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Investigation on Preparation Method of SBS-Modified Asphalt Based on MSCR, LAS, and Fluorescence Microscopy

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Abstract: The preparation method of SBS-modified asphalt has a direct effect on its overall performance. Currently, the optimal process is usually determined by conventional performance properties, such as softening point, ductility, and penetration, which may deviate from practical field performance. This study aims to investigate the influence of different preparation methods on the performance and microstructure of SBS-modified asphalt based on fluorescence microscopy testing, the multiple stress creep recovery (MSCR) test, the linear amplitude sweep (LAS) test, as well as Burgers model fitting. SBS-modified asphalt was prepared with different shear rates, shear temperatures, shear times, development time, and sulfur addition. The results show that the optimal process for preparing SBS-modified asphalt is 2 h of shearing at 180 °C and 4000 r/min, followed by sulfurization and 6 h of development. The performance of SBS-modified asphalt is most notably influenced by sulfurization, which forms C–S bonds to make the polymer network stronger, thereby improving the high-temperature performance as well as the fatigue resistance. However, due to high-temperature sensitivity, C–S bonds may break during development, leading to weakened performance. The performance of SBS-modified asphalt without sulfur addition shows a monotonically increasing trend with the extension of development time.

Keywords: SBS-modified asphalt; preparation method; sulfur; fluorescence microscopy; MSCR; LAS; Burgers model

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1. Introduction

The preparation method of SBS-modified asphalt has a direct effect on its overall performance [1,2]. Abundant studies have been devoted to the optimization of the preparation method of SBS-modified asphalt. Diego O. Larsen believes that the performance of SBS-modified asphalt is affected by the properties of the base asphalt, the weight average molecular weight of the SBS copolymer, and the applied shear rate [3]. Fuqiang Dong believes that there is a critical shear rate, shear temperature, and shear time to optimize the particle-size distribution and performance of SBS-modified asphalt [4]. Derya Kaya conducted the TGA test and concluded that the thermal stability of SBS-modified asphalt can be improved when the shear rate is greater than 3000 rpm; a second small degradation peak appeared at 160 °C, which shifted to the right with the increase in shear time [5]. Yue Xiao performed fluorescence microscopy experiments and concluded that higher shear temperature can improve SBS modifier uniformity and swelling area, and higher shear rate makes the physical dispersion of SBS more uniform, the specific surface area of fine particle size larger, and the area ratio increase due to sufficient swelling effect [6]

However, most work focusing on this topic is based on conventional performance properties, such as softening point, ductility, and penetration [7,8]. These indicators are generally empirical and many studies have suggested that they may deviate from actual road performance. A summary of the related literature is shown in Table 1 [9–13].

Table 1. Recommended preparation methods from the related literature.

Author	Evaluation Indicators	Shear Rate (r/min)	Shear Temp. (°C)	Shear Time (h)	Development Time (h)
Wang, Yan	Softening Point, Penetration, Ductility	—	180	1	3
HOU Yanming	Softening Point, Penetration, Ductility	3000	175	1	1
Dong Fuqiang	Softening Point, Penetration, Ductility	3000	200	1	—
Dong Yufeng	Softening Point, Penetration, Ductility	8000	180	2	1
Mojtaba Mortezaei	Softening Point, Ductility	1500	—	—	—

SBS-modified asphalt is widely used for road construction because of its excellent performance in high-temperature properties and fatigue resistance. Therefore, it appears to be more reasonable and necessary to optimize the preparation method of SBS-modified asphalt based on the corresponding performance of more advanced indicators.

High-temperature rutting resistance is one of the key performance indicators of asphalt binder. The multiple stress creep and recovery (MSCR) test proposed by D'Angelo et al. [14,15] is one of the most popular tests for high-temperature performance evaluation. To further investigate the rheological properties based on the MSCR test, the phenomenological model is frequently adopted [16]. The generalized Burgers model is one of the most widely used models. The work of Saboo et al. [17,18] reported that the Burgers model could closely approximate the creep and recovery response at any combination of loading and unloading cycles.

In terms of the fatigue performance evaluation, the linear amplitude strain (LAS) test is gaining more and more attention these days. The LAS test is a kind of accelerated fatigue test invented by Johnson et al. It applies viscoelastic continuum damage mechanics to the analysis and modeling of test results in order to predict the fatigue life of asphalt under any load condition [19–22].

Furthermore, due to the inherent incompatibility between SBS polymers and plain asphalt, the property of SBS-modified asphalt is also significantly affected by stabilizers. Sun DQ et al. [23] found that SBS-modified asphalt can be improved significantly by reactive blending of SBS and asphalt under high-share mixing at high temperature with elemental sulfur and a vulcanization accelerator. In addition to vulcanization, the production of SBS-modified asphalt usually requires a certain development for better performance; however, long-term development at high temperatures may cause SBS-modified asphalt to age and consequently lead to reduced fatigue life [24,25].

To this end, it is more reasonable and significant to optimize the preparation process of SBS-modified asphalt by using more advanced rheological performance indicators and considering additional influencing factors, such as vulcanization and development.

This study aims to investigate the impact of different preparation methods on the performance of SBS-modified asphalt. To do so, fluorescence microscopy was used to observe the microscopic morphology of the asphalt, and the MSCR test was performed to determine its high-temperature performance. The LAS test was also performed to assess its fatigue performance. Moreover, the Burgers model was also used to fit the creep and recovery curves of the MSCR test, and the instantaneous elastic deformation, delayed elastic deformation, and irrecoverable deformation were quantitatively analyzed. The research results of this article can provide a certain reference for the optimization of the preparation of SBS-modified asphalt.

2. Materials and Methods

2.1. Materials

In this study, plain asphalt ESSO 70# was used to prepare different kinds of SSB modified asphalts. Its relevant performance indexes are shown in Table 2. SBS 791-H was used for preparation. It is a kind of linear polymer with an average molecule weight of

120,000 g/mol, containing 30wt% of styrene. In order to disperse SBS more uniformly in the plain asphalt, 2% aromantic oil was added prior to the addition of SBS, and 0.15% flake elemental sulfur was used as a stabilizer.

Table 2. Main technical indicators of E70# asphalt.

Performance		Unit	The Requirement of 70# Asphalt	Result
Penetration (25 °C, 5 s, 100 g)		0.1 mm	60–80	68
Ductility (5 cm/min)	10 °C	cm	≥15	16.8
	15 °C	cm	≥100	>100
Softening Point		°C	≥46	46.2
Density (15 °C)		g/cm^3	Measured	1.030

2.2. Preparation Method

In this study, the effects of different shear rates, different shear temperatures, and different shear times on the performance of SBS-modified asphalt were investigated. Then, the effects of development time and sulfur addition were investigated based on the optimal preparation method determined above.

