Research Article

Effects of UV Aging on Physical Properties and Physicochemical Properties of ASA Polymer-Modified Asphalt

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The purpose of this study was to characterize and evaluate the effects of UV radiation on the rheological and physicochemical properties of ASA polymer-modified asphalt. The conventional properties (penetration, softening point, ductility, and Brookfield viscosity) of ASA polymer-modified asphalt were tested. Based on rheology, the effect of different UV irradiation times on the high-temperature performance of ASA polymer-modified asphalt was systematically characterized, the thermogravimetric analyzer (TGA) was used to analyze thermogravimetric properties of ASA polymer-modified asphalt, and the micromorphological characteristics of ASA-modified asphalt under different UV irradiation times were characterized by scanning electron microscope (FIB-SEM). The results show that the ASA polymer has a significant effect on the basic properties of asphalt. Compared with the base asphalt, the high-temperature stability of the modified asphalt was decreased, and the low-temperature ductility performance was improved. ASA polymer can effectively reduce the aging effect of ultraviolet radiation on the base asphalt and inhibit the generation of microcracks. ASA polymers improve the thermal stability of asphalt binders. During the aging process, the formation of carbonyl and sulfoxide groups was inhibited, indicating that ASA polymer can effectively delay the UV aging of asphalt.

1. Introduction

Asphalt roads were widely used because of their excellent road performance. However, with the increase of traffic volume and axle load year by year, the performance of ordinary asphalt roads was difficult to meet the requirements. Therefore, the corresponding asphalt modification technology has been proposed by global road researchers. Among them, resin-modified asphalt has always been the focus of attention due to its good high and low-temperature performance, fatigue performance, and wide range of sources [1–4].

During the long-term service period, the coupling effect of various factors such as ultraviolet radiation, temperature, and water on the asphalt material will cause irreversible aging, and the road performance will be significantly deteriorated, which will lead to premature cracks, potholes, and spalling of the asphalt pavement and other adverse diseases [4–6]. Among them, ultraviolet radiation can induce the transformation of chemical components in asphalt, destroy the stability of asphalt colloid structure, and accelerate the performance degradation of asphalt pavement, which was one of the important factors leading to long-term aging of asphalt. In view of this, effectively controlling the ultraviolet aging behavior of asphalt pavement materials was the key factor to realize the long-life development of asphalt pavement [6–10]. However, researchers still have not fully grasped the UV aging mechanism of asphalt. Therefore, it was of great significance for the application and promotion of resin-modified asphalt to study the UV aging process of asphalt binder, especially the role and law of resin modifiers in this process [11–15]. Among them, acrylate-styrene-acrylonitrile (ASA) resin is a copolymer of styrene,
acrylonitrile, and acrylate rubber. Technically, ASA not only maintains the main characteristics of ABS resin but also combines the advantages of weather resistance of PMMA resin and has good anti-ultraviolet radiation performance, so that the application of the product can be extended to outdoor use.

At present, there are few research results on ASA-modified asphalt, especially on its anti-ultraviolet radiation performance. In this paper, conventional tests and Brookfield viscosity tests were carried out on ASA resin-modified asphalt. On this basis, the effect of ultraviolet radiation on the high-temperature rheological properties of ASA-modified asphalt was further studied, and the micromorphological evolution process of ASA-modified asphalt was systematically characterized by means of FIB-SEM. The thermophysical properties and functional group composition of ASA-modified asphalt were studied by thermogravimetric analyzer-Fourier infrared system (TG-FTIR). This research will provide relevant basis for the application of ASA modifier in asphalt pavement materials.

2. Materials and Methods

2.1. Materials

2.1.1. Asphalt. 70# base asphalt was selected as the carrier, and the specific technical indicators are shown in Table 1.

2.1.2. Acrylate-Styrene-Acrylonitrile (ASA). The melting point and density of ASA are 160–200°C and 1.06–1.09 g/cm³, respectively. Acrylate-styrene-acrylonitrile was selected as modifier, and the dosage was 2%, 4%, and 6% of the asphalt mass. The selected ASA molecular structure is shown in Figure 1.

