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Research Article

High- and Low-Temperature Properties and Thermal Stability of Silica Fume/SBS Composite-Modified Asphalt Mortar

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Recently, China has started paying more attention to environmental protection, and the efficient utilization of exhaust gases produced by smelting has emerged as a key problem concern. The silica fume collected from the exhaust gases produced by smelting ferrosilicon or industrial silicon was often used as a cement concrete admixture. Using silica fume as an asphalt modifier can make exhaust gases profitable. In this study, silica fume/SBS composite-modified asphalt mortar was prepared to improve the performance of asphalt. The effects of the silica fume content, temperature, and ratio of filler asphalt on the composite-modified asphalt mortar were studied through the cone penetration, softening point, viscosity, dynamic shear rheological (DSR) test, and bending beam rheometer (BBR) test. The thermal stability of composite-modified asphalt was analyzed through the thermal analysis test. The results showed that with the increase of silica fume content and ratio of filler asphalt, the high-temperature performance of asphalt mortar was improved; the content of silica fume had a great influence on the low-temperature performance of asphalt mortar, the optimum silica fume content was 7%; increasing the ratio of filler asphalt reduced the low-temperature cracking resistance of asphalt mortar; the incorporation of silica fume enhanced the initial decomposition temperature, thermal residual rate, temperature in which the weight loss rate reaches the maximum, and the endothermic peak of maximum temperature and improved the properties of asphalt.

1. Introduction

Asphalt pavement constitutes more than 90% of the expressway in China for its good smoothness, low noise, convenience, and ease of maintenance [1, 2]. Therefore, the study of the high-quality modified asphalt and its mixtures has become an important research area in the field of road traffic, for reducing the early disease and prolonging the service life of asphalt pavement. SBS-modified asphalt is widely used in highway construction because of its excellent comprehensive performance. However, rutting, translation, bleeding, shoving, bulging and cracking, aging, and other diseases arising from the use of the pavement are also difficult problems that have been plaguing road practitioners for long [3–5]. Therefore, it is imperative to study the composite-modified asphalt mixture to meet the requirements of high flow, heavy load, stability, durability, and high strength.

It has been found that silica fume has many properties, such as abundant reserves, low cost, large specific surface area, low thermal conductivity, strong thermal stability, and strong adsorption capacity. It has great advantages in improving asphalt's high- and low-temperature performance, fatigue performance, and water stability [6–11]. Wei et al. studied the diatomite and SBS composite-modified asphalt by using regression analysis and a modified grey model. They found that diatomite could reduce the air void and improve the indirect tensile strength and indirect tensile stiffness modulus of crumb rubber-modified stone mastic asphalt (SMA) [12]. Luo et al. analyzed a modified mechanism of diatomite-modified asphalt by using the infrared (IR) spectrum and scanning electron microscope (SEM). They also inferred a model diagram of the modification process with four components of asphalt [13]. Tan et al. carried out the low-temperature bending test, low-temperature compression test, and contraction coefficient test of matrix asphalt mixture and diatomite-modified asphalt mixture. Their test results indicated that the addition of silica fume could improve the low-temperature performance and increase the contraction coefficient of the mixture [14].

In this paper, the preparation of silica fume/SBS composite-modified asphalt was investigated. The high-temperature performance and low-temperature performance of silica fume/SBS composite-modified asphalt mortar were evaluated by the cone penetration, softening point, viscosity, DSR, and BBR. The mechanism of modification and the microscopic characteristics of composite-modified asphalt were analyzed through a thermal analysis test.

2. Test Preparation

2.1. Materials. The matrix asphalt used in this experiment was AH-90 (Panjin) asphalt, and the modifiers were made of SBS (4601, LCY Rubber Co., Ltd., Huizhou, Guangdong Province of China) and silica fume (Anmei International Trade Industrial Development Co., Ltd., Anshan, Liaoning Province of China). The technical indicators of the raw materials are shown in Table 1.

Table 1: Indicators of raw materials.