To develop (cure) the sheared SBS/asphalt blend, it was stirred continuously for 18 h at a speed of 600 r/min, 180 °C. The blend was sampled every 2 hours (0 h, 2 h, 4 h . . . 18 h) for examination. Two batches of SBS-modified asphalt were prepared, one with 0.16% sulfur and the other with none. The preparation plans used in this paper are depicted in Figure 1. The sample prepared by shearing at 180 °C for 2 h at a rate of 4000 r/min, with sulfur addition and zero development time was used as the control group.

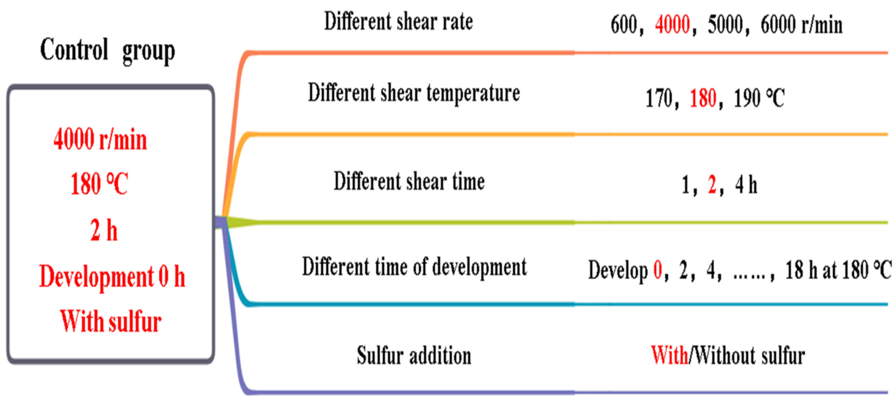


Figure 1. Different preparation methods discussed in this paper.

2.3. Materials Characterization Methods

2.3.1. Fluorescence Microscopy Test

The fluorescence microscopy test is used to observe the microscopic morphology and structure of SBS-modified asphalt obtained from different preparation methods [26]. The instrument used was the LEICA DM2700P. The dark yellow part in the attained picture represents the asphalt phase and the bright yellow part represents the swollen SBS phase [27]. Even though there are studies that use measurement software to calculate the SBS phase area [28], there is disagreement regarding threshold setting. Therefore, this paper does not calculate the SBS phase area but only uses it for visual observation.

2.3.2. MSCR Test

The MSCR test is primarily designed to evaluate the performance of modified asphalt at high temperatures [29–31]. In this paper, a dynamic shear rheometer, DHR-3, from the TA Company was used, and the test was conducted according to AASHTOM 332. Each cycle consists of 1 s of creep and 9 s of recovery. To mimic the high stress and high-temperature field conditions, only the data obtained at 3.2 kPa and a temperature ranging from 76 °C to 82 °C were analyzed. 1 The R and Jnr are calculated based on Equations (1) and (2), respectively:

$$R\% = \frac{\varepsilon_p - \varepsilon_u}{\varepsilon_p} \times 100\% \quad (1)$$

$$J_{nr} = \frac{\varepsilon_u}{\sigma} \quad (2)$$

where: ε_p is the peak strain at 1 s; ε_u is the unrecovered strain at 10 s; σ is the creep stress.

2.3.3. LAS Test

The LAS test can evaluate the fatigue performance of asphalt [32]. The paper uses the DHR-3 dynamic shear rheometer from TA Company for testing, and it is conducted in accordance with the specification AASHTO TP 101-12-UL. The LAS test protocol consists of two steps. In the first step, the general rheological properties of the asphalt are measured, and in the second step, the damaged properties of the asphalt are predicted. In this study, fatigue life (N_f) at 5% and 15% strain levels are calculated from Equation (3).

$$N_f = A(\gamma_{max})^{-B} \quad (3)$$

where: the coefficients A and B depend on the material properties calculated using the simple viscoelastic continuous loss theory (S-VECD). γ_{max} is the applied strain level (5%, 15%).

2.3.4. Burgers Model

The Burgers model is shown in Figure 2, which is composed of a Maxwell model and a Kelvin model in series. According to this model, asphalt deformation can be divided into instantaneous elastic deformation, delayed elastic deformation, and viscous deformation. The term transient elastic deformation refers to the complete recovery of the deformed part upon release of the load. The delayed elastic deformation describes the part of the deformation that gradually recovers from the deformation of asphalt over time. The viscous deformation cannot be recovered from creep deformation. In this study, the Burgers model is used to fit the loading and unloading curves of the MSCR test of asphalt samples at 76 °C. The creep stage is characterized by Equation (4), while the recovery stage is characterized by Equation (5).

$$\varepsilon = \frac{\sigma_0}{E_1} + \frac{\sigma_0}{\eta_1}t + \frac{\sigma_0}{E_2}\left(1 - e^{-\frac{E_2}{\eta_2}t}\right) \quad (4)$$

$$\varepsilon = \frac{\sigma_0}{\eta_1}t + \frac{\sigma_0}{E_2}\left(1 - e^{-\frac{E_2}{\eta_2}t}\right)e^{-\frac{E_2}{\eta_2}(t-1)} \quad (5)$$

where: ε is the deformation at time t, t is the loading time, σ_0 is the external force, E_1 is the spring stiffness of the Maxwell model, η_1 is the viscosity coefficient of the Maxwell model, E_2 is the spring stiffness of the Kelvin model, η_2 is the viscosity coefficient of the Kelvin model.

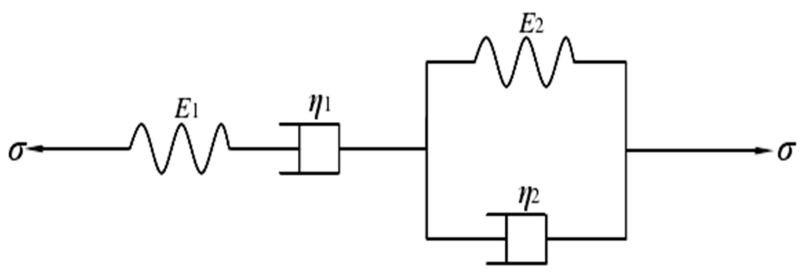


Figure 2. Burgers model.

