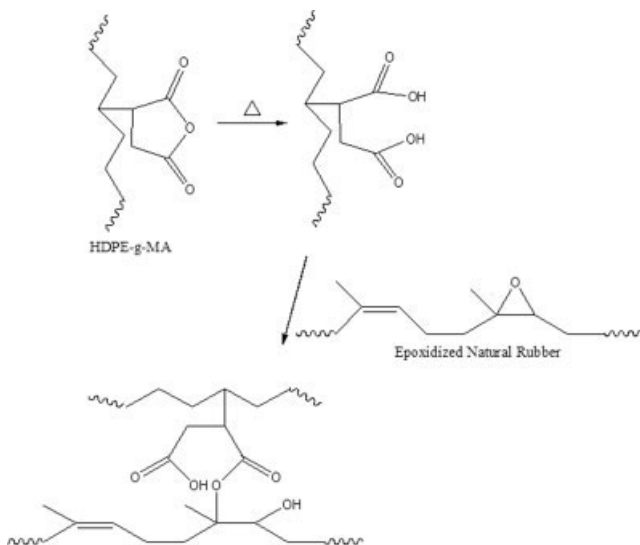
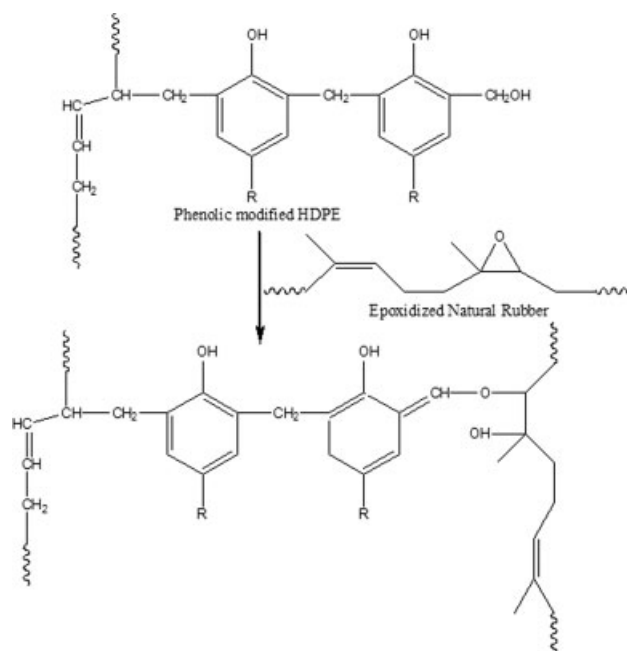


Figure 3 Stress-strain curves of TPEs based on 60/40 ENR-20/HDPE blend with various types of compatibilizer.

failure stresses and strains of ENR-20/HDPE blends at various blend ratios and types of blend compatibilizers. The tensile stress decreased, whereas elongation at break increased, with increasing quantities of ENR in the blend composition. The decreasing trend of tensile strength was attributed to a decrease in the HDPE content. This component exhibited higher strength properties of semicrystallizing thermoplastics. An increase in ENR in the blend ratio caused a lower level of the HDPE and a decrease in the degree of crystallinity of HDPE. This caused a higher capability for extending the specimens, as observed in Figure 4. When the tensile strength and elongation at break were compared at a given quantity of ENR in the ENR-20/HDPE blend, we observed that the blend without compatibilizer showed the lowest ten-



Scheme 5 Proposed mechanism of the compatibilization of ENR/HDPE by HDPE-g-MA.



Scheme 6 Proposed mechanism of the compatibilization of ENR/HDPE by phenolic-modified HDPE.

sile but the highest elongation at break. Therefore, the incorporation of blend compatibilizers contributed to the improvement of the strength properties. Set properties in terms of tension set were also improved by use of the blend compatibilizer, as shown in Figure 6. Also, the blends with PhHRJ-PE and PhSP-PE compatibilizers showed a similar value of tensile strength because of similar molecular characteristics. However, the blend with HDPE-g-MA showed the highest tensile strength. This was attributed to a crosslinking formation in the HDPE phase because of the reaction of residual DCP in HDPE-g-MA with HDPE molecules at high temperatures.

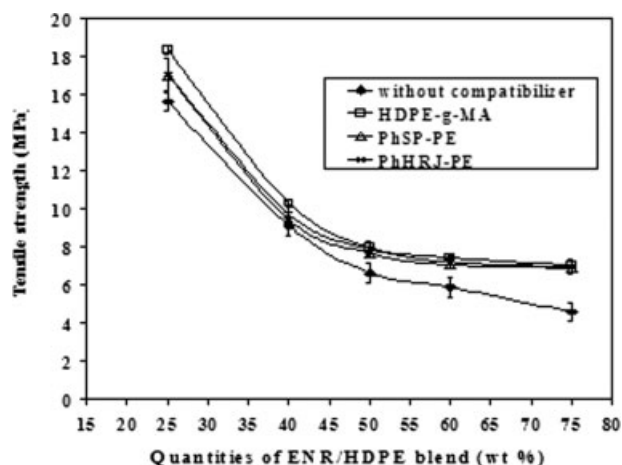


Figure 4 Tensile strength of TPEs based on ENR-20/HDPE blends with various types of compatibilizers and blend ratios.

