

Crumb Tire Rubber and Polyethylene Mutually Stabilized in Asphalt by Screw Extrusion

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ABSTRACT: In order to put recycled rubber and plastics into practical use for asphalt's modification, the polyethylene (PE)/crumb tire rubber (CTR) blends were blended to prevent them from phase separation in asphalt during hot storage. The effect of screw extrusion on storage stability, morphology, and thermal properties of modified asphalt were investigated. The results showed that the storage stability of PE/CTR modified asphalt was significantly improved after screw extrusion in the presence of a compatibilizer. The extruded PE and CTR were more finely and uniformly dispersed in the asphalt. The rheological properties of modified asphalts were improved at both higher and lower temperature's range. The density discrepancy between asphalt and the extruded PE/CTR particles was neutralized by the combination between PE and CTR. The interactions among PE, CTR and asphalt were improved by the compatibilizer. Both the reduction of density difference and the enhanced interactions are critical to obtain a stable modified asphalt system. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 41189.

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INTRODUCTION

Asphalt is widely used for road paving because of its viscoelastic properties and excellent adhesion with mineral aggregates. However, asphalt is susceptible to temperature's variation, which leads to some types of pavement distress, such as permanent deformation, rutting, and load fatigue cracking. Increasing vehicle loads and traffic volumes also draw researcher's attention to developing modifiers to meet these pavement demands. Polymers are usually selected to modify asphalt for improving the pavement's performance.¹⁻⁴

Polymers employed in the asphalt's modification can be classified into three categories: plastic, thermoplastic elastomer, and rubber. Polyethylene (PE) from the first category confers asphalt with longer fatigue life and increased rutting resistance.⁵⁻⁸ However, polyethylene modified asphalt is a thermo-dynamically unstable blend, and phase separation occurs during storage at high temperatures.⁹ One of the approaches to improve the storage stability is to introduce reactive polymers with functional groups, which react with asphalt compositions in the blends.^{10,11} Frequently used reactive polymers are thermoplastic polymers grafted with maleic anhydride (MA) and ethylene-based copolymers containing epoxy rings. However, the reaction between the polymers with asphalt is often uncontrollable. The cost of reactive polymer is too high to

make it into practical use. Crumb tire rubber (CTR) can furnish asphalt with excellent flexibility, noise reduction, reduced load fatigue cracking, decreased permanent deformation besides other desirable qualities.^{12,13} Substitution of virgin material with recycled tire rubber will contribute to solving a waste disposal problem and reducing overall cost of tire disposal. However, the settlement of crumb rubber in the asphalt is still a disadvantage for the widespread use of CTR modified asphalt.

Polyethylene and/or crumb rubber as asphalt modifiers have been extensively investigated, while few study deal with the combination of both by using screw extrusion. Navarro et al. found that the mechanical properties of asphalt could be enhanced by using polyethylene and rubber composite.¹⁴ Polyethylene mainly improves the high-temperature performance, while rubber improves the low-temperature performance.^{14,15} Because of the disparity of properties between polymer and asphalt, such as polarity, molecular structure, solubility parameters, the mixture exhibits the tendency to phase separation during hot storage.¹⁶

In order to stabilize the polyethylene or crumb rubber in asphalt, the polyethylene and crumb rubber were compounded by screw extrusion, and the blends were used to modify asphalt. The mechanical, rheological, and thermal properties of the blend were evaluated in addition to hot storage stability. The

Table I. Recipes and Calculated Density of Each BER Blends

Sample	Content (phr)			Calculated density (g/cm ³)
	HDPE	CTR	LLDPE-g-MA	
BER370	30	70	0	1.08
BER371	30	70	1	1.08
BER373	30	70	3	1.07
BER375	30	70	5	1.07
BER553	50	50	3	1.04
BER733	70	30	3	1.00

chemical reaction between asphalt and the compatibilizer was investigated by Fourier Transform Infrared (FTIR).

EXPERIMENTAL

Materials

Asphalt was provided by China National Petroleum Corporation. Softening point temperature was 45.3°C, penetration at 25°C was 70.1 (0.1 mm).