3. Results and Discussion

3.1. Influence of Shear Rate

3.1.1. Fluorescence Microscopy Test

Samples prepared with shear rates of 600, 4000, 5000, and 6000 r/min were examined by fluorescence microscopy to investigate the morphological characteristics of the asphalt phase and SBS phase. The results are shown in Figure 3.

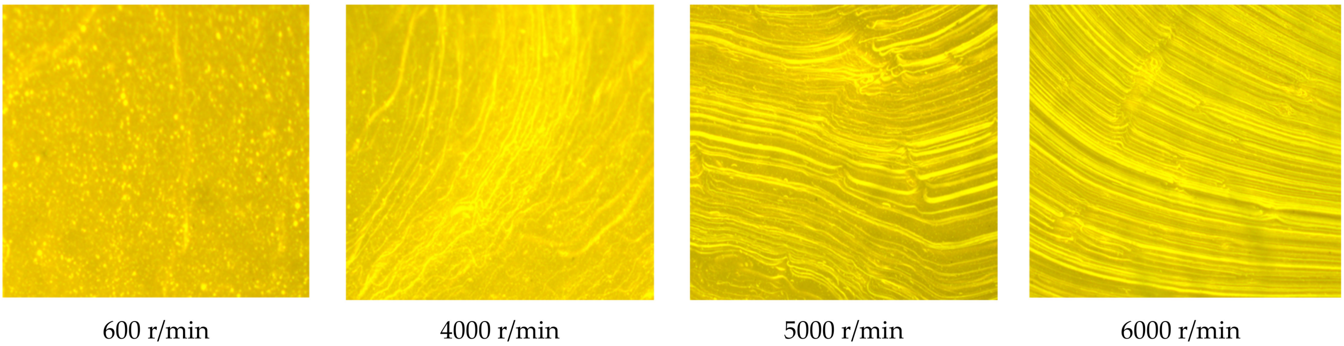


Figure 3. Fluorescence microscopy test of SBS-modified asphalt of different shear rates (100 times).

The SBS phase morphology changed from droplet (600 r/min without network structure) to the filament (4000 r/min with network structure) with an increasing shear rate. The distribution pattern became more uniform and the area ratio gradually increased.

The higher the shear rate, the smaller the SBS particle was sheared and the more evenly distributed the SBS was. It is easier for smaller SBS particles to absorb more light components to achieve a greater degree of swelling, resulting in a stronger network structure, which is beneficial to the overall performance of SBS-modified asphalt.

3.1.2. MSCR Test

For the purpose of studying the influence of shear rate on the high-temperature performance of SBS-modified asphalt, the SBS-modified asphalts were examined with the MSCR test. The calculated R and Jnr are shown in Figure 4.

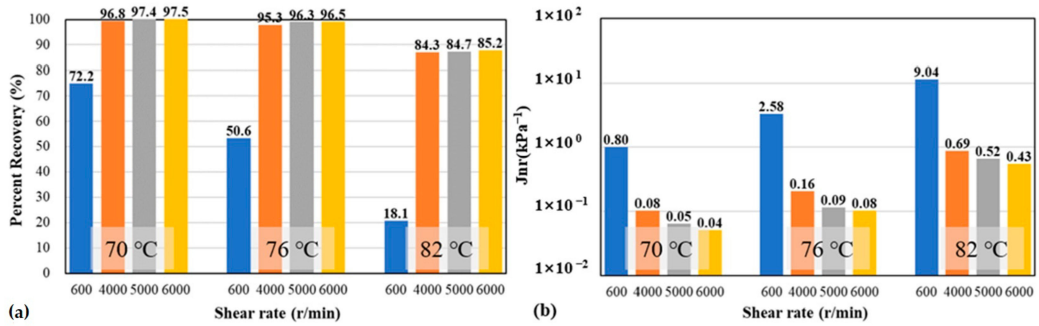


Figure 4. MSCR test results of SBS-modified asphalt under different shear rates (**a**) R, (**b**) Jnr.

According to Figure 4, the SBS-modified asphalt prepared by low-speed shear (600 r/min) did not show a strong elastic network as it shows the lowest R with the highest Jnr. Additionally, it shows the highest temperature sensitivity as its R/Jnr varies noticeably with increased testing temperatures. These results are most likely attributed to the inhomogeneous distribution of SBS.

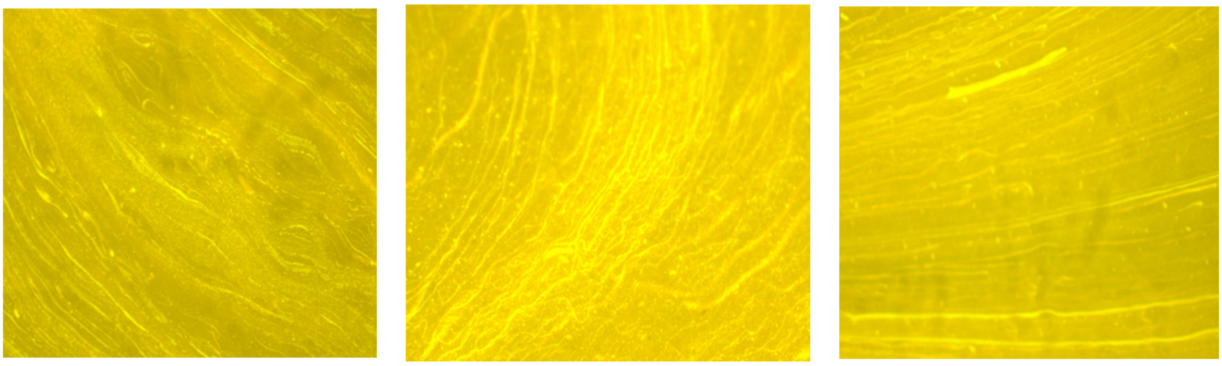
On the other hand, SBS-modified asphalt prepared by high-speed shearing (4000, 5000, 6000 r/min) demonstrates a strong and even network structure, and the SBS is uniformly distributed. Hence the R and Jnr test results are significantly better than those prepared by low-speed shearing (600 r/min). The SBS-modified asphalt prepared at 6000 r/min shows the most uniform microstructure as well as the best MSCR results (high R and low Jnr).

Although the higher the shear rate, the better the performance of SBS-modified asphalt [33], the performance of SBS-modified asphalt prepared at 6000 r/min was only slightly better than that of SBS-modified asphalt prepared at 4000 r/min. For the purpose of efficiency, the recommended shear rate in this paper is 4000 r/min.