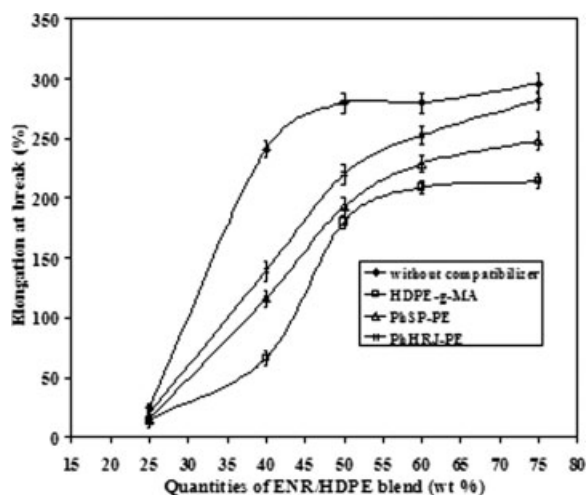


Figure 5 Elongation at break of TPEs based on ENR-20/HDPE blends with various types of compatibilizers and blend ratios.

Tension set properties were measured with specimens with an elongation at break higher than 100%, as the results show in Figure 6. The blend without compatibilizer showed the highest tension set, whereas the blend with PhHRJ-PE exhibited the lowest values. The blends with PhSP-PE and HDPE-g-MA showed intermediate values of tension set. Therefore, the tension set was ordered by type of blend compatibilizer as follows: PhHRJ-PE < PhSP-PE < HDPE-g-MA < Without compatibilizer. So, the incorporation of compatibilizers caused an increase in the elastomeric nature of the blend (i.e., lower in tension set). A chemical interaction between the two phases was the reason for the enhancement of the strength and set properties. That is, the functional groups in the blend compatibilizer reacted with the functional groups in the ENR molecules (Schemes 5

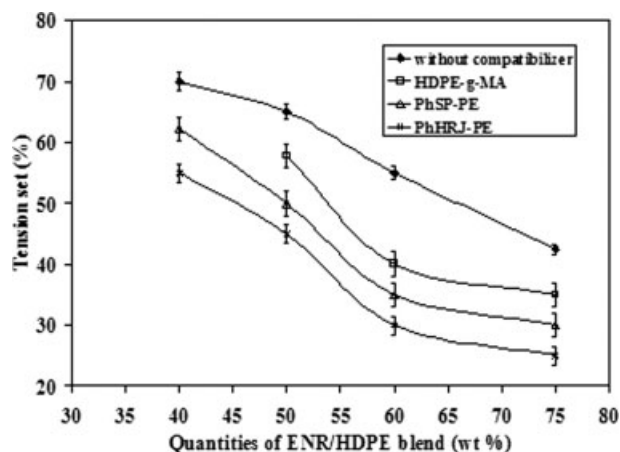


Figure 6 Tension set of TPEs based on ENR-20/HDPE blends with various types of compatibilizers and blend ratios.

and 6). Also, the HDPE segments were capable of being miscible with the HDPE in the blend component. The tensile and set properties agreed with the results from our previous study on the use of PhSP-PE and PhHRJ-PE to enhance the properties of TPE based on NR/HDPE blends.¹⁹

Figure 7 shows the hardness properties of the blends of ENR-20/HDPE. The hardness decreased with increasing content of ENR, which was a soft phase. At the same level of ENR used in the blends, the blend with HDPE-g-MA showed the highest level of hardness, whereas the blend without compatibilizer exhibited the lowest value. This was attributed to the HDPE crosslink formation because of residual DCP, as mentioned earlier. Therefore, the hardness of the blends with various types of blend compatibilizers could be ordered as follows: HDPE-g-MA > PhSP-PE > PhHRJ-PE > Without compatibilizer.

Morphological properties

The etched cryogenic fracture surfaces based on 60/40 ENR-20/HDPE blends with various types of blend compatibilizers are shown in Figure 8. The elastomer phase was preferentially extracted with chloroform. As a result, ENR was dissolved at an elevated temperature. Therefore, cavitations occurred at a location of the ENR phase after the etching process. The two-phase morphology was obvious. Also, at this blend ratio, the rubber and thermoplastic were both continuous phases; that is, there was a cocontinuous phase morphology. Also, smaller cavitations or phase sizes of for the blend with compatibilizers were observed. Therefore, the incorporation of the blend compatibilizers caused a smaller phase morphology in the blends. This was attributed to the

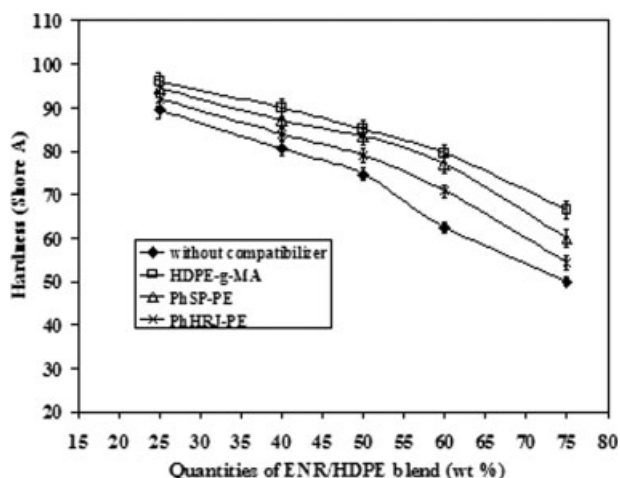


Figure 7 Hardness of TPEs based on ENR-20/HDPE blends with various types of compatibilizers and blend ratios.