High density polyethylene (HDPE, 5070EA, density 0.94 g/cm³) was supplied by CNGC Huajin Chemicals Co., Ltd. LLDPE (DFDA-7042N) was manufactured by Jilin Petrochemical Corporation. Crumb tire rubber (30 mesh, density 1.15 g/cm³) was firstly ground from whole truck tire rubber at ambient temperature and then processed by dynamic devulcanization at 220°C for 2 h in a tank with stirring. The treated crumb tire rubber was provided by Shanghai Lujia Co., Ltd. The characteristics of this crumb rubber are as following: acetone extraction is 19.3%, the soluble rubber is 7.6%, gel content is 73.1%, and swelling ratio by toluene is 3.85. MA-grafted linear low density polyethylene (LLDPE-g-MA) with grafting degree of 0.8–1.0% was supplied by Shanghai Jiuju Co., Ltd.

Preparation of PE/CTR Blend Modified Asphalts

Step 1: Blended PE/CTR (BER) with different content of LLDPE-g-MA and different ratios of PE to CTR were prepared by screw extrusion. The compounds were extruded at 180°C using Twin Screw Extruder (ZE 25A, made by Berstorff GmbH, Germany), followed by pelletizing for use in asphalt modification. The recipes of each blend of BERs are listed in Table I.

Step 2: Asphalt was heated till fluid in an iron container. Then upon reaching at about 180°C, BER (15 wt% of asphalt ternary blend) was added to the asphalt. The ternary blends were blended by using a high shear mixer (made by Shanghai Aidong Electrical Equipment Co. LTD) at 180°C with a shearing speed of 4000 rpm for 30 min.

Conventional Properties and High-Temperature Storage Stability

The conventional properties of modified asphalt like penetration, softening point, and ductility were carried out according to GB/T0604-2011, GB/T0606-2011, and GB/T0605-2011, respectively. The high-temperature storage stability was evaluated through the following procedure. The samples of prepared blends were transferred into an aluminum toothpaste

tubes (25.4 mm in diameter, 140 mm in height). The tubes were sealed and stored vertically in an oven at 163°C for 48 h. After that, the samples were cooled down to room temperature. The aluminum tubes were then cut horizontally into three equal parts. The storage stability of the BER modified asphalt was evaluated by measuring the difference in softening points (ΔSP) between the top and bottom sections. If the value was less than 3.5°C, the blend was considered as stable.

Morphological Analysis

A small drop of asphalt was placed between two heated microscopic glass slides and squeezed to form a thin film. The morphology of the asphalt ternary blends was observed under an optical microscope (Leica, Germany) by using common light source with a magnification of 100 times.

Rheological Analysis

The viscous and elastic behaviors of asphalt ternary blends were characterized by Rotational Rheometer (AR-G2, TA Instrument, USA), using parallel plate geometries (60 mm in diameter) at 1 Hz in temperature range of 50–100°C at a heating rate of 1°C/min.

Dynamic Mechanical Analysis (DMA) tests were carried out in a NETZSCH device, using 15 × 10 × 3 mm samples in single cantilever mode. The liquid nitrogen was used as coolant. All tests were carried out at a frequency of 1 Hz and a strain within the linear viscoelastic region. The temperature ranged from –60°C to 10°C at a heating rate of 2°C/min.

Thermal Analysis

Thermal analysis of asphalt ternary blends was performed by Differential Scanning Calorimeter (DSC, Perkin Elmer, Inc., USA) in a nitrogen atmosphere at a flow rate of 50 mL/min. The data were collected from 40°C to 200°C at a heating rate of 10°C/min.

Interaction Analysis Between Compatibilizer and Asphalt

About 4 g of asphalt and 1 g of LLDPE-g-MA were added into 100 mL toluene in a 250 mL flask fitted with reflux condenser, a mechanical stirrer, and a thermocouple. This mixture was heated and stirred constantly. After 5 h, the reacted product was put into an evaporating dish to vaporize the solvent. The resulting samples were extracted by tetrahydrofuran using Soxhlet Extractor for 24 h. Control tests were carried out by using LLDPE substitute LLDPE-g-MA.

