Research Article

Effect Investigation of Ultraviolet Ageing on the Rheological Properties, Micro-Structure, and Chemical Composition of Asphalt Binder Modified by Modifying Polymer

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Received 23 March 2022; Accepted 6 May 2022; Published 23 May 2022

Academic Editor: Aniello Riccio

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In order to investigate the effect of nano polymer materials on the UV aged performance of asphalt binder, a nano PA modifying agent was selected to prepare modified asphalt. Under the effect of UV radiation, the modifying effect of nano PA on the rheological properties of asphalt was studied. With the extension of UV ageing time, the microstructure variation of PA modified asphalt was characterized by using a focused ion beam scanning electron microscope (FIB-SEM). The thermophysical properties of PA modified asphalt were investigated by thermogravimetric analysis (TG). Furthermore, the functional group compositions of PA modified asphalt were investigated by dynamic FTIR. The results showed that with the extension of UV ageing time, the PA modifier could mitigate the ageing effect of UV radiation on the asphalt binder. PA modifier could achieve an increasing effect on the viscoelasticity of asphalt during the UV ageing process. PA modifier suppressed the generation of microcracks in the surface microzone of UV aged asphalt samples. PA modifier could promote the thermal stability improvement of asphalt binder when subjected to UV radiation. The generation of carbonyl and sulfoxide groups in asphalt was inhibited during the UV ageing period, which indicated that the UV induced reaction of asphalt could be postponed by the PA modifying agent.

1. Introduction

As one type of typical binder, asphalt is widely used in road construction worldwide. Due to its comfort and smoothness, asphalt pavement could achieve major advantages over cement concrete roads [1–3]. Because of the complex chemical composition, the ageing behavior of asphalt is prone to generate and then becomes hard and brittle under the impact of heat, light, and oxygen [4–7], which leads to the spalling and fatigue of asphalt pavement. When asphalt is further subjected to vehicle load in the presence of temperature shrinkage stress, hydrodynamic stress, and other factors, the damaging asphalt materials might result in cracks, potholes, and other failures that cause the sharp deterioration of its performance [8–10]. Currently, the effective control solution of the ultraviolet ageing behavior of asphalt materials mainly refers to adding antiageing modifiers to impart good ultraviolet ageing resistance to asphalt [11, 12]. The commonly used antiageing modifiers mainly include carbon black, nano-oxides, layered silicates, light stabilizers, and layered double-hydroxy composite metal hydroxides [13–15]. Among these modifiers, inorganic modifiers might cause the problems of low-temperature performance deterioration [16–18] and poor compatibility [19–21]. For light stabilizers, the unstable modifying effect on the UV resistance performance of asphalt inhibits the
application in asphalt pavement [22–24]. Recently, as the superior asphalt modifying agent, resin modifiers could not only improve the road performance of asphalt binders but also have the significant improving effect on the anti-ultraviolet ageing performance of asphalt materials [25, 26]. However, since the improving mechanism of resin modifiers on asphalt is still unclear, the application methods and actual effects of resin modifiers applied in asphalt are still unknown.

In this paper, the modifying effect of nano PA on the rheological properties of asphalt was studied following the impact of UV radiation. The microstructure variation of PA modified asphalt was characterized by FIB-SEM. The thermophysical properties of PA modified asphalt were investigated by TG analysis. Furthermore, the functional group compositions of PA modified asphalt were investigated by dynamic FTIR. This study would provide references for PA used into the UV ageing control of asphalt binder, which would promote the application of PA modifying agent in pavement engineering.

2. Materials and Methods

2.1. Materials

2.1.1. Asphalt. 70# asphalt was chosen in this study and its main technical properties are presented in Table 1.

2.1.2. Nano Polymer Modifier. The nano polyamide (PA6) was selected to prepare polymer modified asphalt and the dosage of polymer modified asphalt was 0.2% by asphalt weight. The selected polyamide and its molecular structure are shown in Figure 1. The technical property of PA6 is demonstrated in Table 2.