2.2. Preparation Process of ASA-Modified Asphalt. 70# base asphalt was put into the constant temperature oven of 110°C and kept for 6 h. In the use of high-speed shear apparatus, first of all, the molten base asphalt is maintained at 130°C, and then the corresponding mass of ASA modifier is evenly added into the molten matrix asphalt three times. After each addition, the manual stirring method is used to stir for 1 min to ensure the full mixing of the modifier and the matrix asphalt. The initially mixed ASA-modified asphalt was placed in an oil bath heating device at 160°C ± 5°C for 0.5 h and stirred at a rate of 600 rad/min for 8 min. The specific preparation process is shown in Figure 2.

2.3. Methodology

2.3.1. UV Aging Method. Three 300 W UV lamps were used to simulate outdoor UV radiation. The radiation area was 0.8 m², and the ultraviolet radiation intensity was 35 W/m². During the experiment, the vertical distance between the asphalt sample and the UV lamp was kept at 30 cm. The UV aging testing facility is shown in Figure 3. The conversion formula and calculation results of indoor simulation time and outdoor actual ultraviolet radiation time are shown in formula (1) and Table 2, respectively.

2.3.2. Brookfield Viscosity Test. The Brookfield viscosity test was carried out according to ASTM D4402/D4402M-13, and the experimental temperatures were selected as 90°C, 135°C, and 175°C. It was used to evaluate the apparent viscosity, fluidity, and other properties of ASA-modified asphalt.

2.3.3. Multiple Stress Creep and Recovery Test (MSCR). The MSCR test was carried out according to ASTM-D7405-A10a. The MSCR test was carried out at 64°C, the parallel plate size was chosen to be 25 mm, and the gap was 1 mm. Twenty cycles were run at a stress level of 0.1 kPa, with the first 10 cycles used to condition the samples, followed by 10 cycles at a stress level of 3.2 kPa, for a total of 30 cycles of creep recovery. Each cycle was a creep period of 1 s and an unloading recovery period of 9 s, and the total test time was 300 s. The evaluation indicators were recovery rate (R) and non-recoverable creep compliance (Jnr), calculated as follows:

\[ R = \frac{\gamma_p - \gamma_{nr}}{\gamma_p - \gamma_0} \]

\[ J_{nr} = \frac{\gamma_{nr} - \gamma_0}{\tau} \]

where \( \gamma_p \) is the peak strain in each loading cycle, \( \gamma_{nr} \) is the residual strain in each loading cycle, \( \gamma_0 \) is the initial strain in each loading cycle, and \( \tau \) is the corresponding shear stress. In the calculation, the first 10 cycles under the stress of 0.1 kPa were not involved in the calculation as the sample pre-treatment, and only the data of the last 20 cycles were selected.

2.3.4. SEM Analysis. The morphology evolution of ASA-modified asphalt under ultraviolet radiation was studied by focusing ion beam microscopy (FIB-SEM). Firstly, the asphalt sample is placed on the aluminum plate. Since the surface of the sample does not have conductivity, it is necessary to coat a layer of metal conductivity on the sample. Then, it is placed in a microscope under vacuum, and the sample surface is observed by amplifying 400 and 2000 times, respectively. The indoor UV aging simulation time was 76.25 h, 152.5 h, 228 h, and 305 h.

2.3.5. TG Analysis. The thermal properties of ASA-modified asphalt were characterized by TG 209 F1 thermogravimetric analyzer-Fourier infrared system (TG-FTIR) produced by NETZSCH formula in Germany. According to the reference
**Figure 1:** Acrylate-styrene-acrylonitrile molecular structure and sample appearance.

**Figure 2:** Preparation process of modified asphalt.

**Figure 3:** UV aging testing facility.

**Table 2:** Setting UV aging time.

<table>
<thead>
<tr>
<th>Actual illumination time</th>
<th>3 months</th>
<th>6 months</th>
<th>9 months</th>
<th>12 months</th>
</tr>
</thead>
<tbody>
<tr>
<td>Indoor simulation time</td>
<td>76.25 h</td>
<td>152.5 h</td>
<td>228 h</td>
<td>305 h</td>
</tr>
</tbody>
</table>
of the literature and the preparation of the preliminary test, the quality of the sample is controlled at about 9 mg. The alumina crucible is used in the test. The test temperature is 20°C–500°C, the heating rate is 10°C/min, the flow rate is 50 mL min, and the nitrogen atmosphere is used.