Fourier Transform Infrared (FTIR) Analysis

The samples obtained from the above reaction procedure were heat-compressed into a thin film to conduct FTIR tests using a spectrophotometer (Spectrum 100, Perkin Elmer, Inc., USA), with a resolution of 4 cm⁻¹ and within a wavenumber range of 400–4000 cm⁻¹.

RESULTS AND DISCUSSION

Conventional Properties and High-Temperature Storage Stability

The conventional properties of PE/CTR modified asphalts were shown in Table II. It can be seen that the softening points, ductility and penetration of PE/CTR modified asphalts almost keep no change as increasing the content of LLDPE-g-MA. The ratio

Table II. Effect of LLDPE-g-MA Content on Conventional Properties and Storage Stability of Ternary Asphalt Blends

Sample	Softening point (°C)	Ductility (5°C)/cm	Penetration (25°C)/0.1 mm	Softening points (°C)		
				Top	Bottom	ΔSP
BER370	67	3.8	46.3	78	56	22
BER371	66	4.1	48.6	70	58	12
BER373	65	3.7	43.1	65	62	3
BER375	65	3.7	39.2	63	65	-2
BER553	75	1.6	25.8	72	71	1
BER733	82	1.5	16.0	77	62	15

of PE/CTR obviously influences the conventional properties. The softening points of PE/CTR modified asphalts increase with increasing the ratio of PE in PE/CTR, however, the ductility and penetration show a sharp decrease.

As shown in Table II, the control sample (BER370) exhibits high phase separation at high temperature, which is confirmed by ΔSP value of 22°C. The ΔSP values decrease with increasing LLDPE-g-MA content, which indicates that LLDPE-g-MA act as a compatibilizer to improve the hot storage stability. During the process of BER extrusion, dipolar interactions would take place between HDPE and CTR due to the addition of LLDPE-g-MA.^{17–19} These interactions will help combine the polyethylene and crumb tire rubber together, which prevent the BER from phase separation. As shown in Table II, the hot storage stability of the modified asphalt also depends on the ratio of PE to CTR, too much PE (e.g., 70/30) in the PE/CTR blends will lead to a phase separation. This indicates that except the combination of PE and CTR (Figure 1), the density discrepancy between asphalt and extruded PE/CTR particles is also very important for the storage stability. According to the calculated densities of

PE/CTR blend in Table I, PE/CTR (70/30) blend is 1.00 g/cm³, which makes it easily float to the top of the modified asphalt because of swelling by the light fraction of the asphalt.

Optical Microscopy

The effect of LLDPE-g-MA contents on morphologies of modified asphalts is shown in Figure 1. It can be clearly seen that large and irregular shaped HDPE-rich phase disperses in the brown asphalt-rich phase, which indicates the immiscibility among CTR, HDPE, and asphalt. With even a very low concentration of LLDPE-g-MA (1 phr, BER371), a completely different morphology is obtained [Figure 1(b)]. Spherical HDPE particles combine with irregular shaped CTR disperse in the brown bituminous phase, indicating a good interfacial adhesion between HDPE and CTR. With the increase of LLDPE-g-MA concentration (BER373, BER375), the size of HDPE droplets further decrease, as shown in Figure 1(c) and (d). These result in a significant enhancement of storage stability of asphalt ternary blends, which is evident in blends BER373 and BER375 with ΔSP value of 3°C and -2°C, respectively. Comparison of the morphology of Figure 1(c) and (d), BER375 shows an