Materials	Property	Value
Matrix asphalt	Penetration (25°C, 0.1 mm)	83
	Ductility (15°C, cm)	>100
	Softening point (°C)	46
	Penetration index	−1.4
	Dynamic viscosity (60°C, Pa·s)	175
	Density (g/cm^3)	1.004
SBS	Block ratio	30/70
	Tensile strength (MPa)	21.6
	Elongation at break (%)	850
	Relative molecular mass	28×10^4
Silica fume	Appearance	Grey
	SiO_2 content (%)	>92
	pH	6~8
	Particle size distribution (μm)	0.1~0.3
	Granularity (mesh)	300~500
	Loose density (g/cm^3)	0.1~0.2
	Tight heap density (g/cm^3)	0.3~0.4
	Specific surface area (m^2/g)	>25
	Burning loss (%)	<5
	Impurity content (%)	<8
	Water content (%)	<5

2.2. Preparation of Composite-Modified Asphalt. In this study, a high-speed shear stirred emulsifier (FLUKOAF25) for modified asphalt was used to prepare modified asphalt. Firstly, the matrix asphalt was heated to a molten state. Then, SBS (mass fraction 4%) and dry silica fume were added into the matrix asphalt. Shearing was initiated at a speed of 5000 r/min at 170°C to 190°C. The shearing duration was performed for one hour. Finally, the finished silica fume/SBS composite-modified asphalt was made by stirring for 30 min. Composite-modified asphalt mortars were obtained by adding fillers into the modified asphalt.

The effects of silica fume content on composite-modified asphalt were studied by fixing the ratio of filler asphalt at 1.0 and varying the amount of silica fume (mass fraction 1%, 4%, 7%, 10%, and 13%). Similarly, the effects of the ratio of filler asphalt on composite-modified asphalt were studied by fixing the content of silica fume at 7% and varying the ratio of filler asphalt (0.6, 0.8, 1.0, 1.2, and 1.4).

2.3. Experimental Methods

2.3.1. Cone Penetration Test. Silica fume/SBS composite-modified asphalt mortar is a heterogeneous material, and the discreteness of data would be very large if the conventional penetration test is used to evaluate its denseness [15–17]. In this paper, the cone penetration test was used to evaluate the consistency of the asphalt mortars. The angle of the cone was 30°, and the cone was made of stainless steel. The inner diameter of the dish was 70 mm, and the inner depth was 45 mm. The total weight of the cone, the connecting rod, and the balancing weight were 195 g. The test temperature was 25°C (ASTM D5/D5M-2013).

2.3.2. Softening Point Test. The softening point test is another test that can be used to measure the denseness of the asphalt mortar. The ring and ball method was used to determine the softening point (ASTM D36/D36M-2014).

2.3.3. Brookfield Viscosity Test. Brookfield viscosity is an important index of rheological properties of asphalt. The high-temperature performance of asphalt is evaluated by measuring the torque of the shaft rotating at certain speed in the asphalt. In this study, the test was conducted at a temperature of 177°C (ASTM D4420).

2.3.4. Dynamic Shear Rheological (DSR) Test. The flow characteristics of the polymer materials are evaluated by rotational shearing at a certain speed. In this paper, a stress control mode was adopted. The stress level was 0.1 kPa, and angular frequency was 10 rad/s. The diameters of the samples were 25 mm (the thickness is 1 mm) and 8 mm (the thickness is 2 mm) (AASHTO T315-09).

2.3.5. Bending Beam Rheometer (BBR) Test. The flexural creep stiffness (S) and creep rate (m) were obtained by studying the stiffness modulus of a bituminous beam under creep load in the BBR test. The low-temperature cracking resistance of asphalt mortars is evaluated from S and m. The size of the bituminous beam: the length was 102.00 mm, the width was 12.70 mm, and the height was 6.35 mm (AASHTO T313-09).

2.3.6. Thermal Analysis Test. These experiments were conducted in the analysis and testing center of Northeast Forestry University. The data were acquired using the synchronous thermogravimetric analyzer (STA409PC,

Figure 1: Silica fume.

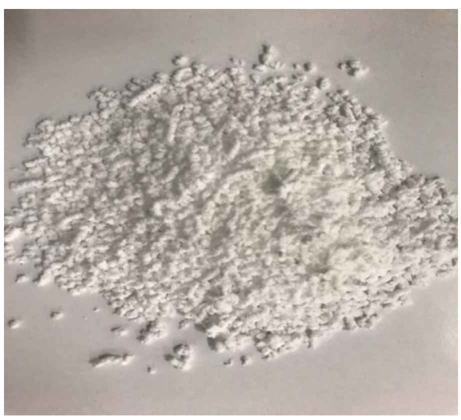

Figure 2: SBS.

