Effect of linear low density-polyethylene grafted with maleic anhydride (LLDPE-g-MAH) on properties of high density-polyethylene/styrene–butadiene–styrene (HDPE/SBS) modified asphalt

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HIGHLIGHTS

• A kind of asphalt modifier was prepared by melt blending.
• The low-temperature performance of HDPE/SBS modified asphalt was improved.
• Softening points difference between the top and bottom sections of samples were sharply reduced.
• LLDPE-g-MAH had no significant effect on high-temperature property and rheological character.

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ABSTRACT

A kind of modifier composed of high density-polyethylene (HDPE) and styrene–butadiene–styrene (SBS) was prepared by melt blending with different contents of linear low density-polyethylene grafted with maleic anhydride (LLDPE-g-MAH). HDPE/SBS modified asphalt composites were prepared. Effects of LLDPE-g-MAH on classical performance, hot storage stability, and dynamic rheological behaviors of HDPE/SBS modified asphalt were investigated. Compared with samples without LLDPE-g-MAH, penetration and ductility of HDPE/SBS modified asphalt were both increased, in the meantime, softening point and the maximum failure temperature were decreased. The research results showed that low-temperature performance was improved, while LLDPE-MAH had no positive effect on high-temperature property. In addition, softening point difference between the top and bottom sections of samples was sharply reduced. A fairly homogeneous dispersion system of the modifiers in the asphalt matrix was observed by microscope. A conclusion that LLDPE-g-MAH had no significant effect on high-temperature property and rheological character can be drawn from the rheological studies.

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1. Introduction

In recent years, with the increasing volume of traffic, larger-scaled vehicles and the increasing proportion of overloading vehicles, the quality requirement for the road surface of modern transportation is higher and higher. Ideal performance demands that the binder must be able to withstand low temperatures and the resulting thermal stresses that develop pavement shrinks, resist repeated loading and unloading without exhibiting fatigue failure, withstand loading to prevent permanent deformation, and be capable of being compacted, transported, and stored at safe temperatures [1,2].

From this perspective, application properties of ordinary oil asphalt cannot accommodate the increasing traffic intensity, axle loads and meet the demand for comfort, because of its severe temperature susceptibility. Therefore, it is essential to modify asphalt [3,4]. Polymer modified bitumen (PMB) has been growing rapidly in road paving applications over the last decade [5]. Polyethylene (PE) has been found to be one of the most effective polymer additives with reducing the rutting under heavy loads. Unfortunately, the high crystalline nature of PE makes badly compatible with asphalt. PE particles separate from the asphalt when stored at high temperature, which further limits the application of PE modified asphalts in paving [1].

From previous studies, we can know that the compatibility between polymer and asphalts and stability of modified asphalt depend on many factors, such as asphalt composition, polymer characteristics, preparation processes and so on. In order to resolve this problem, many researchers have been trying to find effective methods, such as adding of sulfur [6], incorporating silica into LDPE [4], chemical reactive blending [7], steric stabilizers [8]. On the
basis of the common physical modification of the asphalt, the chemical reactive blending and adding the graft copolymer were valuable to obtain better performance [9,10].

It is well known that plastomeric polymers can improve rut resistance, but they are inferior to elastomers [11,12]. In addition, plastomeric polymers cannot improve low-temperature performance of asphalt. Elastomers can improve fatigue resistance and cracking resistance, but it is limited to improve heat resistance. Thus, combining the advantages of plastomeric polymer such as PE, EVA and elastomers such as SBS, SBR and incorporation of these materials into PMA would be an effective method of recycling polymer to enhance the properties of asphalt pavement [13–17].

In the present work, high-density-polyethylene (HDPE) modified asphalts with good high-temperature storage stability were prepared by blending SBS and graft copolymer into HDPE and mixing into asphalt. We selected tentatively LLDPE-g-MAH as the graft copolymer to improve the compatibility between asphalt and HDPE, and to enhance interaction between modifiers. The effects of LLDPE-g-MAH on the classical properties, high-temperature storage stability, rheological properties of HDPE/SBS modified asphalt were analyzed.