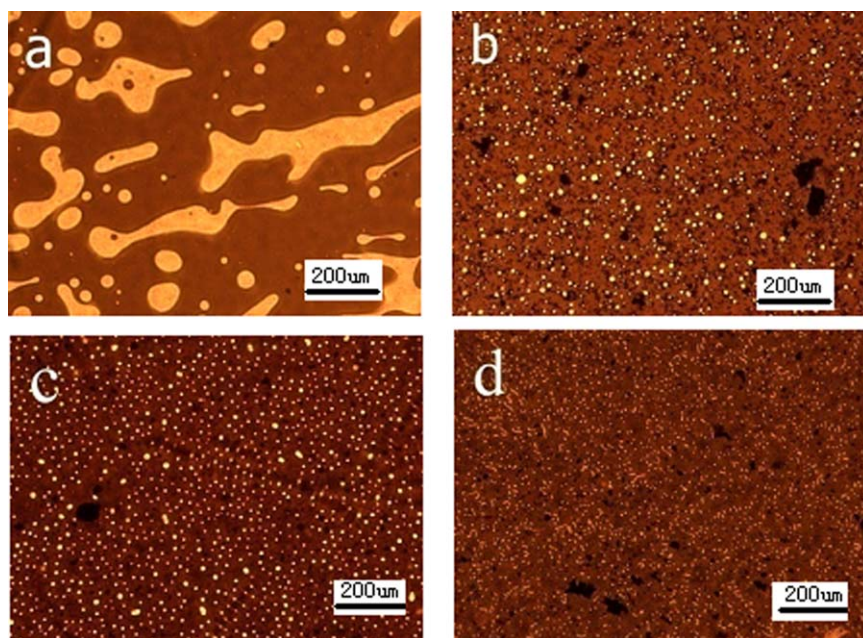


Figure 1. Optical photomicrographs of asphalt ternary blends modified with different BERs. (a)BER0, (b) BER 1, (c) BER 3, (d) BER5. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

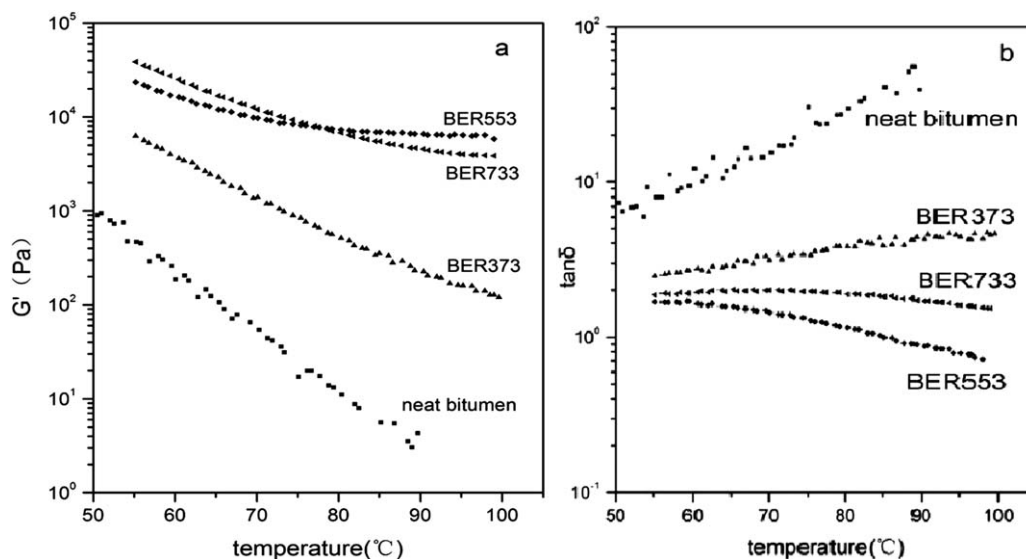


Figure 2. Effect of the different ratio of HDPE to CTR on the viscoelastic properties of asphalt ternary blends at higher temperature. (a) Storage modulus, (b) Loss tangent.

interconnected network. These morphological analyses support the assumption that LLDPE-g-MA act as a good compatibilizer in modification system.

Rheological Analysis

The effect of HDPE/CTR ratio on viscoelastic properties of the asphalt ternary blends were characterized to gain more insight into the mechanical response of the material. As can be seen from Figure 2(a), the storage modulus increases with increasing ratio of HDPE to CTR, they are much higher than that of asphalt. Such increase supports that BER contributes to improving high-temperature properties of modified asphalt. Compared to the asphalt, a significant decrease in the loss tangent values ($\tan \delta$) is obtained for asphalt ternary

blends [Figure 2(b)], indicating the BER modifier enhances the elasticity of asphalt blends.²⁰ Moreover, as the ratio of HDPE to CTR increases, the slope of $\tan \delta$ to temperature decrease [Figure 2(b)], illustrating that temperature susceptibility of asphalt ternary blends decrease with increasing HDPE ratio.^{14,21} The enhanced elasticity and reduction in temperature susceptibility indicate a greater degree of intermolecular association within asphalt ternary blends.²² Finally, a G' crossover between BER553 and BER733 was observed around 75°C [Figure 2(a)] and $\tan \delta$ value of BER733 modified asphalt (HDPE concentration of around 10.5%, CTR of 4.5%) was higher than BER553 modified asphalt (HDPE concentration of around 7.5%, CTR of 7.5%). This data illustrates that HDPE seems to influence the BER/asphalt binder's