NETZSCH Instrument Company, Germany). The atmosphere was nitrogen gas atmosphere, the rate of ventilation was 120 ml/min, the heating rate was 20°C/min, and the range of temperature was 35~600°C Figures 1 and 2.

3. Results and Discussion

3.1. Cone Penetration. As could be seen from Figure 3, the cone penetration of the asphalt mortar decreased with the increase in the content of silica fume. The results showed that the shear strength of the asphalt mortar was enhanced by the addition of silica fume. The saturated and aromatic components in the asphalt were sucked into the pores of silica fume, which increased the content of asphaltene and pectin relatively. Thus, asphalt became thicker, the shear strength increased, and the cone penetration decreased [18, 19]. Figure 4 shows that the cone penetration of asphalt mortar decreased with the increase in filler content. It could be seen that increasing the ratio of filler asphalt greatly improves the high-temperature performance of asphalt mortar.

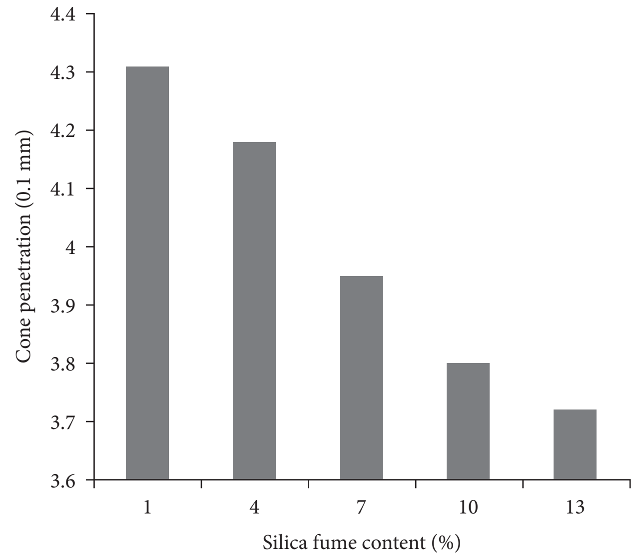

Figure 3: Influence of the content of silica fume on the cone penetration.

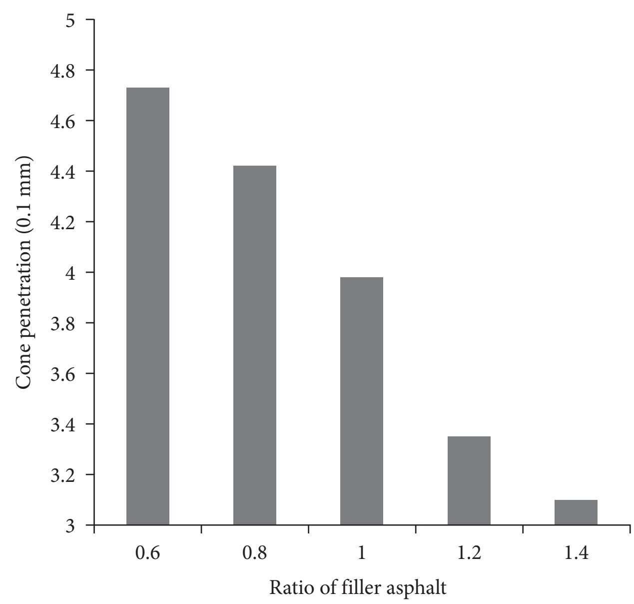

Figure 4: Influence of the ratio of filler asphalt on the cone penetration.

3.2. Brookfield Viscosity at 177°C and Softening Point. According to the data in Figure 5, the viscosity at 177°C and the softening point of silica fume/SBS composite-modified asphalt mortar increased obviously with increasing the content of silica fume. These data suggested that the small particle size and large specific surface area of silica fume cause the particles to come in closer contact with the asphalt. The strong adsorbability of silica fume was beneficial in strengthening the intermolecular force and increasing the thickness of the structural asphalt [18]. Thus, the coherence between the asphalt and the aggregate was enhanced, and the high-temperature stability of the asphalt mortar was improved.