2.3.6 Dynamic FTIR Analysis. The main functional groups of base asphalt and ASA-modified asphalt and the changes of functional groups under UV aging were analyzed by TG 209 F1 thermogravimetric analyzer-Fourier infrared system (TG-FTIR) produced by NETZSCH Germany. The test conditions were as follows: scanning range: 4000–400 cm$^{-1}$, scanning times: 8, and resolution: 4 cm$^{-1}$. The test temperature was 20°C to 600°C. Thermogravimetric analysis and infrared spectroscopy are simultaneous tests. In order to avoid the termination of the infrared spectroscopy when the thermal gravimetric test reaches the maximum temperature required, the test time of infrared spectroscopy is increased.

3. Results and Discussion

3.1. Conventional Test. The conventional test results under different UV aging time and ASA modifier dosage are shown in Figure 4. It can be seen from the diagram that UV radiation significantly reduces the ductility and penetration of 70# base asphalt and improves the softening point of asphalt. From 0 h to 305 h, the softening point of 70# base asphalt increased by 11.9°C, and the ductility and penetration decreased by 12.3 cm and 17.7 (0.1 mm), respectively. This indicates that UV radiation makes base asphalt harden and improves its deformation resistance but deteriorates its low-temperature ductility. This reflects the effect of ultraviolet radiation on the colloidal structure of asphalt. Under the action of ultraviolet radiation, the light components in asphalt gradually change to the recombinant components such as colloid and asphaltene while volatilizing, thereby increasing the content of the recombinant components, which leads to higher high-temperature resistance of asphalt.

For ASA-modified asphalt, the conventional test results under UV irradiation showed a similar trend to that of 70# base asphalt, but the influence degree was significantly different. From 0 h to 305 h, the softening point of 2%, 4%, and 6% ASA-modified asphalt increased by 4.8°C, 8.1°C, and 8.1°C, respectively. The penetration decreased by 11.8 cm, 10.1 cm, and 6.9 cm, respectively. The ductility decreased by 11.5 cm, 11.3 cm, and 10.7 cm, respectively. With the extension of UV radiation time, the variation range of conventional test results of ASA-modified asphalt was significantly smaller than that of 70# base asphalt, indicating that ASA modifier can effectively inhibit the influence of UV radiation on the basic performance and improve the UV radiation resistance of asphalt.

In addition, it can be found from Figure 4 that in the process of UV radiation, with the increase of dosage, the softening point difference of ASA-modified asphalt was stable within a certain range, showing only a small fluctuation. The difference between ductility and penetration decreased gradually with the increase of dosage, indicating that increasing the dosage of ASA modifier within a reasonable range was conducive to further improving the anti-UV aging performance of asphalt. Therefore, ASA-modified asphalt with 6% dosage was selected for further performance characterization.

3.2. MSCR Test. The creep curves of 70# base asphalt under different UV aging time are shown in the above figure. It can be seen from Figure 5 that as the UV aging time gradually increases to 228 h, the shear strain of the 70# base asphalt gradually decreases, which indicates that the UV radiation induces the hardening of the base asphalt and significantly improves the deformation resistance, which was basically consistent with the conventional performance test. In addition, the high-temperature rheological properties of asphalt can be characterized by two high-temperature rheological indicators, the recovery rate $R$ and the non-recoverable creep compliance $J_{nr}$.

It can be seen from Figure 5 that under the load level of 0.1 kPa, from 0 h to 228 h, the recovery rate $R$ of 70# base asphalt increases by 4.96 times, and the non-recoverable creep compliance decreases by 23.6%. However, when the aging time continued to extend to 305 h, $R$ decreased by about 68.6%, and $J_{nr}$ increased by 27.6%, which fully proved that in the early stage of aging, the aging effect of ultraviolet radiation on 70# base asphalt was significant, but as the aging continued, the elastic recovery performance of 70# base asphalt was seriously deteriorated.