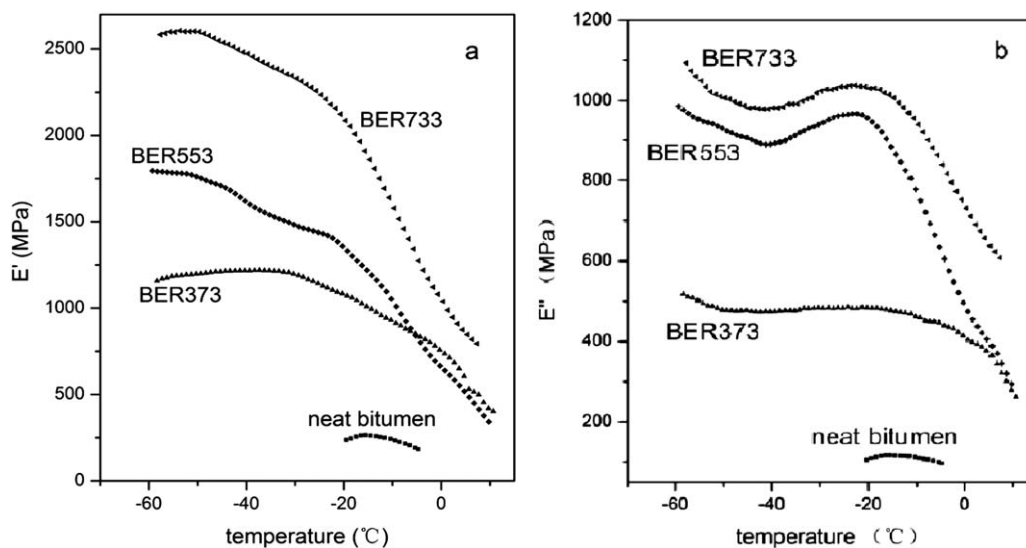


Figure 3. Effect of the different ratio of HDPE to CTR on the viscoelastic properties of asphalt ternary blends at lower temperature. (a) Elastic modulus, (b) loss modulus.

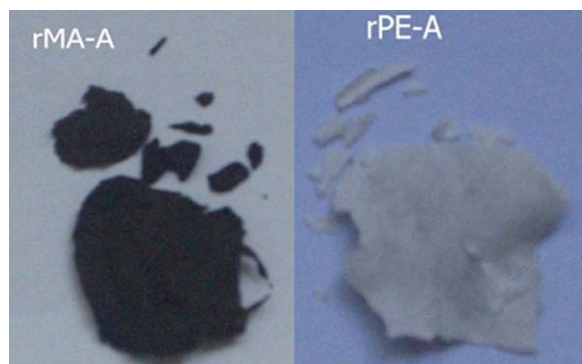


Figure 4. Physical appearance of rMA-A and rPE-A. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

elasticity below 75°C. However, CTR exerts a major influence on material elasticity. These results can be explained on the basis of the HDPE and CTR rheological properties. As we know, the modulus of polymer would decrease with increasing temperature especially at higher temperatures (30–100°C). While CTR is partially vulcanized rubber which behaves more like inert filler, the reduction of modulus with temperature is less obvious than HDPE.

As shown in Figure 3, the viscoelastic properties of asphalt ternary blends depend on the relative proportion of each polymer in the lower temperature range. The larger ratio of HDPE to CTR, the higher the viscoelastic values of modified asphalt. This may contribute to a better relaxation of stress in paving application.²³ Such increase is more evident as BER improve material performance at low temperature. An apparent glass transition-behavior is around -20°C for BER553 and BER733. Comparing modified asphalts of BER553 and BER733, the higher CTR content (for BER553 modified asphalt), the lower the glassy region. This also implies that CTR improve asphalt's lower in-service temperature performance.