As is shown in Figure 6, the softening point and the Brookfield viscosity of the asphalt mortar were improved greatly because of the increase in the ratio of filler asphalt. It was similar to the researches conducted by Wu et al. [20] and

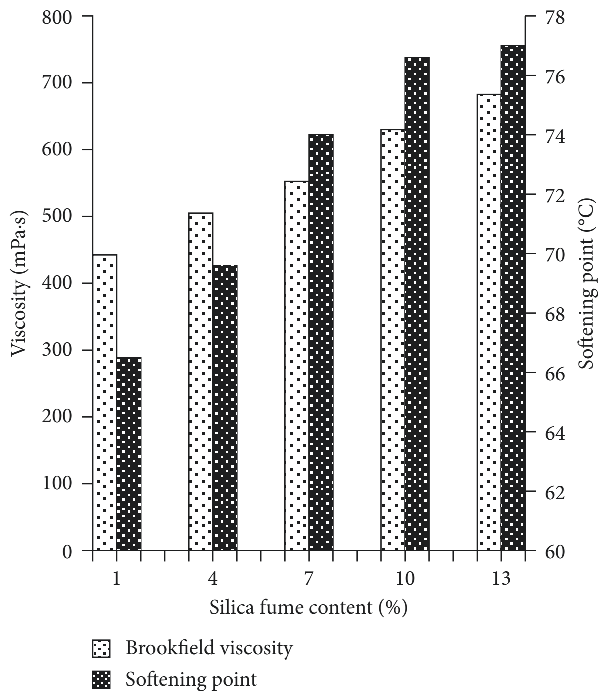

Figure 5: Influence of the content of silica.

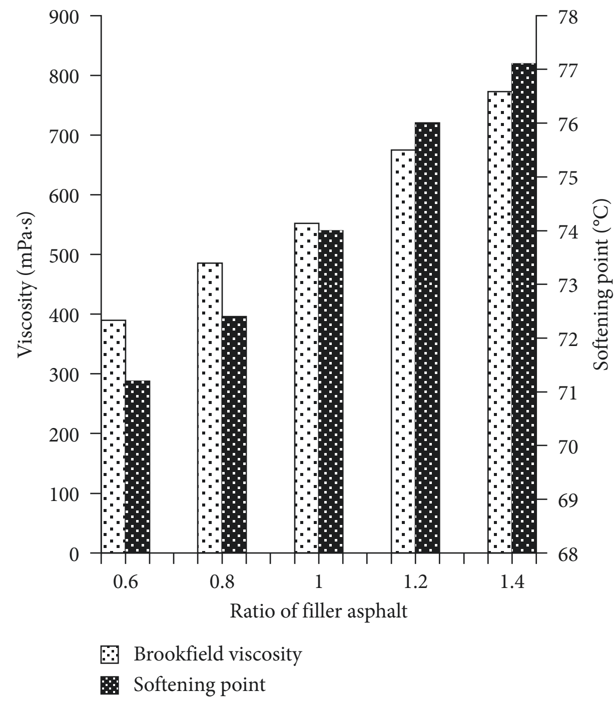

Figure 6: Influence of the ratio of filler asphalt on softening point and Brookfield viscosity.

Zhang et al. [21]. They assumed that the increase of the content of filler would enhance the adsorption between asphalt and filler. Therefore, the ratio of structural asphalt was increased, and the viscosity and strength of asphalt were enhanced.

3.3. DSR. According to the data from Table 2, as the content of silica fume increased, the complex shear modulus (G^*) increased gradually, whereas the phase angle (δ) decreased gradually. It could be concluded that the addition of silica fume enhanced the ability of asphalt mortar to resist stress, increased the relative proportion of the elastic components in the asphalt, and reduced the permanent deformation. The rutting factor ($G^*/\sin\delta$) increased with the increase in the content of silica fume. With increase in the content of silica fume, the ability of the asphalt mortar to resist the high-temperature rutting increased.

When the content of silica fume was the same, through comparing G^* and δ of 58°C, 64°C, 70°C, 76°C, and 82°C, it was found that with the increase in the test temperature, the complex shear modulus (G^*) of asphalt mortar decreased and δ increased. The results showed that the asphalt mortar was a typical temperature-sensitive material. The change in temperature could change the ability of asphalt mortar to resist stress rapidly. As the temperature increased, some of the elastic components of the asphalt mortar changed into viscous components, resulting in the decrease of $G^*/\sin\delta$, which indicated the decline of high-temperature performance of the asphalt mortar.