2. Experiment

2.1. Materials

Base asphalt, with 60/80 penetration grade, was obtained from the Shenghua refinery in Shan Dong province, China. The base properties and chemical composition of base asphalt are shown in Table 1.

HDPE, Grade 5000S, was produced by Yanshan Petrochemical Company. SBS, Grade 791H, was provided by Raling Petro-Chemical Company of Sinopec Corp. Co., LLDPE-g-MAH was obtained from the market; its graft level is 0.9%.

The engineering properties of the HDPE and SBS are presented in Table 2.

2.2. Preparation of composite modifier

The composite modifier, with HDPE and SBS as the matrix, LLDPE-g-MAH as the compatibilizer with different content, carbon black (CB) as the filler in definite proportion by weight, was prepared by melt blending. HDPE, SBS, LLDPE-g-MAH and CB were transferred into the twin-screw extruder preheated at 165 °C first, extruding grain at a constant mixing speed of 30 r/min, then composite modifier was obtained and can be used in the following step.

2.3. Preparation of HDPE/SBS modified asphalt

The HDPE/SBS modified asphalts were prepared by three steps. First, the base asphalt was heated to 180 °C and melted in an iron container, the composite modifier of HDPE/SBS was added into the asphalt and stirred at a moderate or low speed at 160 °C for 0.5 h to obtain a finely dispersed minor phase. Then, the sample was heated to 180 °C and sheared at the rate of 4000 r/min for 40 min. During this process, a little sulfide was added into HDPE/SBS modified asphalt. Last, the sample was stirred at 140 °C for 20 min.

2.4. Performance measurements

Conventional performance was mainly composed of softening point, penetration, ductility measured according to Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering [18].

The storage stability of the modified asphalts was measured as following steps: the sample (50 g) was poured into an aluminum toothpaste tube (25 mm in diameter and 140-mm in height). The tube was sealed and stored vertically in an oven at 160 °C/C for 48 h, then cut horizontally into three equal sections, after it cooled to room temperature. The samples from the top and bottom sections were melt and placed in small molds labeled A and B to evaluate storage stability by measuring the softening points of them, respectively. If the difference of the softening points between A and B were less than 2.5 °C, the sample will be regarded to have good storage stability.

A drop of bitumen was first sandwiched between a slide, and a cover slip, and then put onto sample heater. Bitumen was heated and slowly pressed into a thin layer between slide and cover slip. Its distribution behavior was observed by using optical microscopy and micrographs were taken using a photo camera.

A dynamic shear coefficient (Anton paar made in Austria, Type H-PST1020) was used for dynamic mechanical analysis of asphalt binders. The viscous and elastic behavior of asphalt binders was characterized at intermediate and high service temperatures at a rate of 10 rad/s. The samples were “sandwiched” between two parallel plates with a diameter of 25 mm and a gap of 1 mm. The test temperature was raised by ramping rate of 6 °C one cycle, and the test was terminated when the G’/sin δ was equal to 1. Temperature sweeps were applied at a fixed frequency of 10 rad/s and variable strain. The rheological parameters were measured for calculating viscoelastic parameters such as complex modulus (G’), phase angle (δ), and rut factor (G’/sin δ).

3. Results and discussion

3.1. Effects of LLDPE-MAH on classical properties of HDPE/SBS modified asphalt

In technical specifications for construction of highway asphalt pavements, penetration and low temperature ductility are usually used to evaluate the low-temperature performance of asphalt. The effects of LLDPE-MAH on low-temperature properties of HDPE/SBS modified asphalt were illustrated in Table 3.