2.2. Preparation Process of PA Modified Asphalt. 70# asphalt was put into the oven at 110°C and kept for 6h. Then, the temperature of the melted asphalt was kept at 130°C. The amount of PA was weighed and added into molten asphalt. In the process of adding the modifier, the stirring was kept performing to promote the mixing between asphalt and PA. Finally, the prepared PA modified asphalt was stirred at 600 rad/min for 8 minutes, and then the preparation of modified asphalt was completed.

2.3. Methodology

2.3.1. UV Ageing Method. This study selected the ultraviolet iron lamp for the ultraviolet ageing of asphalt binder and the power of the UV lamp was 300W. The lamp was 30 cm away from the sample and the radiation area was approximately 0.8 square meters. The UV ageing testing facility is shown in Figure 2. The unit ultraviolet radiation intensity was 375 W/m2. The setting UV ageing time was calculated by using formula 1, whose results are shown in Table 3:

Indoor UV ageing time = \frac{Outdoor solar radiation energy}{Power of indoor UV radiation}.

2.3.2. Brookfield Viscosity Test. The Brookfield viscosity test was carried out in accordance with ASTM D4402/D4402M-13. The testing temperature of Brookfield viscosity included 90°C, 135°C, and 175°C, which provided evidence for evaluating the ability of PA modified asphalt to resist shear deformation.

2.3.3. Multiple Stress Creep and Recovery Test (MSCR). 25 mm flat plates were used in the MSCR test and the spacing was 1 mm. The test temperature was 64°C. The MSCR test was run for 20 cycles at the stress level of 0.1 kPa, of which the first 10 cycles were used to condition the sample. The other 10 cycles were performed at the stress level of 3.2 kPa. Meanwhile, MSCR contained a total of 30 cycles of creep recovery. Each cycle had a creep phase of 1s and an unloading recovery phase of 9s. The total test time was 300s.

The evaluation indicators included recovery rate (R) and irrecoverable creep compliance (Jnr), and the calculation formula is as follows:

\[ R = \frac{\gamma_p - \gamma_{\text{irr}}}{\gamma_p - \gamma_0}, \]

\[ J_{\text{irr}} = \frac{\gamma_{\text{irr}} - \gamma_0}{\tau}. \]

Here, \( \gamma_p \) is the peak strain in each loading cycle, \( \gamma_{\text{irr}} \) is the 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 pretreatment, and only the data of the last 10 cycles were selected.

2.3.4. SEM Analysis. The focused ion beam scanning electron microscopy (FIB-SEM) was used to investigate the
microstructure variation of PA modified asphalt with the extension of UV ageing time. The magnification of the FIB-SEM test was 200 and 2000 times. The samples mainly contained the asphalt film subjected to UV ageing of 76.25 h, 152.5 h, 228 h, and 305 h, respectively.

2.3.5. TG Analysis. TG 209 F1 thermogravimetry produced by NETZSCH in Germany was used to measure the thermophysical properties of PA modified asphalt. The quality of the asphalt sample was controlled around about 9 mg. The alumina crucible was used in the TG experiment. The test temperature ranged from 30 °C to 500 °C and the heating speed was 10 °C/min. The test was carried out in the nitrogen atmosphere with the gas flow rate of 50 mL/min.

2.3.6. Dynamic FTIR Analysis. The thermal stability and compositions of a functional group of PA modified asphalt were investigated by a fast speed thermogravimetric analyzer (TGA/DSC + Nicolet IS50, USA) coupled with an FTIR spectrometer (Mettler Toledo Thermo Fisher, USA). The heating period was from 20 °C to 600 °C and the heating rate was 10 °C/min. The test was performed in the nitrogen flow with a speed of 100 mL/min.