The relationship between the rutting factor and the ratio of filler asphalt is illustrated in Figure 7. It could be observed that the rutting factor ($G^*/\sin\delta$) increased gradually with the increase in the ratio of the filler asphalt. It is indicated that the increase of the content of filler is helpful to improve the high-temperature performance of the asphalt mortar. Adding filler into asphalt mortar would increase the volume of mortar significantly because of the specific surface area of the filler. Therefore, the proportion of structural asphalt would increase and the cohesive force of asphalt mortar would be improved. Thus, the high-temperature stability of the asphalt mortar would be improved.

In order to obtain rheological properties of asphalt in wider frequency range, the frequency sweep test was carried out. As is shown in Figures 8 and 9, with the increase of frequency, the complex modulus of three kinds of asphalt increased. However, the phase angle had a downtrend as a whole. The complex modulus of silica fume/SBS composite-modified asphalt at low frequency (high temperature) was larger than SBS-modified asphalt, and the complex modulus was smaller than SBS-modified asphalt at high frequency (low temperature). On the contrary, it was proved that the addition of silica fume further improved the high- and low-temperature performance of asphalt.

3.4. BBR. The creep rate (m) reflects the stress relaxation performance of the asphalt mortar and the sensitivity of the stiffness with time, and the creep stiffness (S) reflects the flexibility of the mortar. The higher the value of m was, the better the anticracking performance of the asphalt mortar at low temperature would be. Moreover, the smaller the S was, the better the flexibility of the asphalt mortar at low temperature would be. As could be seen from Figure 10, with the increase in silica fume content, the bending creep stiffness of asphalt mortar at −12°C, −18°C, and −24°C

Table 2: Results of DSR.

Test parameters	Temperature (°C)	Silica fume content (%)				
		1	4	7	10	13
G^* (kPa)	58	19.8	20.091	20.125	20.156	20.847
	64	9.716	9.847	10.123	10.234	10.319
	70	4.966	5.086	5.222	5.229	5.286
	76	2.693	2.724	2.771	2.771	2.805
	82	1.531	1.538	1.544	1.556	1.58
Δ (°)	58	76.14	76.12	75.52	75.07	74.33
	64	79.3	78.81	77.6	77.17	76.66
	70	81.44	81.33	79.85	78.89	78.39
	76	84.46	83.88	83.2	81.19	79.66
	82	84.94	84.71	83.97	82.56	80.45
$(G^*/\sin\delta)$ (kPa)	58	20.394	20.695	20.785	20.860	21.652
	64	9.888	10.038	10.474	10.496	10.605
	70	5.022	5.145	5.305	5.329	5.396
	76	2.706	2.740	2.791	2.804	2.872
	82	1.537	1.545	1.553	1.569	1.602

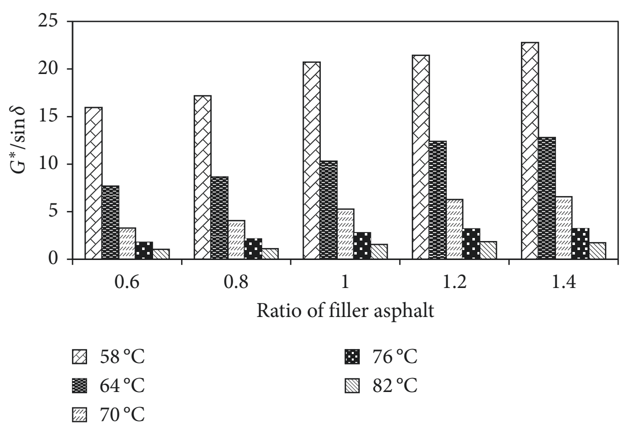

Figure 7: Influence of the ratio of filler asphalt on the rutting factor.