Compared with those modified asphalts in absence of LLDPE-g-MAH, such as sample D, penetration and ductility of the asphalt became higher after the addition of LLDPE-g-MAH, and the changes are more obvious with increasing LLDPE-g-MAH content, demonstrating that low-temperature properties of HDPE/SBS compound modified asphalt was improved by LLDPE-g-MAH and influence of LLDPE-g-MAH on low-temperature properties of HDPE/SBS compound modified asphalts was dependant on LLDPE-g-MAH content. At low LLDPE-g-MAH content, such as sample A, penetration and low temperature ductility increased only a little, showing that LLDPE-g-MAH had no obvious influence on low-temperature property of HDPE/SBS compound modified asphalt. When the LLDPE-g-MAH content was increased, such as sample B and C, the penetration and ductility of the modified asphalt increased significantly, indicating a better low-temperature property of these compound modified asphalts. But unfortunately, even when LLDPE-g-MAH content was increased to 0.4%, penetration and ductility of the compound modified asphalt was still lower than base asphalt. This was dependant on HDPE and SBS, as we all know that, HDPE, as plastomeric polymer, cannot improve even destroy low-temperature performance of asphalt.

Effects of LLDPE-MAH on High-temperature property of HDPE/SBS modified asphalt were showed in Table 4 and Fig. 1. According to Strategic Highway Research Program (SHRP), softening point and the maximum failure temperature (Tshsp) are usually used to characterize the high-temperature property of asphalt. The maximum failure temperature is calculated by the complex modulus divided by the phase angle, and its value is the corresponding temperature when G’/sin δ is equal to 1.0 KPa [4,14]. The higher the softening point or corresponding temperature, the better high-temperature property. The above analysis showed that the introduction of LLDPE-g-MAH decreases the softening point and Tshsp to some extent, and the higher LLDPE-g-MAH content, the lower softening point and Tshsp, indicating that the LLDPE-g-MAH has no positive effect on the high-temperature properties of the HDPE/SBS compound modified asphalts. The addition of LLDPE-g-MAH resulted in less resistant permanent deformation at high temperatures (higher resistance to rutting) compared with the original modified asphalt.

### Table 1

<table>
<thead>
<tr>
<th>Properties</th>
<th>Value</th>
<th>Chemical composition (%)</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Penetration 25 °C, d mm</td>
<td>67.1</td>
<td>Asphaltenes</td>
<td>12.13</td>
</tr>
<tr>
<td>Softening point, °C</td>
<td>50.1</td>
<td>Resins</td>
<td>22.82</td>
</tr>
<tr>
<td>Ductility 10 °C, cm</td>
<td>21.1</td>
<td>Saturates</td>
<td>11.93</td>
</tr>
<tr>
<td>Density 15 °C, g/cm³</td>
<td>1.004</td>
<td>Aromatics</td>
<td>53.12</td>
</tr>
</tbody>
</table>
The above research proves that LLDPE-g-MAH had significant effect on low-temperature properties of HDPE/SBS modified asphalt, but had no positive influence on its high-temperature property. As we all know that the engineering properties of modified asphalt were strongly influenced by the characteristics of each constituent that formed the material. When asphalt was mixed with melting blends of HDPE, SBS and LLDPE-g-MAH, the resulting system was a composite in which the asphalt was the matrix, HDPE and SBS can be considered as the filler and modifier, the LLDPE-g-MAH played a role of dispersion agent and compatilizer. The interaction that occurs between asphalt binders and LLDPE-g-MAH is a chemical course, in addition, the light components from the base bitumen are not only absorbed into HDPE and SBS particles, but also can enter into LLDPE graft polymer particles, the absorption of the light composite make the particles of modifiers and compatilizer swell, and result in an increase in their volume, cause the structure of asphalt changes from sol to gel due to loss of the light components [15]. The increase volume of modifiers and additive particles during the mixing and reaction with the base asphalt cause the change of its classical property.

3.2. Effects of LLDPE-MAH on High-temperature storage stability of HDPE/SBS modified asphalt

Softening point variation and phase compatibility have been used to evaluate the high-temperature storage stability if phase separation occurred. Difference of softening point between top and bottom section of samples after the hot storage stability test can be no higher than 2.5 °C, indicating that there is no substantial phase separation or interactions between the polymers and the asphalts during mixing, which is strong enough to resist phase separation of the modified asphalts. In addition, optical microscopy was employed to observe the distribution behavior of modifier particles. For true stability station, the top section of the sample should have the similar dispersion characteristics with the bottom section. To determine the influence of LLDPE-MAH, storage stabilities of compound modified asphalts with LLDPE-g-MAH and in absent of LLDPE-g-MAH were compared in Table 5.