3. Results and Discussion

3.1. Rheological Properties

3.1.1. Brookfield Viscosity Test. The Brookfield viscosity tests of different UV ageing time and temperature conditions are
mainly shown in Figures 3 and 4. It could be analyzed from Figure 3 that the ultraviolet ageing effect could increase the apparent viscosity of 70# asphalt significantly. Under different temperature conditions, the Brookfield viscosity of 70# matrix asphalt was increased with the extension of UV ageing time, but the increasing range would firstly increase and then decrease with the increase of temperature. When the UV ageing time extended from 0 h to 305 h at 90 °C, the apparent viscosity of 70# asphalt increased by about 32.1%. When the temperature rose to 135 °C and 175 °C, the apparent viscosity of 70# asphalt increased by about 82.1% and 61.9%, which showed that under different temperature conditions, ultraviolet ageing had different effects on the apparent viscosity of asphalt materials. The significant increase in viscosity also reflected the effect of ultraviolet ageing on the asphaltene components of asphalt. The effect of ultraviolet ageing would increase the content of asphaltenes, thereby raising the viscosity of asphalt, which would promote the viscosity rise of asphalt and improve the resistance of the asphalt to high-temperature effects.

As for PA modified asphalt, it could be seen from Figure 4 that the variation trend of the apparent viscosity of PA modified asphalt with temperature and UV ageing time was basically the same as that of 70# matrix asphalt, but the specific values were obviously different. When the UV ageing time extended from 0 h to 305 h, the apparent
viscosity of PA modified asphalt increased by about 23.6%. As the temperature continued increasing to 135 °C and 175°C, the apparent viscosity of PA modified asphalt increased by 50.3% and 48.4%, respectively. The increasing rate of the apparent viscosity of PA modified asphalt during UV ageing was obviously lower than 70# asphalt, which showed that the addition of a PA modifier could retard the effect of UV ageing on apparent viscosity and improve the resistance ability of asphalt to UV radiation.

3.1.2. MSCR Test. The creep recovery of 70# asphalt under the influence of different ultraviolet ageing times is shown in Figure 5 and Table 4. It could be analyzed from Figure 5 that the cumulative strain of 70# asphalt decreased gradually at 0.1 kPa level when the ultraviolet ageing time increased to 228h. This demonstrated that at the initial stage of ultraviolet ageing, the elastic components of 70# asphalt increased with UV ageing time and the elastic recovery performance of asphalt was improved remarkably. The relevant results of MSCR were also reflected in the change of the elastic recovery rate $R$ of asphalt binder. When the ultraviolet ageing time extended from 0h to 228h, the elastic recovery rate $R$ increased by approximately 635%. As the UV ageing time extended, the elastic recovery performance of 70# asphalt was degraded and the elastic recovery rate $R$ decreased by about 68.6%, which indicated that the viscous component's content of 70# asphalt might be increased and the antideformation performance was degraded obviously. Under the 3.2 kPa stress level, the elastic recovery rate of 70# asphalt has stayed at the low level and its irreversible deformation was easy to generate, which supposed that under

![Figure 4: Brookfield viscosity testing results of PA modified asphalt subjected to UV radiation at different times: (a) 90°C, (b) 135°C, (c) 175°C, and (d) Comparison.](image-url)
Figure 5: MSCR testing results of 70# asphalt subjected to UV radiation at different times: (a) 0.1 KPa and (b) 3.2 KPa.

| Asphalt type | Ageing time/h | 0.1 kPa | 3.2 kPa |
|--------------|---------------|---------|---------|
|              | R (%)         | J_{nr}  | R (%)   | J_{nr}  |
| 70#          | 76.25         | 1.48    | 0.38    | 0.18    | 0.45    |
|              | 152.5         | 2.89    | 0.37    | 0.18    | 0.45    |
|              | 228           | 7.35    | 0.29    | 0.24    | 0.39    |
|              | 305           | 2.31    | 0.37    | 0.2     | 0.43    |
| PA           | 76.25         | 3.84    | 0.33    | 0.23    | 0.41    |
|              | 152.5         | 6.63    | 0.27    | 0.28    | 0.40    |
|              | 228           | 6.77    | 0.24    | 0.31    | 0.35    |
|              | 305           | 4.90    | 0.28    | 0.38    | 0.32    |

Table 4: Calculating values of $R$ and $J_{nr}$.