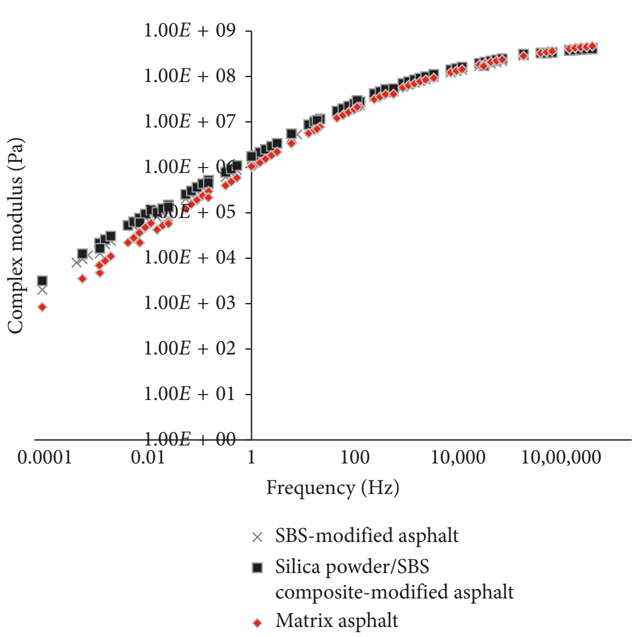

Figure 8: Complex modulus master curve.

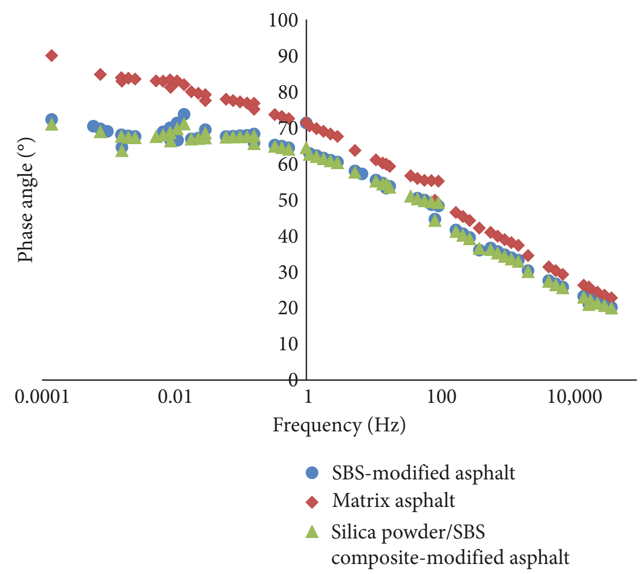

Figure 9: Phase angle master curve.

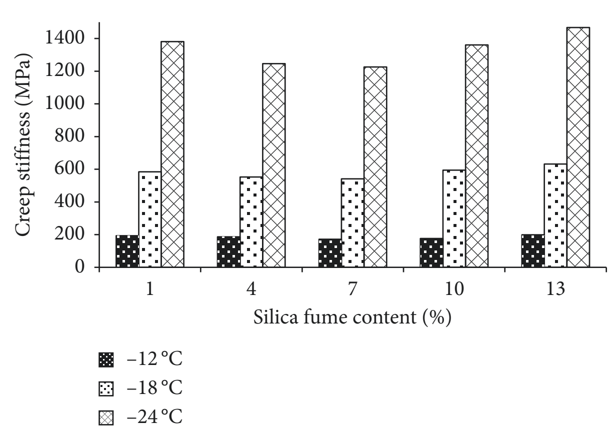

Figure 10: Influence of the content of silica fume on the creep stiffness.

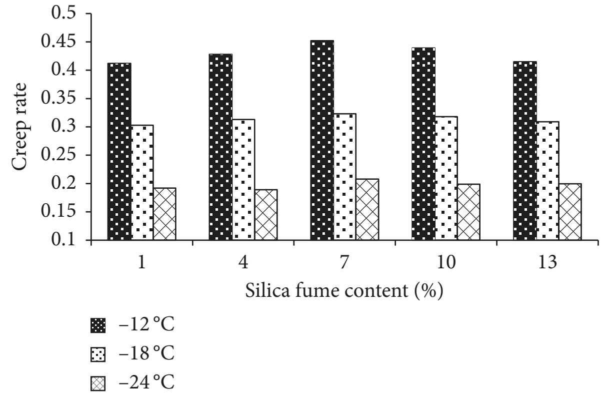

Figure 11: Influence of the content of silica fume on the creep rate.

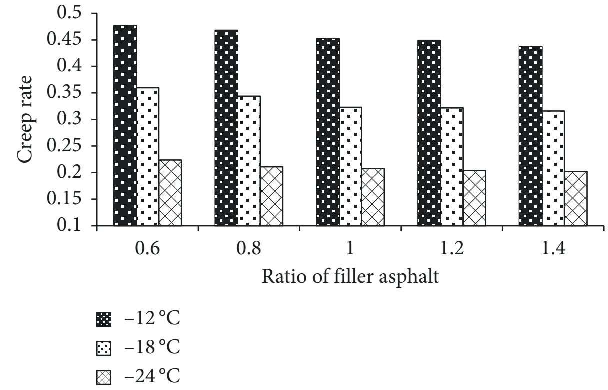

Figure 13: Influence of the ratio of filler asphalt on the creep rate.