FTIR Analysis

The LLDPE-g-MA/asphalt was named as rMA-A (r means reaction), and LLDPE/asphalt as rPE-A. As shown in Figure 4, the rMA-A sample of black color can be considered as the compos-

ite of asphalt and polymer, while the other white one only contains pure LLDPE. It illustrates that chemical bonds generated between LLDPE-g-MA and asphalt during reaction procedure. The interaction occurs in the two samples were characterized by FTIR shown in Figure 5.

The spectrum of LLDPE-g-MA shows characteristic cyclic anhydride peaks at 1788 cm^{-1} and carboxylic acid groups (due to partially hydrolysis) at 1712 cm^{-1} . These bonds are significantly reduced in rMA-A and a new small and wide absorption presents at 1592 cm^{-1} , which could also be seen in asphalt. In addition, a small peak at 1537 cm^{-1} , belongs to the O=C–N stretching vibration of amides is observed in rMA-A. These results indicate that cyclic anhydride opened and reacted with some nitrogen-components of asphalt during reaction procedure, further evidenced by the newly-generated peak in 802 cm^{-1} in rMA-A. The special peak at 1063 cm^{-1} of LLDPE-g-MA transformed into two peaks at 1092 and 1025 cm^{-1} (S=O) after reacting with asphalt.

Based on the FTIR analysis of rPE-A in Figure 5, except for various groups of $-\text{CH}_2-$, $-\text{CH}_3$, $-\text{CH}$, and $-\text{OH}$ of LLDPE, numerous peaks also appeared in rPE-A. The peaks at 1166 and 1102 cm^{-1} are assigned to the stretching vibration of S=O group. These peaks presented at around 950 – 1050 cm^{-1} are attributed to ether group. Above mentioned new peaks may relate to the absorption of low-molecular component in asphalt of LLDPE.

Thermal Analysis

In order to verify the interaction between LLDPE-MA and asphalt, DSC curves of asphalt ternary blends with different LLDPE-g-MA content were shown in Figure 6. The effect of LLDPE-MA on the melting points (T_m), heat of fusion (ΔH_f) of the modified asphalt is shown in Table III. All the PE's melting peaks of modified asphalts are shifted from 131.2°C to around 120°C . The swelling of PE yield a smaller crystalline phase,²⁴ and the light molecular weight components are mixed with a homogeneously crystalline polymer.²⁵

As shown in Table III, the heat of fusion of all asphalt samples are larger than calculated value (4.5% of pure HDPE $\Delta H_f = 7.241\text{ J/g}$). However, the discrepancy between observed

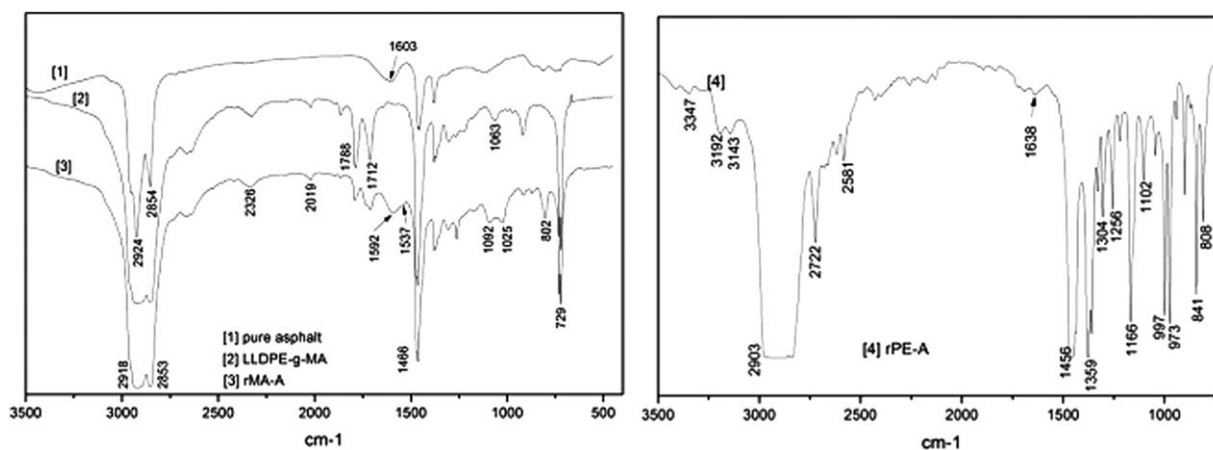


Figure 5. Infrared spectra of LLDPE-g-MA/asphalt reacted product and control LLDPE/asphalt reacted product at 110°C .