3.2. Influence of Shear Temperature

3.2.1. Fluorescence Microscopy Test

To investigate the influence of different shear temperatures on the microstructure of SBS-modified asphalt, samples prepared at 170, 180, and 190 °C were examined by fluorescence microscopy. The results are shown in Figure 5.



170 °C 180 °C 190 °C

Figure 5. Fluorescence microscopy test of SBS-modified asphalt at different shear temperatures (100 times).

According to Figure 5, most SBS polymers show a filamentous appearance and the rest were droplets. The SBS area ratio shows a rank as follows: 180 °C > 190 °C > 170 °C. It is important to prepare SBS-modified asphalt at the appropriate temperature. At low temperatures, the molecules are not sufficiently active, which results in a low degree of blending between SBS and the asphalt. At high temperatures, the SBS molecule might be susceptible to thermal oxidative degradation, which in turn affects the strength of the polymer network structure. In this sense, 180 °C seems to be an intermediate and suitable preparation temperature.

3.2.2. MSCR Test

In order to investigate the influence of shear temperature on the SBS-modified asphalt's high-temperature performance, the above samples were sent for MSCR testing, and the calculated R and Jnr are shown in Figure 6.

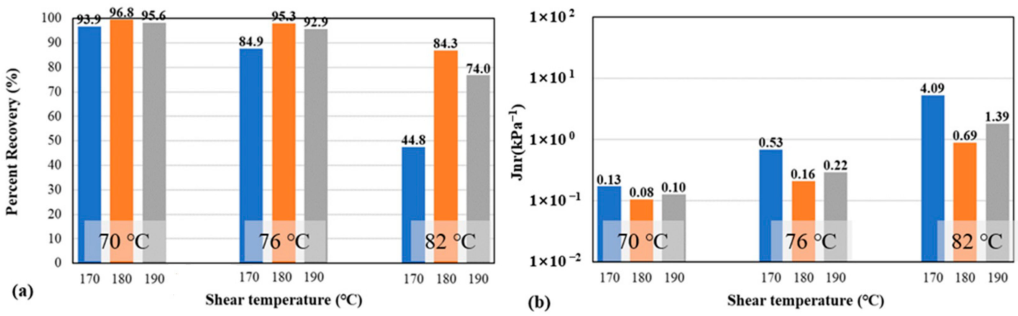


Figure 6. MSCR test results of SBS-modified asphalt under different shear temperatures (**a**) R, (**b**) Jnr.

For the SBS-modified asphalt prepared at 170 °C, due to its poor uniformity and weak network structure strength, it shows the lowest R with the highest Jnr. SBS-modified asphalt prepared at 180 °C and 190 °C show similar performance at the testing temperature of 70 °C, but when the testing temperature reaches 82 °C, the performance of SBS-modified asphalt prepared at 180 °C is superior to that of SBS-modified asphalt prepared at 190 °C.

These results indicate that the preparation temperature of 170 °C may not be high enough to facilitate the swelling and blending of the SBS polymer, while 190 °C might be too high to avoid thermal oxidative degradation of the SBS polymer. When SBS-modified asphalt is prepared at 180 °C, the best MSCR results are recorded [6]. For this reason, a shearing temperature of 180 °C is recommended.

3.3. The Influence of Shear Time

3.3.1. Fluorescence Microscopy Test

Fluorescence microscopy tests were conducted on samples sheared at 1, 2, and 4 h to determine the morphological characteristics of the asphalt phase and the SBS phase. The results are shown in Figure 7.

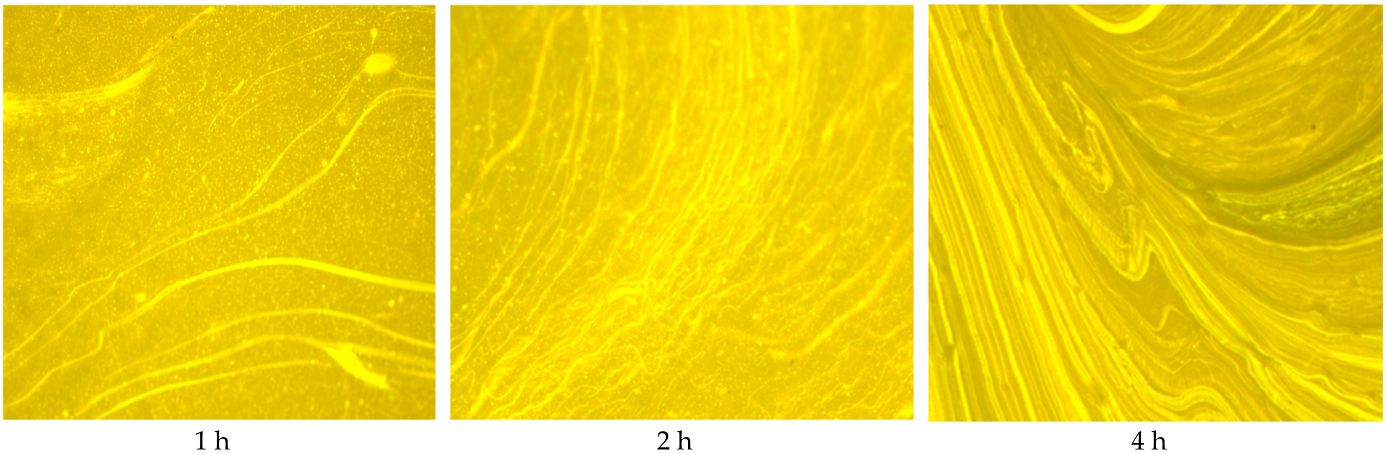


Figure 7. Fluorescence microscopy test of SBS-modified asphalt with different shearing times (100 times).

It is seen that with increasing shearing time, the SBS distribution becomes more uniform, and the area ratio gradually increases. It seems extended shearing time facilitates the swelling of SBS-modified asphalt.

3.3.2. MSCR Test

In order to study the influence of shear time on the performance of SBS-modified asphalt, the above samples were examined with the MSCR test. The calculated R and Jnr are shown in Figure 8.