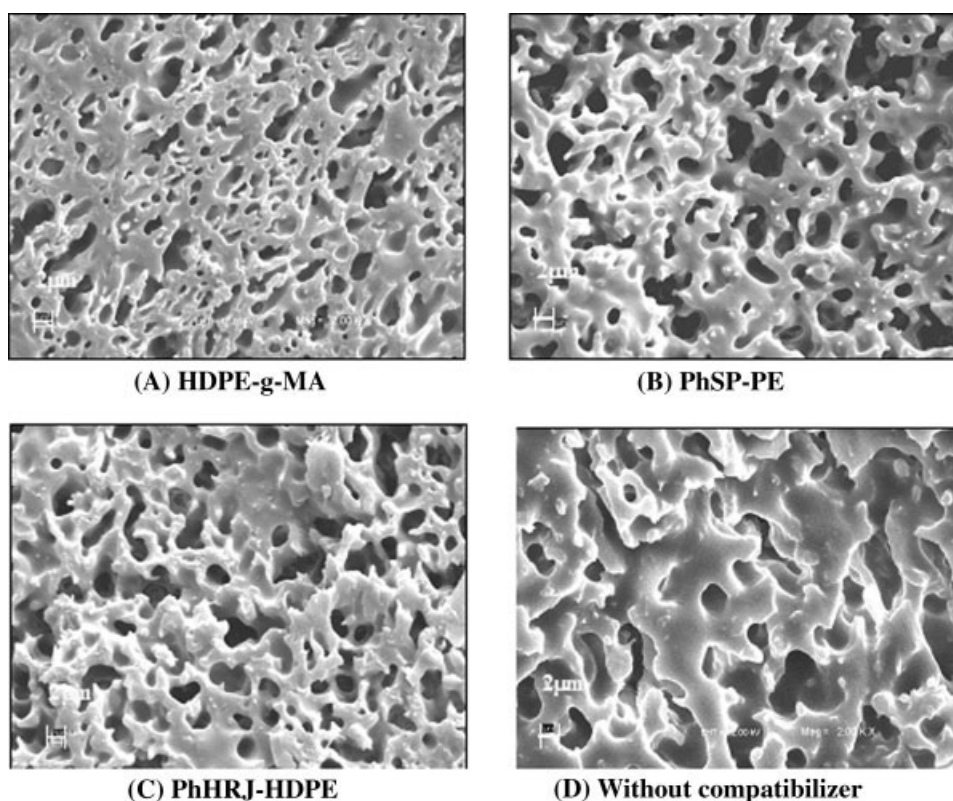


Figure 8 Scanning electron micrographs of TPEs based on 60/40 ENR-20/HDPE blends with various types of compatibilizers.

interaction between the ENR and HDPE phase via compatibilizer molecules. This caused a higher interfacial area and, hence, interfacial forces between the phases. The blend with HDPE-g-MA showed the smallest phase morphology. Therefore, it exhibited the highest strength properties (Fig. 4). The blend with PhSP-PE and HRJSP-PE exhibited a similar size but a slightly larger phase morphology than that of the blend with HDPE-g-MA. This caused these two types of the blends to exhibit lower strength properties than that of the blend with HDPE-g-MA compatibilizer. However, the blend with phenolic-modified compatibilizers exhibited a higher tendency to recover from prolonged elongation than that of the blend with HDPE-g-MA (Fig. 6). This was attributed to the crosslink structure of HDPE in the blend with HDPE-g-MA, which caused rigidity in the blend.

CONCLUSIONS

ENR-20/HDPE blends with various types of blend compatibilizers exhibited superior tensile, hardness, and set properties than blends without a compatibilizer. This was attributed to the interaction between ENR and HDPE molecules via a link of compatibilizer molecules. They contained polar functional groups (i.e., succinic anhydride, succinic acid, or

phenolic groups), which could have possibly reacted with oxirane groups in the ENR molecules. Also, the HDPE segments in the compatibilizer molecules were prone to compatibilize with the HDPE molecules in the blend components. Among the blend compatibilizers, HDPE-g-MA gave the highest strength properties but the lowest tension set properties. This was a result of the crosslink formation of HDPE because of the reaction of residual DCP in the system. A high strength and rigid material were consequent. The 60/40 ENR-20/HDPE blend exhibited a cocontinuous phase morphology. Smaller phase sizes were observed in blends with a high level of interaction, such as the blend with HDPE-g-MA. This corresponded to the greater hardness and strength properties of this type of material

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