Figure 6: MSCR testing results of PA modified asphalt subjected to UV radiation at different times: (a) 0.1 KPa and (b) 3.2 KPa.
the high stress levels, ultraviolet ageing would aggravate the generation and accumulation of permanent deformation of 70# asphalt. From the analysis of Figure 6 and Table 4, it could be known that with the extension of UV ageing time, the cumulative strain of PA modified asphalt decreased significantly, which was different from the trend that the cumulative strain of 70# asphalt first dropped and then increased. This might indicate that the PA modifier could achieve the modifying effect on the viscoelasticity of asphalt during the ultraviolet radiation process. As for the elastic recovery performance, the \( R \) value changing rule of PA modified asphalt was similar to that of 70# asphalt, but the testing value was obviously different. Under different ultraviolet radiation times, the \( R \) of PA modified asphalt was better than 70# asphalt (208h was slightly lower than 70# asphalt) and the drop rate of \( R \) value at 305h was much lower than that of 70# asphalt (68.6%), only 27.6%. Under the 3.2 kPa stress level, the \( R \) value of PA modified asphalt was also obviously lower than that of 70# asphalt. At the same time, the \( J_{nr} \) of PA modified asphalt decreased obviously with the prolonging of ultraviolet radiation. The longer the UV ageing time was, the larger the \( J_{nr} \) decreased, which demonstrated that the PA modifier could improve the

\[ \text{Figure 7: Microstructure of 70# asphalt at different UV ageing stages. (a) 76.25 h-200 time. (b) 152.5 h-200 time. (c) 228 h-200 time. (d) 305 h-200 time. (e) 76.25 h-2000 time. (f) 152.5 h-2000 time. (g) 228 h-2000 time. (h) 305 h-2000 time.} \]
antiageing performance and high temperature stability of asphalt materials by mitigating the ageing effect of UV radiation.

3.2. SEM Analysis

3.2.1. 70# Asphalt. As shown in Figure 7, at the early stage of UV ageing, microcracks appeared gradually on the surface of 70# asphalt. As UV ageing time extended, the morphology and characteristics of the microcracks on the surface of asphalt were significantly different. After 76.25 hours of ultraviolet radiation, obvious cracks appeared on the surface of 70# asphalt. As the ultraviolet radiation time extended to 152.5 h, the cracks on the surface of asphalt expanded more regularly and the depth increased.

After 228h of ultraviolet radiation, the direction of surface cracks changed from up and down to left and right. The enlarged surface block distribution was more serious, which might suppose that the ageing degree of 70# asphalt had increased significantly. When the UV irradiation time reached 305h, the cracks began to change vertically and the

Figure 8: Microstructure of PA modified asphalt at different UV ageing stages. (a) 76.25h-200 time. (b) 152.5 h-200 time. (c) 228 h-200 time. (d) 305 h-200 time. (e) 76.25h-2000 time. (f) 152.5 h-2000 time. (g) 228 h-2000 time. (h) 305 h-2000 time.
Figure 9: TG analysis results of 70# asphalt at different UV ageing stages: (a) 76.25 h, (b) 152.5 h, (c) 228 h, and (d) 305 h.
Figure 10: TG evaluating indices of 70# asphalt at different UV ageing stages.
depth further increased. Furthermore, the roughness of the asphalt surface increased visibly.

3.2.2. PA Modified Asphalt. It could be seen from Figure 8 that with the prolongation of ultraviolet radiation time, no obvious microcracks appeared on the surface of PA modified asphalt, which indicated that the addition of nano PA could alleviate the generation of microcracks on the asphalt surface induced by ultraviolet radiation. When the ultraviolet radiation time reached 76.25 h, a certain number of wrinkles appeared on the surface of PA modified asphalt sample and the wrinkles extended vertically from the middle trunk to the surroundings, which promoted forming a grid on the surface of the PA modified asphalt. Under the condition of 152.5 h ultraviolet radiation, the wrinkles on the surface of PA modified asphalt were relatively gentle and only part of the area appeared grid distribution. When reached 228 h, the smoothness of the PA modified asphalt surface was further increased and only the middle part had grooves. When the ultraviolet radiation time reached 305 h, the surface roughness of the PA modified asphalt increased, which indicated that the control effect of the PA modifier on the ultraviolet ageing of asphalt became weakened.