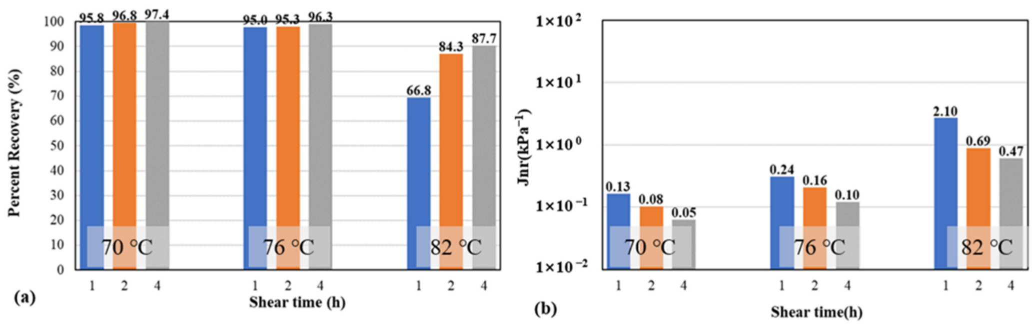


Figure 8. SBS-modified asphalt MSCR test results under different shear times (**a**) R, (**b**) Jnr.

At the temperature range of 70 °C to 82 °C, a 1 h shearing time shows the weakest elastic network, hence its MSCR results are also the worst (low R with high Jnr). SBS-modified asphalt that was sheared for 2 h and 4 h had better results, with 4 h shearing time exhibiting the best results.

Based on above results, the longer the shearing period, the more uniformly the SBS is dispersed in the plain asphalt, the greater the area ratio, and the stronger the network structure is [33]. Although the performance of SBS-modified asphalt sheared for 2 h is slightly lower than that of SBS-modified asphalt sheared for 4 h, the difference is insignificant and 4 h shearing might be too costly and ineffective during industrial production. Therefore, the recommended shearing time is 2 h.

3.4. The Influence of Sulfur and Development Time

3.4.1. Fluorescence Microscopy Test

The SBS-modified asphalt with and without sulfur was tested by fluorescence microscopy after high-temperature development [34–36]. The test results are shown in Figure 9.

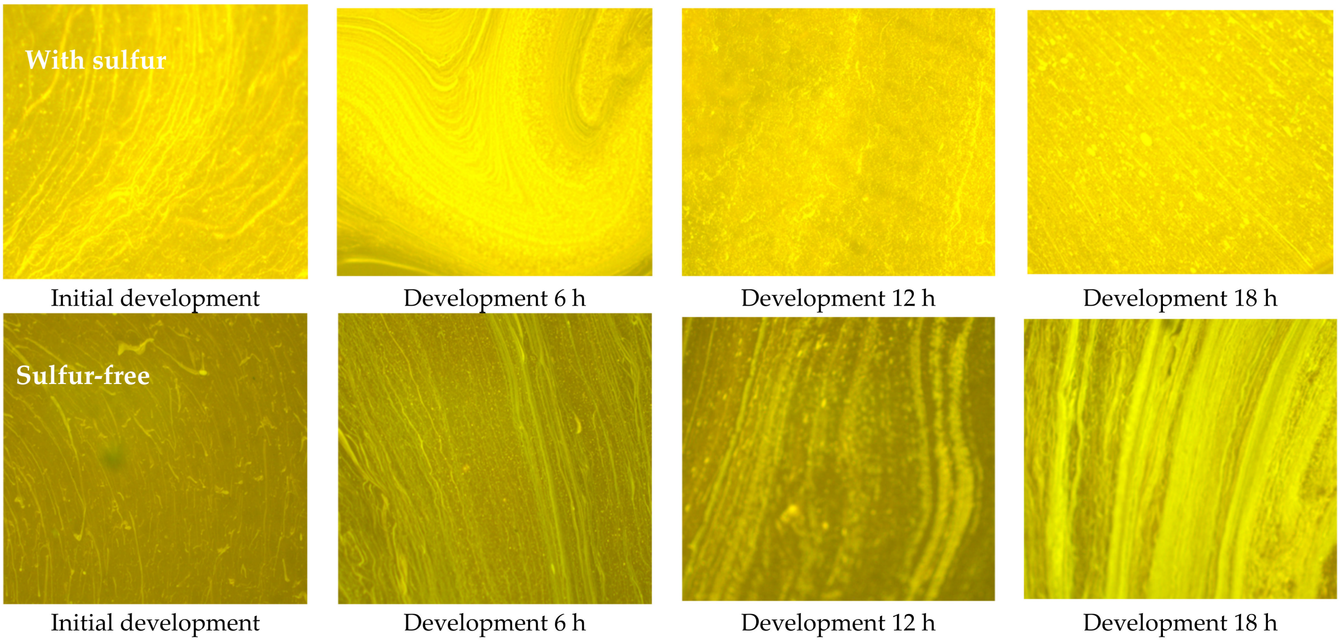


Figure 9. Fluorescence microscopy tests of SBS-modified asphalt with and without sulfur added during high-temperature development (100 times).

For SBS-modified asphalt with sulfur addition, there is a clear filamentous SBS polymer network from the start to the 6 h development time. After 6 h of development, the filamentous structure becomes unclear and it might be indicative of the degradation of the SBS polymer network.

SBS-modified asphalt without sulfur addition shows a filamentous SBS polymer structure which grows more obvious with extended development time. The width of the filamentous SBS also increases, suggesting the structure is strengthened during the development.

Related studies [37–39] have reported that the sulfur-crosslinked SBS network might be more vulnerable to thermal storage. The C–S bond introduced by sulfurization is sensitive to heat and might break during the development stage of the modified asphalt production. On the other hand, SBS-modified asphalt without sulfur does not contain heat-sensitive C–S bonds, so the development only facilitates its swelling and thus a stronger polymer network is observed with extended development time.

3.4.2. MSCR Test

Figure 10 shows the MSCR test results of SBS-modified asphalt with and without sulfur added after high-temperature development. It is first seen that at every stage of development, sulfur addition always improves the asphalt performance.