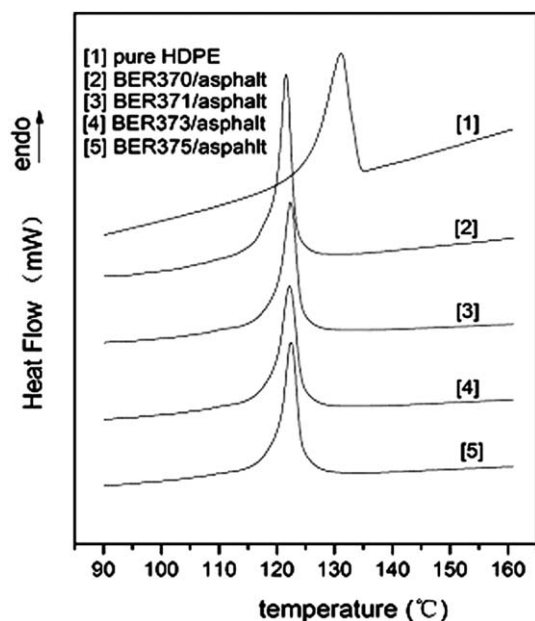


Figure 6. DSC curves of HDPE and asphalt ternary blends with different LLDPE-g-MA content.

and calculated ΔH_f of BER1 modified asphalt has a bit of large error range due to the serious phase separation⁷; so here we just compare the last three samples. With increasing LLDPE-g-MA content, ΔH_f of modified asphalt increases. It implies that greater amount of heat is needed to fuse the polymer in asphalt matrix, which may be caused by the change of crystallinity of HDPE.

Hypothesis of Stabilization Mechanism

Generally speaking, asphalt can be divided into Saturate, Aromatic, Resin and Asphaltene (SARA) components according to their polarity. The saturate fraction consists of nonpolar material including linear, branched, and cyclic saturated hydrocarbons (naphthenes). Aromatics, which contain one or more aromatic rings, are slightly more polarizable. The remaining two fractions, resins and asphaltenes, have polar substituents. When

Table III. Effects of LLDPE-MA Contents on Thermo Behavior of Asphalt Ternary Blends

Sample	HDPE content (%)	LLDPE-MA content (%)	T_m (°C)	ΔH_f (J/g)
Pure HDPE	100	0	131.2	160.9
BER370/asphalt	4.5	0	121.6	9.1
BER371/asphalt	4.5	0.15	122.4	7.3
BER373/asphalt	4.5	0.45	122.1	8.6
BER375/asphalt	4.5	0.75	122.4	9.2

PE and CTR were put into the asphalt, PE is swollen by absorption of the nonpolar low-molecular weight components,²⁶ while CTR mainly interact with aromatic fraction according to their polarity.

Based on the above discussion, hypothesis of the stabilization mechanism of asphalt/HDPE/CTR system is shown in Figure 7. The stabilization of PE or CTR in asphalt can be analyzed in terms of thermodynamic, kinetic, and steric effects. Firstly, the thermodynamic stabilization of polymer in the asphalt can be improved by increasing the adhesion between polymer and asphalt. Some chemical interactions occur between the functional groups of BER, such as maleic anhydride, double-bond of CTR, carboxyl groups of carbon black, and active groups in asphalt like amino-group, hydroxyl, and carboxyl groups. Secondly, the kinetic stabilization of polymer particles can be improved according to Stoke's law, reducing the size of polymer, decreasing the density difference between polymer and asphalt, and increasing the viscosity of asphalt are all helpful for the stabilization. During the processing, BER, LLDPE-g-MA can enhance the compatibility of HDPE and CTR due to the interaction between maleic anhydride groups and the functional groups of both carbon black and CTR.²⁷ Devulcanization and mechanical milling also promoted the interfacial interaction between CTR and HDPE.^{28,29} The size of PE and CTR could be greatly reduced by compatibilization of LLDPE-g-MA and screw extrusion. The reduction of density difference is mainly due to