3.3. TG Analysis

3.3.1. 70# Asphalt. Figure 9 shows the thermogravimetric test results of 70# asphalt at different UV ageing times. It could be seen from Figures 9 and 10 that with the extension of the UV ageing time, the initial decomposition temperature of 70# matrix asphalt decreased significantly. When the UV ageing time reached 76.25 h, the initial decomposition temperature of 70# asphalt was about 232.6 °C. When the UV ageing time extended to 305 h, the initial decomposition temperature of 70# matrix asphalt was 158.8 °C, reduced by about 31.7%, which indicated that the UV ageing effect would reduce the thermal weight loss onset temperature of asphalt, thereby accelerating the thermal weightlessness behavior of asphalt materials. From the results of DTG analysis in Figure 11, it could be analyzed that the thermal weight loss rate of 70# matrix asphalt also changed significantly after UV ageing. After 76.25 hours of UV ageing, the thermal weight loss rate of 70# asphalt was the largest, while the difference in thermal weight loss rate of asphalt subjected to other ageing times was relatively small. As for the thermal weight loss rate, the thermal weight loss rate of 70# asphalt at different UV ageing times was similar, which was about 79%.

3.3.2. PA modified asphalt. From the TG/DTG curve of PA6 modified asphalt (Figures 12–14), it could be known that the initial decomposition temperature of PA modified asphalt rose gradually with the increase of ageing time, which indicated that UV ageing treatment could increase the stability temperature of asphalt material under the high temperature. However, by comparison, the extension of the UV ageing time would lower the increase rate of the initial decomposition temperature, which demonstrated that the effect of the PA modifier on the thermal stability of asphalt was gradually degraded in the later stage of UV ageing. At the same time, the mass loss of PA6 modified asphalt reached more than 80%. With the extension of the ageing time, the weight loss rate increased significantly, reaching the maximum weight loss rate at 228 h UV ageing time, which was about 94.6%. But as the UV ageing time reached 305 h, the weight loss rate dropped to about 80%, which showed that the UV ageing time had a greater impact on the weight loss rate. However, the UV ageing time was further extended and the weight loss rate would return to the level of the initial ageing stage.

3.4. Dynamic-FTIR

3.4.1. 2D-3D Spectrogram Analysis. According to the testing results in Figure 15, the strong absorption peak position of 70# asphalt under the action of ultraviolet radiation was shown in 2800~3000 cm⁻¹ and the absorption peak in this range might be mainly derived from the stretching vibration peak of the methylene group and the stretching vibration peak of the methyl group. In addition, there were moderately strong carbonyl functional groups at 1700 cm⁻¹. The
Figure 12: TG analysis results of PA modified asphalt at different UV ageing stages: (a) 76.25 h, (b) 152.5 h, (c) 228 h, and (d) 305 h.
Figure 13: DTG analysis results of PA modified asphalt at different UV ageing stages; (a) 76.25 h, (b) 152.5 h, (c) 228h, and (d) 305h.
Figure 14: TG evaluating indices of PA modified asphalt at different UV ageing stages.

Figure 15: 2D-3D spectrogram of 70# asphalt at different UV ageing stages. (a) 76.25 h-2D. (b) 76.25 h-3D. (c) 152.5 h-2D. (d) 152.5 h-3D. (e) 228h-2D. (f) 228h-3D. (g) 305h-2D. (h) 305h-3D.
production of these functional groups was mainly due to the reaction of the internal components of the asphalt caused by the temperature rising during the dynamic FTIR test. For example, the carbonyl group was produced by the oxygen absorption reaction and the sulfoxide group was produced by the reaction of the asphalt containing sulfide.