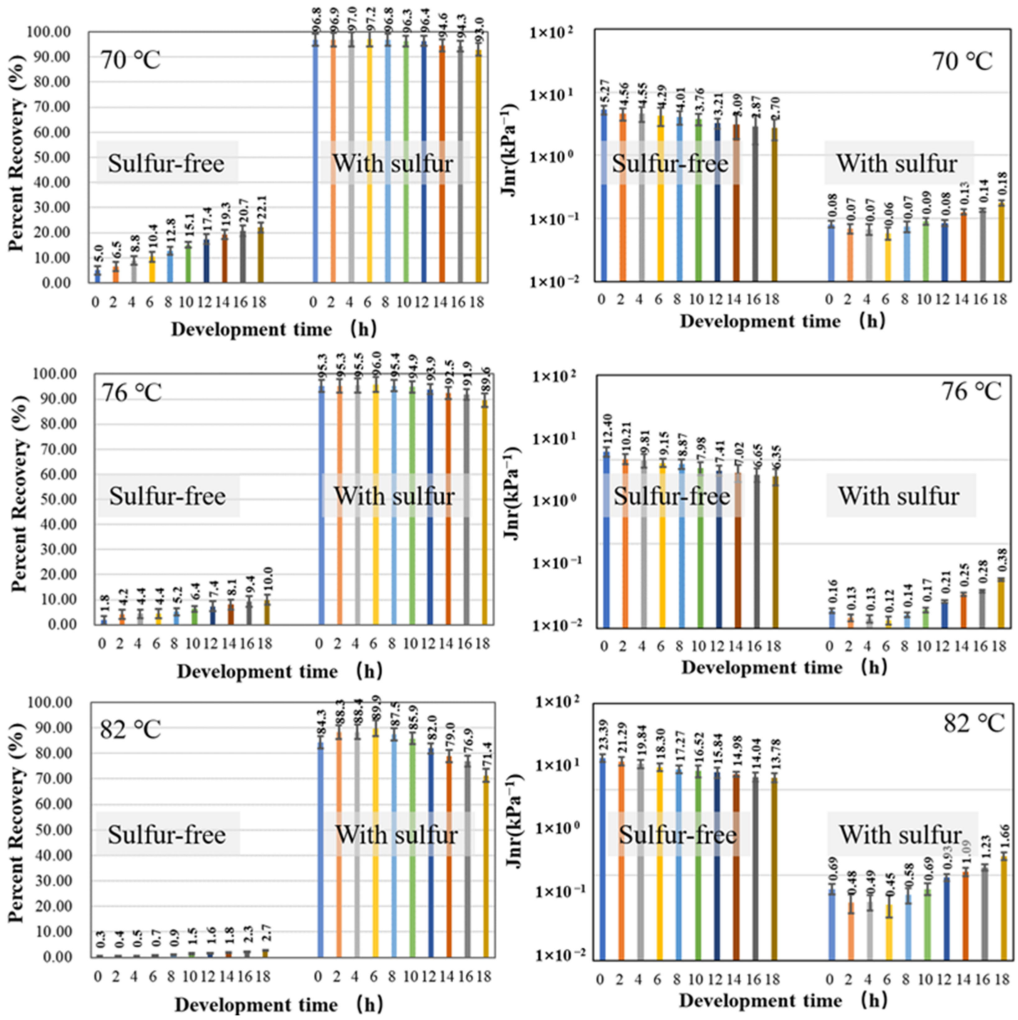


Figure 10. Comparison of MSCR test results of SBS-modified asphalt with or without sulfur.

For sulfur-free SBS-modified asphalt, the development process has a positive effect on the swelling of the SBS polymer. Thus, the longer the development process, the greater the performance improvement is.

Regarding sulfur-added SBS-modified asphalt, its performance shows a slight improve from the start to 6 h, followed by a small reduction until the end of the development process (6–18 h), rendering the h development showing the optimal performance.

When sulfur is added, it crosslinks with the asphalt and SBS polymer to form C–C and C–S bonds, which can improve the mechanical performance, but the bond energy of C–S is relatively low, making it easily broken by heat [40]. During the development process, both SBS polymer swelling and C–S bond breaks occur. On one hand, SBS swelling improves the performance of SBS-modified asphalt. On the other hand, when the development time is too long and the negative effect of the C–S bond break prevails, a reduced performance might be observed.

3.4.3. LAS Test

During extended development, the SBS-modified asphalt faces the risk of aging and weakened fatigue resistance. Therefore, the LAS test is conducted to evaluate the samples' fatigue resistance. The results are shown in Figure 11. Without sulfur, the evolution trend of fatigue life against development time is inconsistent. When sulfur is added, the fatigue life of SBS-modified asphalt first increases and then decreases. The asphalt's fatigue life reaches its maximum after 8 h of development. It is also noted that when the development time exceeds 16 h, the asphalt's fatigue life is lower than its initial state (e.g., 4423 versus 4723 for 5% strain level).