Obviously, for asphalts modified by HDPE/SBS without LLDPE-g-MAH, the difference in the softening points was obvious; demonstrating that the phase separation was serious and the stable modified asphalt could not be obtained. When the addition of LLDPE-g-MAH was increased, an obvious decrease tendency of phase separating can be observed. Table 5 showed that, for HDPE/SBS modified asphalt with 0.13% LLDPE-g-MAH (simple A), the difference of softening point was obvious as well, but it was much smaller compared with sample D, which had no LLDPE-g-MAH. When LLDPE-g-MAH was increased to 0.27% (simple B), the storage stability of HDPE/SBS compound modified asphalts was improved significantly, difference of softening point between

### Table 2
Properties of HDPE and SBS.

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Density (g/cm³)</th>
<th>Melt flow rate (g/10 min)</th>
<th>Elongation at break (%)</th>
<th>Tensile strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>0.95</td>
<td>0.9</td>
<td>800</td>
<td>23</td>
</tr>
<tr>
<td>SBS</td>
<td>0.94</td>
<td>0.5</td>
<td>750</td>
<td>18</td>
</tr>
</tbody>
</table>

### Table 3
Low-temperature properties of HDPE/SBS compound modified asphalt.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Mass ratio (HDPE/SBS/LLDPE-g-MAH)</th>
<th>Low-temperature performance of modified asphalt</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Penetration (25 °C, 0.1 mm)</td>
<td>Ductility (10 °C, cm)</td>
</tr>
<tr>
<td>A</td>
<td>4/1.3/0.13</td>
<td>51.1</td>
</tr>
<tr>
<td>B</td>
<td>4/1.3/0.27</td>
<td>58</td>
</tr>
<tr>
<td>C</td>
<td>4/1.3/0.4</td>
<td>67</td>
</tr>
<tr>
<td>D</td>
<td>4/1.3/0</td>
<td>50.8</td>
</tr>
</tbody>
</table>

a The percentage contents of HDPE, SBS, LLDPE-g-MAH were calculated based upon the weight of base asphalt.

### Table 4
Effect of LLDPE-g-MAH content on the high-temperature properties of HDPE/SBS composite modified asphalt.

<table>
<thead>
<tr>
<th>LLDPE-g-MAH content (%)</th>
<th>Property</th>
<th>Softening point, °C</th>
<th>T&lt;sub&gt;SHRP&lt;/sub&gt;, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
<td>64.3</td>
<td>80.2</td>
</tr>
<tr>
<td>0.13</td>
<td>61.1</td>
<td>59.6</td>
<td>73.8</td>
</tr>
<tr>
<td>0.27</td>
<td>59.6</td>
<td>58.9</td>
<td>72.1</td>
</tr>
<tr>
<td>0.4</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table 5
Storage stability of HDPE/SBS modified asphalts as measured by softening point.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Softening point (T&lt;sub&gt;SN&lt;/sub&gt;(°C))</th>
<th>Original sample</th>
<th>Top section&lt;sup&gt;b&lt;/sup&gt;</th>
<th>Bottom section&lt;sup&gt;b&lt;/sup&gt;</th>
<th>ΔT&lt;sub&gt;SN&lt;/sub&gt;&lt;sup&gt;c&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>61.1</td>
<td>73.9</td>
<td>58.9</td>
<td>15</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>56.9</td>
<td>60.65</td>
<td>57.9</td>
<td>2.75</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>59.65</td>
<td>58.9</td>
<td>58.75</td>
<td>0.15</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>64.3</td>
<td>78.2</td>
<td>52.7</td>
<td>25.5</td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup> Mass ratio of HDPE/SBS/LLDPE-g-MAH was the same as samples A, B, C, & D listed in Table 3.
<sup>b</sup> Stored in an oven at 163 °C for 48 h.
<sup>c</sup> ΔT<sub>SN</sub> = the difference of softening point between top section sample and bottom section sample.