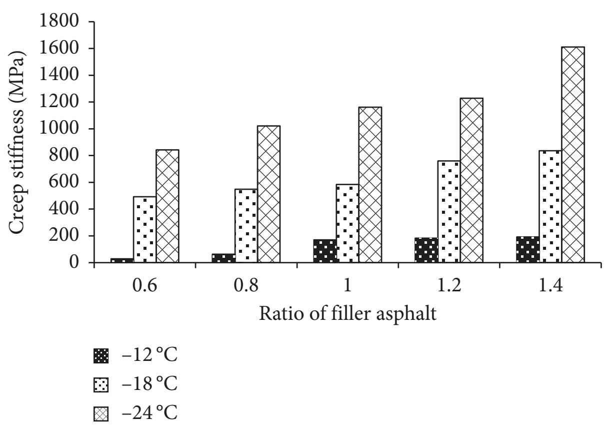

Figure 12: Influence of the ratio of filler asphalt on the creep stiffness.

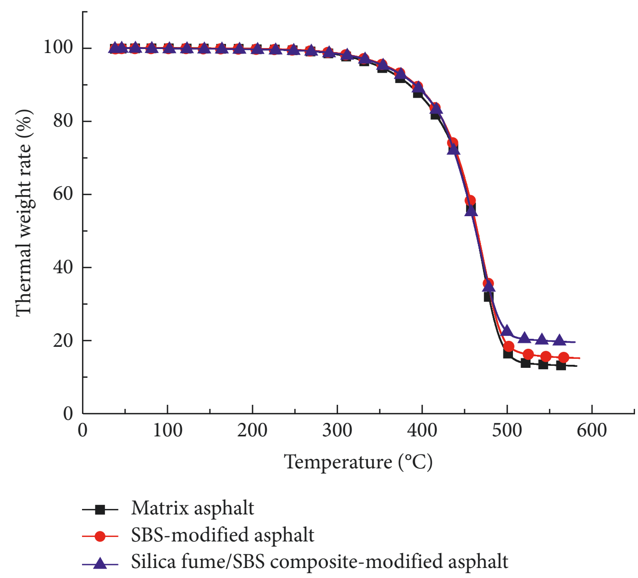

Figure 14: TGA curve.

decreased initially and then increased, indicating that a small amount of silica fume was helpful to improve the low-temperature anticracking performance, however, more than 7% would damage its low-temperature performance. It could be seen from Figure 11 that the creep rate of asphalt mortar increased initially and then decreased with the increase in the silica fume content, indicating that the stress relaxation ability was the best for approximately 7% silica fume content. Therefore, the recommended percentage of silica fume is not more than 7%.

Figures 12 and 13 show that the bending creep stiffness of the asphalt mortar increased and the creep rate decreased with the increase in the ratio of silica fume. It was indicated that the addition of filler would reduce the fluidity of structural asphalt and reduce the flexibility of the mortar. Therefore, the increase in the ratio of filler asphalt was not beneficial to the improvement of the anticracking performance of the asphalt mortar at low temperature.

4. Thermal Analysis Test

The thermogravimetric test will generate a thermogravimetric analysis (TGA) curve, which takes the temperature or time as the transverse coordinate and the mass change as the longitudinal coordinate. The pyrolysis properties of the asphalt, such as the initial decomposition temperature, maximum decomposition rate, and the mass residual rate, can be obtained from the TGA curve. The differential thermogravimetry (DTG) curve is the first derivative of the TGA curve to temperature (or time), and it reflects the relationship between the change rate of the quality of asphalt (dm/dt) and temperature (or time): $dm/dt = f(T)$ or $f(t)$. The rate of weight loss can be analyzed using the DTG curve. The differential scanning calorimetry (DSC) test shows that the peak, peak width, and peak area of the DSC curve are closely related to the microstructure characteristics of the sample. The macroperformance of composite-modified asphalt is mainly affected by the aggregation state of asphalt and modifier, while the aggregation state of asphalt and modifier in the range of endothermic peak temperature is also reflected in the location and area of endothermic peak [22]. The thermal analysis curves of matrix asphalt, SBS-modified asphalt, and silica fume/SBS composite-modified asphalt are shown in Figures 14–16.