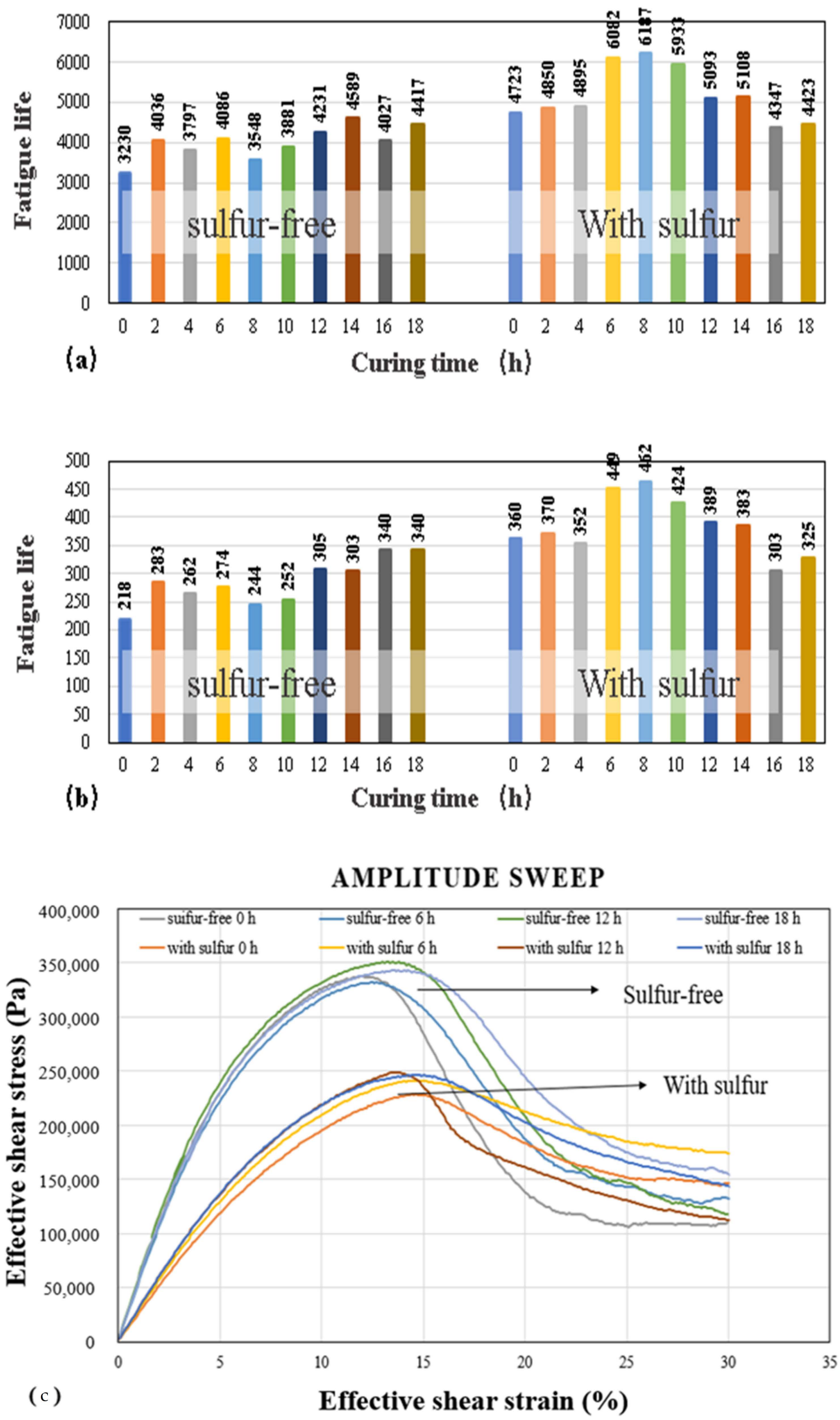


Figure 11. The effect of sulfur on SBS-modified asphalt LAS test. (**a**) 5% strain, (**b**) 15% strain, (**c**) amplitude scan.

The stress–strain curve obtained from the LAS test is shown in Figure 11c. It shows that at 25 °C, SBS-modified asphalt with sulfur has lower peak stress and stiffness, which suggests it is less likely to fracture during fatigue loading. It shows that the chemical reaction between sulfur and the SBS-modified asphalt can improve the strength and toughness of SBS-modified asphalt, thus prolonging the fatigue life [41].

Based on previous performance testing, it can be concluded that without sulfur addition, the networks structure strength, high-temperature performance, and fatigue performance of SBS-modified asphalt are significantly weakened. Sulfur crosslinks the asphalt and SBS to form C–C and C–S bonds, effectively improving the asphalt's performance [42].

However, during the development process of the sulfur-added SBS-modified asphalt, the SBS polymer swells during the high-temperature development process on the one hand, which improves the performance of the SBS-modified asphalt. On the other hand, the chemical bonds formed between sulfur and the SBS-modified asphalt are gradually destroyed due to the high temperature, resulting in impaired performance [43]. The performance of sulfur-added SBS-modified asphalt is affected by the coupling effect of these two aspects. It appears that SBS swelling dominates the first 6 h of high-temperature development. Afterwards, the degradation of the sulfurized polymer network prevails. As for the SBS-modified asphalt without sulfur, it faces less degradation during development, and thus its performance always shows an increasing trend.

3.5. ANOVA Analysis

ANOVA was performed for each influencing factor regarding the R derived from the MSCR test. The results are shown in Table 3. The p-values of all influential factors, except for different development times (without sulfur), were less than 0.05. As a result, the performance of SBS-modified asphalt is greatly affected by the preparation process.

Table 3. ANOVA analysis for each influencing factor.

Factors	F	p-Value	Significant
Shear rate	186.90	9.51×10^{-6}	Yes
Shear temperature	210.08	1.32×10^{-4}	Yes
Shear time	8938.50	7.50×10^{-8}	Yes
Development time (with sulfur)	1951.09	8.30×10^{-20}	Yes
Development time (without sulfur)	3.15	9.25×10^{-2}	No

The p-values for different development times (with sulfur) were the lowest, indicating that the swelling of SBS during high-temperature development and the cross-linking effect of sulfur have a significant effect on the performance of SBS-modified asphalt. Shear rate, shear time, and shear temperature also have a significant effect on modified asphalt properties. This is attributed to the influence of these three factors on the particle size and morphology of the modifiers in asphalt.

The p-values of different development times (without sulfur) are more than 0.05, indicating that high temperature development has no significant effect on the performance of modified asphalt without sulfur. The contrast of different development time (with/without sulfur) highlights the significance of sulfur addition during preparation.

3.6. Burgers Model

3.6.1. Viscoelastic Parameters

The Burgers model was employed to further investigate the creep and recovery behavior characterized by the MSCR test. The creep and recovery curve of 3.2 kPa MSCR was fitted using the Burgers model, and the testing temperature was 76 °C.

The obtained parameters of the Burgers model are shown in Table 4. E_1 represents the stiffness of the instantaneous elastic deformation part of the model, and the instantaneous elastic deformation is inversely proportional to the stiffness. Therefore, the smaller the E_1, the greater the instantaneous elastic deformation. η_1 is the viscosity of the dashpot in the model, which is inversely proportional to the unrecoverable viscosity deformation in the fitting formula. The larger the η_1, the smaller the unrecoverable deformation, and the better the asphalt's permanent deformation resistance. E_2 and η_2 are the stiffness and viscosity of the parallel spring and dashpot in the model. E_2 and η_2 have a combined effect on delayed elastic deformation. In the fitting formula, the delayed elastic deformation is proportional to E_2/η_2 [44].

Table 4. Viscoelastic parameters of SBS-modified asphalt Burgers model under different preparation factors.

		E_1	η_1	E_2	η_2	E_2/η_2	R^2
Control group	180 °C 2 h 4 kr/min	10,753.81	45,463.02	3111.48	2818.09	1.10	0.998
Shear temp	170 °C	11,893.32	26,875.47	2884.09	3215.36	0.90	0.999
	190 °C	11,730.07	31,649.12	2895.69	3120.81	0.93	0.998
Shear time	1 h	11,306.03	43,955.77	3045.24	2873.82	1.06	0.998
	4 h	10,006.02	35,684.36	3242.67	2810.33	1.15	0.997
Shear rate	0.6 kr/min	16,679.76	16,679.76	3006.52	4367.08	0.69	0.995
	5 kr/min	10,471.18	46,808.48	3303.54	2916.80	1.13	0.998
	6 kr/min	10,066.70	48,072.47	3441.86	2875.24	1.20	0.998
With sulfur and development	2 h	10,604.07	46,056.28	3344.01	2952.62	1.13	0.997
	4 h	10,393.33	47,056.93	3316.30	2917.26	1.14	0.997
	6 h	10,062.92	50,057.22	3509.16	2941.64	1.19	0.996
	8 h	10,471.50	48,414.18	3438.70	3060.36	1.12	0.996
	10 h	11,018.38	38,875.70	2839.78	3137.07	0.91	0.998
	12 h	11,428.87	35,474.39	2718.87	3313.29	0.82	0.997
	14 h	11,836.66	28,201.39	2593.87	3472.96	0.75	0.997
	16 h	12,562.11	20,889.82	2544.19	3536.98	0.72	0.998
	18 h	13,249.11	18,101.61	2534.70	3693.99	0.69	0.997
Without sulfur and development	0 h	2,125,361,005.26	3311.92	67,262.80	103,170.29	0.65	0.998
	2 h	2,125,277,467.87	3378.16	57,149.72	84,043.71	0.68	0.997
	4 h	2,125,067,312.98	3426.98	48,912.93	68,891.45	0.71	0.995
	6 h	2,124,971,149.91	3463.64	24,788.23	34,351.01	0.72	0.997
	8 h	2,124,957,213.26	3481.19	22,713.78	31,546.92	0.72	0.997
	10 h	2,124,906,147.42	3551.98	18,567.98	25,091.86	0.74	0.996
	12 h	2,124,836,164.03	3597.67	16,371.09	21,901.41	0.75	0.998
	14 h	2,124,471,819.42	3624.29	15,263.32	20,083.32	0.76	0.998
	16 h	2,124,857,872.72	3693.38	13,865.74	18,007.45	0.77	0.997
	18 h	2,124,910,622.34	3743.54	12,653.30	16,135.94	0.78	0.997