![Fig. 1. Curve of G’/sin(δ) versus temperature for HDPE-SBS modified asphalt with different content of LLDPE-g-MAH.](image-url)
top and bottom section of samples was 2.75 °C, extremely close to 2.5 °C, but it failed to meet the standard of stable asphalt. At the LLDPE-g-MAH content of 0.4% (simple C), difference of softening point between top and bottom section of samples was significantly narrowed to 0.15 °C, indicating that stable modified asphalt was obtained. So it can be concluded that LLDPE-g-MAH has a positive effect and its proper content is important to improve the high-temperature storage stability.

Furthermore, optical microscopic observation as the direct approach was also used to study distribution behavior of compound particles and the phase interface between modifiers and base asphalt at high temperature, and directly determine the storage stability of HDPE/SBS modified asphalts melt blended with LLDPE-g-MAH. Micrographs of composite material modified asphalts were shown in Fig. 2. In order to investigate the influence of LLDPE-g-MAH, HDPE/SBS content was set up to be constant value. At low LLDPE-g-MAH content or without LLDPE-g-MAH, as shown in Fig. 2A and D, the phase separation was serious, composite coalescent particles dispersed in asphalt, indicating that homogeneous system could not be developed at lower LLDPE-g-MAH. From Fig. 2, the dispersion characteristic of B and C was improved significantly compared with A and D, when LLDPE-g-MAH content was increased. We can also learn that almost no obvious phase interface was observed, particle size became much smaller, and uniformly distributed in asphalt, indicating that homogeneous system was developed under these contents. Furthermore, micrographs of the top and the bottom samples of compound modified asphalt containing LLDPE-g-MAH after 48 h storage were also shown in Fig. 3. With increasing LLDPE-g-MAH content, dispersion behavior of the top and bottom section samples was tending to unanimity. The dispersion character of compound modified asphalts morphology is in accordance with the conclusion drawn from the storage stability test.

A good compatibility between the polymer and the asphalt is desired to prevent separation during storage, pumping, and applying the asphalt and to achieve the expected properties in the pavement [13]. In this paper, LLDPE-g-MAH was selected as the compatible agent for the following reasons: (1) LLDPE-g-MAH has a PE-attracting group (LLDPE) and a hydrophobic (oil-attracting MAH) group. The former group has good affinity to HDPE and the MAH group can interact with the alkaline group of asphalt. It was added into HDPE/SBS modified asphalt system, as a surfactant, its structure cannot only decrease the interfacial tension between the asphalt and modifiers, but also has the function to connect the asphalt and the modifier. (2) LLDPE-g-MAH, but not HDPE-g-MAH, was selected to improve the compatibility between asphalt and modifies for the structure difference between HDPE and LLDPE, for the crystallinity of LLDPE is lower than HDPE. Due to the differences in producing method, there are lots of short branched-chains in the LLDPE structure, but as for HDPE, there is only a long chain, almost no branched-chain. The structure difference leads to the different compatibility with the asphalt. Structure with more branched-chains has the better affinity towards asphalt constituents and SBS particles. Thus LLDPE-g-MAH was used not only for its surfactant structure, but also for its better affinity towards components of asphalt and SBS than HDPE-g-MAH, which can bring about the physical and chemical interaction among HDPE, SBS and asphalt, also improve the interaction between HDPE and asphalt, SBS and asphalt. All these interaction can develop a steric barrier which can resist phase separation.

In brief, the LLDPE-g-MAH can improve compatibility through decreasing interface tension and enhancing the interaction between modifiers and asphalt, which are effective to develop stable modified asphalt system.