It was worth noting that compared with 0.1 kPa, the $R$ value and $J_{nr}$ value of 70# base asphalt decreased and increased significantly under the load of 3.2 kPa, respectively, which was because the asphalt material was more prone to irreversible deformation under high stress load.

It can be seen from Figure 6 that the shear strain of ASA-modified asphalt gradually decreases with the prolongation of UV aging time, which was similar to the change trend of 70# base asphalt. However, the difference was that the shear strain of ASA-modified asphalt decreases less than 70# base asphalt with the prolonging of aging time. This shows that ASA modifier can improve the anti-ultraviolet aging performance of asphalt. In addition, it can be found that the ASA-modified asphalt under different aging time conditions was smaller than the 70# base asphalt, which indicates that the ASA modifier optimizes the viscoelastic mechanical properties of the asphalt material.

It can be found from Figure 6 that under the load level of 0.1 kPa, the $R$ value of ASA-modified asphalt was greater than that of base asphalt in the early stage of aging, and the change trend was similar to that of 70# base asphalt, but its growth rate was lower than that of 70# base asphalt. At 228 h, the $R$ value of ASA-modified asphalt increased by 29.8%. Under the load of 3.2 kPa, the $R$ value of ASA-modified asphalt was greater than that of 70# base asphalt. It was worth noting that as the aging time was extended to 228 h, the $J_{nr}$ value gradually increases, and the variation
range was greater than that of the 70# base asphalt, which was opposite to the trend of the $R$ value and needs further research.

3.3. SEM Analysis. It can be seen from Figure 7 that a large number of microcracks in the same direction have appeared on the surface of 70# base asphalt at 76.25 h of aging. With the extension of time, 70# base asphalt surface microcracks develop in many directions, and the number, length, and depth also gradually increase and connect with each other. When the aging time was 152.5 h, the microcracks are broken and the cracks are more obvious.

After aging for 228 h, the number of microcracks increased further, some microcracks cracked, and the crack width expanded. After 305 h, the crack length further developed, and edge warping occurred, indicating that the aging degree of 70# base asphalt deepened.

From Figure 8, it can be found that ASA-modified asphalt also has a large number of microcracks when aging for 76.25 h. Compared with the base asphalt with the same aging time, ASA-modified asphalt has more microcracks and larger depth. However, with the continuous aging, the number of microcracks on the surface of ASA-modified asphalt decreased significantly, and the length of microcracks also decreased significantly.

When aging for 305 h, ASA-modified asphalt surface showed a certain number of folds, with no obvious microcracks. This shows that the addition of ASA modifier accelerates the appearance of microcracks on asphalt surface under short-term UV radiation. However, with the extension of aging time, ASA modifier was beneficial to inhibit the development of microcracks on the surface of asphalt.

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![Figure 4: Conventional test results under different UV aging times. (a) Softening point. (b) Penetration. (c) Ductility.](image-url)
materials and gradually eliminate the microcracks that have occurred, which greatly improves the UV radiation resistance of asphalt materials. This may be due to the gradual heating up of ASA-modified asphalt surface under the action of ultraviolet radiation. At the same time, under the influence of its excellent ductility, ASA-modified asphalt has a trend of slow deformation and gradually eliminates microcracks.

3.4. TG Analysis. Figure 9 shows the thermogravimetric curve diagram of 70# base asphalt under different aging times. It can be seen from Figure 9 and Table 3 that with the prolongation of aging time, the initial decomposition temperature of 70# base asphalt gradually decreased, from 232.6°C in 76.25 h to 158.8°C in 305 h, and the difference between the decomposition temperature at the beginning and the end of the aging time reached 73.8°C, resulting in the late asphalt in the later stage. The thermal decomposition starts at a lower temperature, indicating that the light stability of the base asphalt was poor without modification, and the ultraviolet radiation aggravates the thermal weight loss behavior of the asphalt material. For the weight loss rate, the base asphalt under different aging time conditions always remained stable at about 79%.