3.6.2. Strain Attributed to Different Components in Burgers Model

The asphalt samples strain in three ways confronting loading. The sum of instantaneous elastic deformation and delayed elastic deformation is the recoverable deformation. The larger the recoverable deformation ratio, the better the elasticity and hence, general mechanical performance. Viscous deformation is the unrecoverable part. The smaller the viscous deformation, the better the asphalt deformation resistance.

The distribution of these three types of deformation during the entire loading process is shown in Figure 12. The SBS-modified asphalt prepared with 2 h shearing at 180 °C and RPM 4000 r/min was used as the control group, and the viscous deformation ratio of the control group is labeled with a black horizontal line.

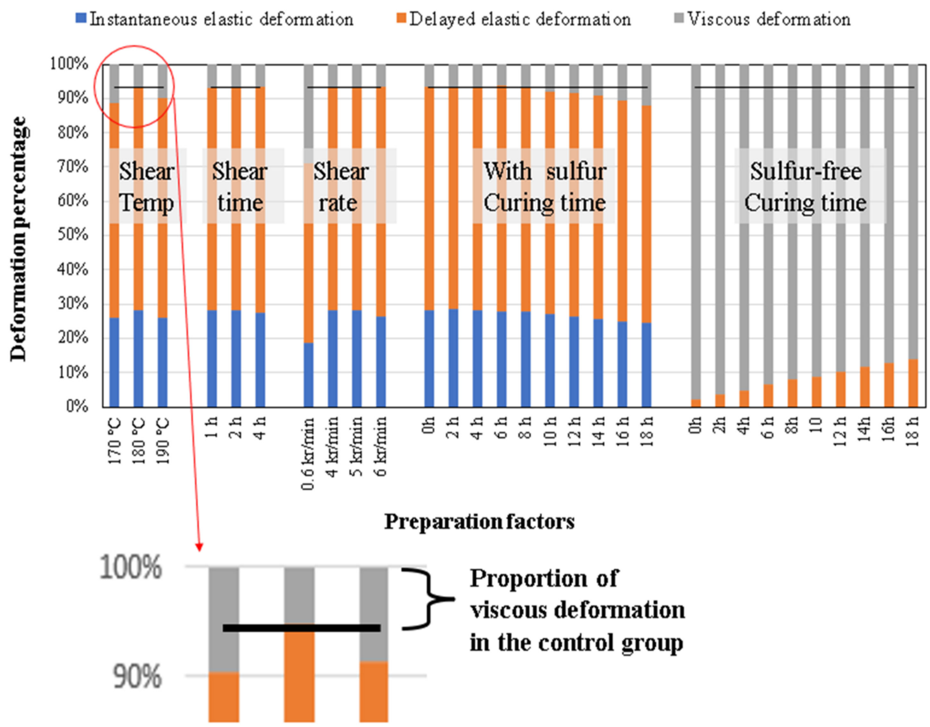


Figure 12. Cumulative proportions of the three deformations at the end of loading.

Based on Figure 12, the viscous deformation rank of shearing temperature is 170 °C > 190 °C > 180 °C, suggesting 180 °C is the optimal preparation temperature. When it comes to shear time, the rank is: 1 h > 2 h > 4 h, suggesting the longer the shear time, the better the SBS-modified asphalt performance. It is also observed that the higher the shear rate, the smaller the viscous deformation is.

Regarding the influence of the development time for the sulfur-added SBS-modified asphalt, the proportion of viscous deformation decreased from 0 h to 6 h and then increased from 6 h to 18 h. The SBS-modified asphalt developed for 18 h has a larger proportion of viscous deformation than that of zero development time, indicating that the fully developed asphalt has an even lower high-temperature performance. For SBS-modified asphalt without added sulfur, the instantaneous elastic deformation is almost zero, and the viscous deformation decreases with the increase of development time. Thus, SBS-modified asphalt without sulfur always shows better performance with extended development time. These data are consistent with the MSCR testing results.

4. Conclusions

This study investigated the influence of different preparation methods on the critical performance and microstructure of SBS-modified asphalt. Different variables, including shearing rate, shearing temperature, shearing time, development time, and sulfur addition were considered. The LAS test and MSCR test were conducted to evaluate the asphalt's high-temperature resistance and fatigue resistance. Moreover, the Burgers model was employed for viscoelasticity analysis. The detailed conclusions are as follows.

- The recommended preparation method is 2 h of shearing at 180 °C, 4000 r/min, with sulfur addition and developing for 6 h. Sulfur addition and the shear rate have the most significant impact on the performance of SBS-modified asphalt;
- The microstructure captured by the fluorescence image shows that an intermediate shearing temperature, high shearing rate, and extended shearing time render the distribution of SBS polymer more even;
- Sulfurized SBS-modified asphalt shows optimal performance after 6 h of development (25% decrease in Jnr and 28.7% increase in N_f), and extended development hinders its

performance (137.5% increase in Jnr and 6% decrease in N_f), which is due to the heat sensitivity of the C–S and S–S bonds;

- Without sulfur, SBS-modified asphalt shows monotonously improving performance with extended development time (48.7% decrease in Jnr and 36.7% increase in N_f). However, after 18 h of development, it still demonstrates much lower high-temperature performance and slightly lower fatigue performance than sulfurized SBS-modified asphalt.

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