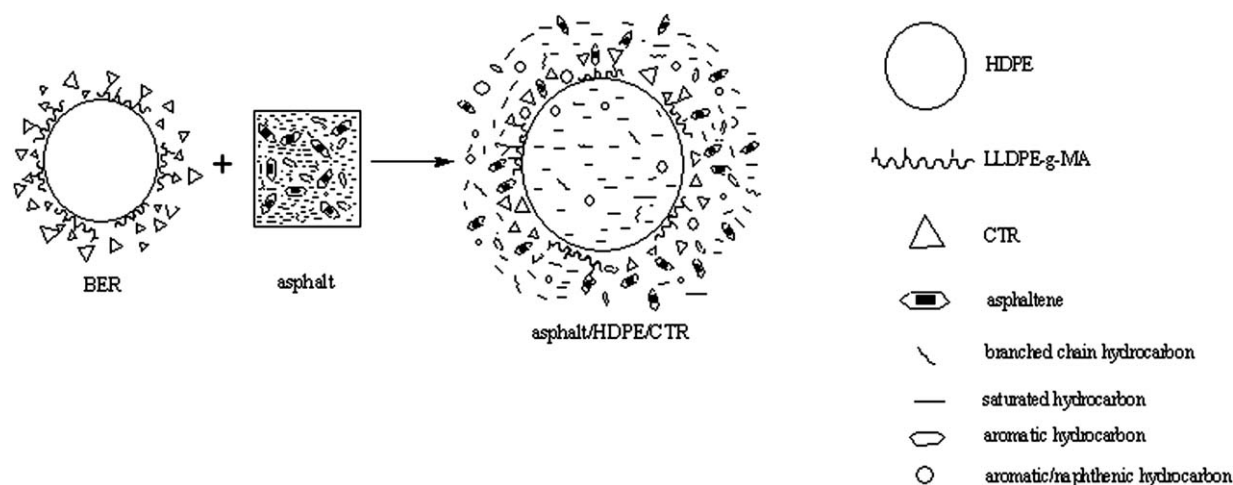


Figure 7. Schematic diagram of interaction in asphalt/HDPE/CTR system.

the buoyancy force of HDPE (0.94 g/cm^3) is offset by the gravitational force of CTR (1.15 g/cm^3), resulting in drop in the falling velocity,¹⁵ leading to more stable suspension of BER particles. Thirdly, the surface of PE was covered with the finely dispersed crumb rubber, which also prevents the coalescence of the PE particles in the asphalt. In conclusion, the PE and CTR could be stabilized in the asphalt through screw extrusion in the presence of compatibilizer in terms of thermodynamic, kinetic and steric effects.

CONCLUSIONS

The PE/CTR blends with excellent compatibility with asphalt were prepared by screw extrusion with the incorporation of LLDPE-g-MA as compatibilizer. The storage stability of modified asphalt samples can be enhanced through increasing the content of LLDPE-g-MA. The ΔSP value decreased and the morphology of polyethylene and crumb tire rubber became finer in the asphalt matrix. The elastic and loss modulus of the modified asphalts significantly increased and thermal susceptibility reduced at both low- and high-temperatures' range. Thus, the extruded PE/CTR modifier is verified as sufficiently to withstand rutting or permanent deformation at high temperature, and resist cracking at low temperature.

Thermal analysis and FTIR analysis revealed chemical interactions between anhydride maleic groups and functional groups in asphalt. Therefore, the incorporation of LLDPE-g-MA is critical to prevent polyethylene or CTR from phase separation in asphalt at high temperature.

Finally, a hypothesis of stabilization mechanism is presented. Chemical interactions and changes in density discrepancy are considered as two main reasons to remain stable at high temperature for the extruded blend modified asphalts.

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