Figure 10 shows the DTG curves of 70# base asphalt under different aging times. It can be seen from the figure that the aging time has a significant effect on the weight loss rate of the 70# base asphalt. The weight loss rate of 70# base asphalt was the largest when it was aged for 76.25 h, reaching 12%. With the aging, the weight loss rate gradually decreased and reached about 8% at 305 h.

Figure 5: MSCR testing results of 70# base asphalt subjected to UV radiation of different times. (a) 0.1 kPa. (b) 3.2 kPa. (c) R value. (d) Jnr value.
Figure 6: MSCR testing results of ASA-modified asphalt subjected to UV radiation of different times. (a) 0.1 kPa. (b) 3.2 kPa. (c) $R$ value. (d) $J_{nr}$ value.

Figure 7: 70# base asphalt under different UV aging times of microscopic morphology. (a) 76.25 h-200 times. (b) 152.5 h-200 times. (c) 228 h-200 times. (d) 305 h-200 times. (e) 76.25 h-2000 times. (f) 152.5 h-2000 times. (g) 228 h-2000 times. (h) 305 h-2000 times.
Figures 11 and 12 show the TG curve and DTG curve of the ASA-modified asphalt, respectively. It can be seen from Figure 11 and Table 4 that the initial decomposition temperature of ASA-modified asphalt aged 76.25 h was 170.3°C, and the initial decomposition temperature gradually increases with the prolongation of aging time. ASA-modified asphalt aged for 228 h was 204°C, which was 33.7°C higher than the initial decomposition temperature of 76.25 h. This indicates that UV radiation improves the thermal stability of ASA-modified asphalt.

Compared with 70# base asphalt, the variation range of the initial decomposition temperature of ASA-modified asphalt decreases significantly with the prolongation of aging time. At the same time, it can be found from Figure 12 that the weight loss rate of ASA-modified asphalt under different aging times was basically the same. This proves that ASA modifier can improve the anti-ultraviolet radiation performance of asphalt material from a microscopic point of view.

**3.5. Dynamic-FTIR.** From Figure 13, the infrared spectrum of the base asphalt was mainly distributed in 2800–3000 cm\(^{-1}\), and the infrared absorption peaks in this range mainly represent the stretching vibration peaks of...
methylene and methyl groups. At the same time, there are moderately strong carbonyl functional groups in the range of about 1700 cm$^{-1}$. These functional groups are generated by the reaction of the internal components of the asphalt caused by the rise of the test temperature. For example, the carbonyl group was generated by the oxygen absorption reaction, and the sulfoxide group was generated by the reaction between the asphalt sulfide and the outside world. With the increase of UV aging time, the infrared spectral range of base asphalt tends to expand. This shows that as the UV radiation promotes the formation of new substances inside the base asphalt, it proves the deepening of the aging degree.

Table 4 shows the functional group index of 70# base asphalt under different aging times. The functional group index of 70# base asphalt basically shows an increasing trend. Compared with 76.25 h of aging, the carbonyl, sulfoxide, and aromatic functional group indices of 305 h of aging were increased by 323.8%, 807.4%, and 593.8%, respectively. This fully shows that long-term ultraviolet radiation will induce serious aging effect of 70# base asphalt.

<table>
<thead>
<tr>
<th>Aging time</th>
<th>Initial decomposition temperature/°C</th>
<th>Weight loss/°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>76.25 h</td>
<td>170.3°C</td>
<td>80.56%</td>
</tr>
<tr>
<td>152.5 h</td>
<td>190.8°C</td>
<td>83.15%</td>
</tr>
<tr>
<td>228 h</td>
<td>204°C</td>
<td>81.28%</td>
</tr>
<tr>
<td>305 h</td>
<td>194.5°C</td>
<td>77.75%</td>
</tr>
</tbody>
</table>

Figure 11: TG test results of ASA-modified asphalt under different UV aging times.