3.3. Effects of LLDPE-MAH on rheological properties of HDPE/SBS modified asphalt

Asphalt, as viscoelastic material, displays different characteristics of application properties in paving industry. Thus, it is crucial to examine the rheological properties of asphalt. In order to evaluate the influence of LLDPE-g-MAH on rheological properties of asphalt.
asphalt, all HDPE/SBS modified asphalt with different LLDPE-g-MAH content were prepared as the same procedure and the rheological tests were essentially non-destructive, differences in their rheological behavior are attributed exclusively to their composition and molecular structure of the polymer modifier [16]. The rheological behavior of the modified asphalt was summarized in Figs. 4–7.

Parameters in the following figures would be introduced respectively. Two important parameters complex modulus ($G'$) and phase angle ($\delta$) can be measured directly from rheological tests.
modified asphalts with different content of LLDPE-g-MAH graft polymer.

Isochronal plots of phase angle $\delta$ for purely elastic behavior and 90°, the in-phase component of $G'$, the out-phase component of $G'$, and the complex modulus $G'$.

Under a range of temperatures at a definite frequency. The former parameter $G'$ related to the material strength is the ratio of the peak stress to the peak strain, reflecting the total stiffness, it represents the shear stiffness of the bitumen under the conditions of testing. And the latter phase angle ($\delta$) defined as the phase difference between the peak stress and strain, provides the information about the ratio between elastic and viscous response during the shearing process and represents the time delay between the applied stress and the measured strain with limiting values of 0° for purely elastic behavior and 90° for purely viscous behavior [17,19]. Another two parameters $G'$ and $G''$ can be calculated from the $G'$ and phase angle ($\delta$): $G''$, the in-phase component of $G''$, is the shear storage modulus can be used to examine elastic character of modified asphalt; $G'$, the out-phase component of $G''$, is the loss modulus, and can determine the viscous behavior of modified asphalt.

Effect of LLDPE-g-MAH on the storage modulus $G'$ was studied and the results were shown in Fig. 4. $G'$ represents the completely recoverable part of the mechanical energy used during deformation; it is usually used to determine the elastic behavior of the modified asphalt. As shown in Fig. 4, for all modified asphalt samples, with increasing of temperature, value of $G'$ reduced smoothly, indicating the decreased elastic behavior with increasing temperature. Under identical conditions (graft polymer concentration), samples prepared with lower LLDPE-g-MAH content were more elastic than those prepared with higher LLDPE-g-MAH. At a fixed temperature, the higher LLDPE-g-MAH content, the worse of the elastic character, showing that introduction of LLDPE-g-MAH had no positive effect on elastic character of HDPE/SBS modified asphalt, and more LLDPE-g-MAH express the worse elastic behavior.

From the above tendency, we can know that the degree of elasticity is dependent on the LLDPE-g-MAH amount. HDPE/SBS modified asphalt with lower LLDPE-g-MAH content express more elastic than those with more LLDPE-g-MAH, it could be explained that the introduction of LLDPE graft polymers can enhance the interaction between modifiers and asphalt. In addition, LLDPE-g-MAH also have the function of polymer modifiers, that also can be swollen by light component, and change the four components of asphalt. Thus, the viscoelastic behavior of HDPE/SBS modified asphalt was changed.

The relationship between phase angle $\delta$ and the complex modulus $G'$ was described in Fig. 5. It’s another useful method to represent rheological data without including frequency or temperature [13]. For all samples, with increasing complex modulus $G'$, phase angle $\delta$ show a similar decreasing tendency. Slope changes in the $\delta$ ($G''$) profiles of HDPE/SBS modified asphalts, indicative of an elastic–viscoelastic behavior that entails structure changes, are attributed to the introduction of the graft polymer as this behavior can cause the change of the whole modified asphalt system. From Fig. 5, we can also know that the addition of LLDPE-g-MAH brought about the decrease of phase angle, and the reduction was the function of LLDPE-g-MAH concentration. Addition of relatively small amounts of LLDPE-g-MAH dramatically changes the elastic–viscoelastic character of HDPE/SBS modified asphalts, the more LLDPE-g-MAH, the closer to the curve of original modified asphalt without LLDPE-g-MAH. In addition, for the samples with any graft polymer content, black diagrams of them are all above the original modified asphalt, indicating that the rheological behavior of a HDPE/SBS modified asphalt system including LLDPE-g-MAH was not as well as the system without graft polymer.