According to Figure 16, under the effect of ultraviolet radiation, the 2D and 3D spectra of PA modified asphalt reflected the changing tendency different from that of 70# asphalt. Especially at 76.25h and 305h, the FTIR spectrum of PA modified asphalt showed obvious multiple peak changes, such as the wavenumber position of 2800–3000 cm\(^{-1}\) at 76.25h and 3500–3900 cm\(^{-1}\) at 305h. This variation of the functional group indicated that the PA modifier had an effect on the composition and proportion of the internal functional groups of asphalt during the dynamic ultraviolet ageing process, which might be the reason for the improving mechanism of the antiultraviolet ageing performance of PA modified asphalt.

### 3.4.2. Functional Group Indices.

It could be analyzed from Figure 17 that under the UV ageing time of 228h and 305h, the degree of ultraviolet ageing of 70# asphalt increased significantly. Compared with unaged asphalt, the carbonyl group content of 305h UV-aged asphalt increased by 782%, the sulfoxide group content increased by 807%, and the aromatic functional group content increased by about 594%. The exponential growth of carbonyl and sulfoxide groups was obvious, which indicated that the chemical composition
of 70# asphalt was greatly affected by ultraviolet radiation. Long-term ultraviolet radiation would accelerate the ageing of asphalt and change the chemical properties of asphalt. The PA modified asphalt would still produce a corresponding functional group under the influence of ultraviolet radiation, but its growth rate and exponential growth were obviously less than 70# asphalt. This result might indicate that the PA modifier reacted with asphalt and promoted the aromatization of asphalt components, which strengthened the internal structure stability of asphalt and inhibit the UV ageing behavior of asphalt material.

According to Figure 18, the aromatic functional group index of PA modified asphalt continued increasing stably throughout the whole ageing stage. The growth of the carbonyl group has mainly happened between 152.5h and 228h of UV ageing, which indicated that the oxidation reaction speed in this period was relatively fast and the acceleration of the combination with oxygen led to the

Figure 17: Functional group variation of 70# asphalt at different UV ageing stages: (a) $I_{\text{Ar}}$, (b) $I_{\text{C=O}}$, (c) $I_{\text{S=O}}$, and (d) Comparison.
formation of the carbonyl group. The carbonyl index decreased significantly after 305h, which might be because of the ageing film on the asphalt surface hindered the progress of the chemical reaction induced by UV radiation.

4. Conclusions

Based on the results presented in this paper, the following conclusions could be drawn:

(i) The PA modifier could promote the viscoelasticity improvement of asphalt during the ultraviolet radiation process, which could improve the antiageing performance and thermal stability of asphalt materials.

(ii) When the UV irradiation time reached 305h, the cracks began to change vertically and the depth further increased. Furthermore, the roughness of the asphalt surface increased. The addition of nano PA could alleviate the cracks on the asphalt surface caused by ultraviolet ageing.

(iii) Initial decomposition temperature of PA modified asphalt rose gradually with the extension of UV ageing time, which indicated that UV ageing treatment could increase the core temperature of
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 do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

Acknowledgments

This paper describes research activities mainly requested and sponsored by Shaxi Transportation Holdings Group Co., Ltd. Technical Project (Grant No. 19-JKKJ-20), Guangdong Basic and Applied Basic Research Foundation Project (Grant No. 2019A151510348), and Guangdong Provincial Natural Science Foundation Project (Grant No. 2019A1515101397), Guangzhou HuaHui Traffic Technology Co., Ltd. Technical Project under Grant No. 21HK0242 and Guangdong GuanYue Highway and Bridge Co., Ltd. Enterprise Mission Project under Grant No. GDKTP2021009700. That sponsorship and interest are gratefully acknowledged Special Fund for Science and Technology Innovation Strategy of Guangdong Province (Grant Nos. pdjh2021a0149, pdjh2021b0161).

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