Figure 12: DTG curves of ASA-modified asphalt under different UV aging times.
Figure 14 shows the infrared spectrum of ASA-modified asphalt under different aging times. It can be seen that there was no significant difference in the spectra of ASA-modified asphalt under the four aging times, indicating that UV radiation has little effect on the chemical composition of ASA-modified asphalt. It can be found from Table 6 that the functional group index of ASA-modified asphalt was basically consistent with the development trend of 70# base asphalt. However, the difference was that the functional group index of ASA-modified asphalt increases less than the base asphalt with the aging time. Compared with the aging 76.25 h, the carbonyl, sulfoxide, and aromatic functional group indices of the ASA-modified asphalt aged 228 h increased by 147.3%, 337.4%, and 313.3%, respectively. This shows that the addition of ASA modifier significantly improves the aging stability of asphalt materials. It was worth noting that the functional group index decreased significantly after aging for 305 h, which may be due to the surface hardening of the asphalt after long-term aging, which delayed the progress of UV aging.

**Table 5**: Functional group index of 70# base asphalt at different UV aging stages.

<table>
<thead>
<tr>
<th>Aging time</th>
<th>$I_{C=O}$</th>
<th>$I_{Ar}$</th>
<th>$I_{S=O}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>76.25 h</td>
<td>0.05</td>
<td>0.025</td>
<td>0.05</td>
</tr>
<tr>
<td>152.5 h</td>
<td>0.051</td>
<td>0.051</td>
<td>0.051</td>
</tr>
<tr>
<td>228 h</td>
<td>0.062</td>
<td>0.029</td>
<td>0.057</td>
</tr>
<tr>
<td>305 h</td>
<td>0.047</td>
<td>0.034</td>
<td>0.049</td>
</tr>
</tbody>
</table>

**Table 6**: Functional group index of ASA-modified asphalt at different UV aging stages.

<table>
<thead>
<tr>
<th>Aging time</th>
<th>$I_{C=O}$</th>
<th>$I_{Ar}$</th>
<th>$I_{S=O}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>76.25 h</td>
<td>0.0567</td>
<td>0.0278</td>
<td>0.0354</td>
</tr>
<tr>
<td>152.5 h</td>
<td>0.0736</td>
<td>0.0475</td>
<td>0.063</td>
</tr>
<tr>
<td>228 h</td>
<td>0.1402</td>
<td>0.1216</td>
<td>0.1465</td>
</tr>
<tr>
<td>305 h</td>
<td>0.0645</td>
<td>0.0788</td>
<td>0.044</td>
</tr>
</tbody>
</table>
4. Conclusions

Based on the above findings, the following conclusions can be drawn:

(i) ASA modifier can reduce the high-temperature performance and deformation resistance of asphalt materials and improve the low-temperature ductility of asphalt materials, and the modification effect on the above three properties increases with the increase of the dosage. Compared with 2% ASA-modified asphalt, the softening point and penetration of 6% ASA-modified asphalt decreased 5.5°C and 3.7 (0.1 mm) respectively, and its ductility increased 1.1 cm.

(ii) ASA modifier improves the viscoelasticity and anti-UV aging properties of asphalt materials during UV radiation. According to MSCR test, from 0 h to 228 h, the recovery rate \( R \) of 70# base asphalt increases by 4.96 times, and the irrecoverable creep compliance decreases by 23.6%. However, the \( R \) value of ASA-modified asphalt increased by 29.8%.

(iii) In the process of UV aging, microcracks appeared on the surface of 70# base asphalt and gradually developed with the extension of aging time. ASA modifier can effectively inhibit the formation and development of microcracks caused by UV radiation.

(iv) The initial decomposition temperature of 70# base asphalt changes greatly with the extension of aging time. The addition of ASA modifier can effectively improve the UV aging stability of asphalt materials. During the aging process, the initial decomposition temperature of ASA-modified asphalt changed only to 33.7°C, while that of base asphalt changed to 158.8°C.

(v) The extension of UV aging time has little effect on functional group index of ASA-modified asphalt, which explains the excellent UV aging resistance of ASA-modified asphalt from microscopic point of view.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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