The correlations between complex modulus and temperature, phase angle $\delta$ and temperature were analyzed in Figs. 6 and 7. Polymers have a significant effect on enhancing elasticity of asphalt. Increasing complex modulus (elastic modulus) reflects a promising rutting resistance at high temperature [5]. And phase angle isochrones have been used to characterize the elastic response of polymer modified asphalt. As $\delta$ is generally considered to be structure sensitive [16].

As illustrated in Figs. 6 and 7, the isochronal $\delta$ and $G'$ curves were function of temperature and LLDPE-g-MAH content for asphalt blends. Over the temperature range examined, $G'$ decreased smoothly with increasing temperature. But phase angles increased with increasing temperature. Figs. 6 and 7 also showed that $G'$ and $\delta$ were smaller than those with LLDPE-g-MAH, and blends with more LLDPE-g-MAH corresponded to the lower phase angle $\delta$ and higher $G'$, indicating a less elastic behavior and a higher susceptibility to rutting, which was in accordance with the viewpoint got from the high-temperature performance. Then we can draw the conclusion that the addition of LLDPE-g-MAH had no positive contribution to the viscoelastic character of the HDPE/SBS modified asphalt. It should be noted that this study was carried out at the definite HDPE/SBS content and a fixed frequency. We only concentrate on the rheological parameters changed with temperature, there are still some limitations. Notwithstanding its limitation, this study does suggest the negative effect of LLDPE-g-MAH, and these problems might be improved and even solved if we change base asphalt or the ratio of HDPE and SBS. Despite these shortcomings,
this study can clearly evaluate the influence of LLDPE-g-MAH on rheological property of HDPE/SBS modified asphalt.

4. Conclusions

LLDPE-g-MAH was used as an additive of the HDPE/SBS modified asphalt to enhance some of its performance. Penetration and low temperature ductility tests were used to evaluate low-temperature property of HDPE/SBS modified asphalt, the result showed that the penetration at 25 °C and ductility at 10 °C increased significantly with the addition of LLDPE-g-MAH. LLDPE-g-MAH has a greatly positive influence on the low-temperature performance of HDPE/SBS modified asphalt. Softening point and the maximum failure temperature were used to analysis the high-temperature property of HDPE/SBS modified asphalt, and we can know that the softening point and rutting resistance parameter $T_{SRHP}$ (corresponding temperature at $G'/\sin(\delta) = 1 \text{ KPa}$) of HDPE/SBS modified asphalt were reduced with the addition of LLDPE-g-MAH. LLDPE-g-MAH had almost no obvious and even negative effect on the high-temperature performance of HDPE/SBS modified asphalt. In addition, Softening point variation and optical micrographs were employed to investigate the storage stability of HDPE/SBS modified asphalt, and the results confirmed that only a little LLDPE-g-MAH can dramatically decrease the difference of softening point between top section sample and bottom section sample. LLDPE-g-MAH enhanced significantly the high temperature storage stability of HDPE/SBS modified asphalt by improving the compatibility between the asphalt and modifiers. It is of great benefit for LLDPE-g-MAH to disperse HDPE and SBS particles in base asphalt, and develop homogeneous and stable system. After addition of LLDPE-g-MAH, the size of modifiers particles became smaller, and degree of phase separation was reduced. At a certain range of LLDPE-g-MAH content, distribution behavior of modifiers particle from the top and bottom section was extremely the same after stored for 48 h. At last, Some work on rheological properties of HDPE/SBS modified asphalt was conducted. We concluded that LLDPE-g-MAH had almost no significant contribution to the visco-elastic character of the HDPE/SBS modified asphalt. More LLDPE-g-MAH correspond to higher phase angle $\delta$ but lower complex modulus and worse elastic behavior, indicating the asphalts exhibit a more susceptibility to rutting.

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References