The analysis of the TGA curve showed that the thermal weight loss rate of the matrix asphalt was 100% when the

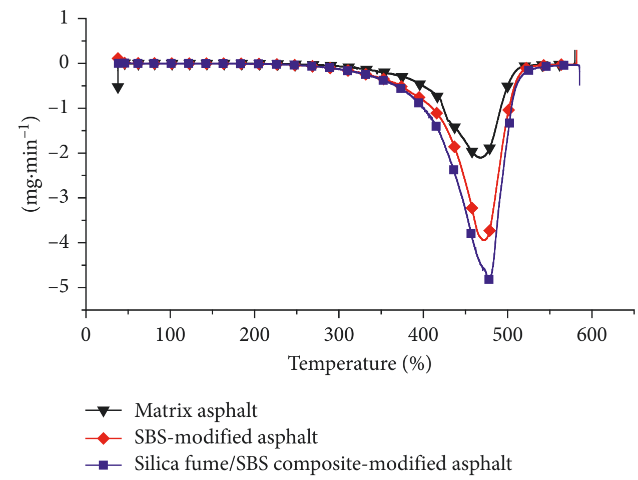

Figure 15: DTG curve.

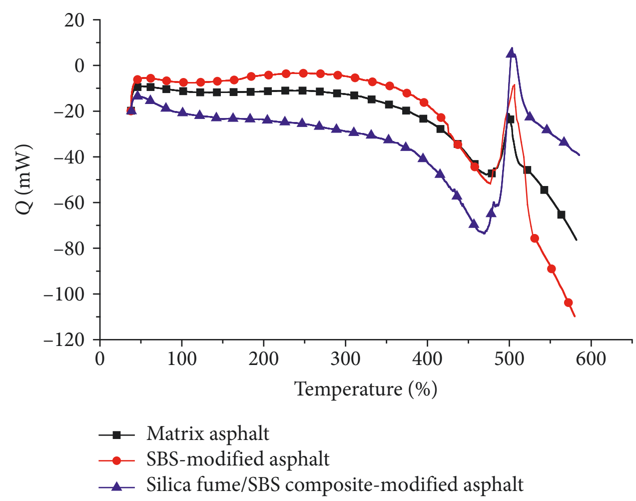

Figure 16: DSC curve.

temperature was within the range of (35°C, 220°C). When the temperature increased to 220°C, the thermal weight loss rate of matrix asphalt began to decrease, which indicated that the matrix asphalt began to decompose at 220°C. When the temperature was above 350°C, the TGA curve decreased rapidly and the mass residual rate of the sample decreased rapidly until it was almost lost. At the end of the test, the mass residual rate was approximately 12%. In the same way, the SBS-modified asphalt was decomposed at 235°C, and the final mass residual rate of the sample was approximately 16%. The silica fume/SBS composite-modified asphalt was decomposed at about 255°C, and the final mass residual rate of the sample was approximately 20%.

By analyzing the DTG curve, it was found that the matrix asphalt had a peak at 468°C, indicating that the decomposition rate of the asphalt was the fastest at 468°C. When the weight loss rate of SBS-modified asphalt reached the maximum value, the temperature was 474°C, and the temperature of the weight loss rate of the silica fume/SBS composite-modified asphalt reached the maximum value at 477°C.

As we could see from the DSC curve, the slope of the curve was larger when the temperature of the matrix asphalt was 35°C and 50°C. Then, the curve began to slow down when the temperature exceeded 50°C. It was shown that with the increase of temperature, the heat absorption of asphalt was larger before the softening point was reached, and the absorption heat was reduced after reaching the softening point. When the temperature reached 350°C, the DSC curve dropped rapidly and the heat absorption increased obviously. The curve reached the peak of an endothermic peak at 468°C and the area of the absorption peak was 2681 mJ. When the temperature reached 475°C, the curve of SBS-modified asphalt reached the peak of heat absorption and the area of the absorption peak was 3189 mJ. When the temperature reached 473°C, the curve of silica fume/SBS composite-modified asphalt reached the peak of heat absorption and the area of the absorption peak was 6014 mJ. Compared with the matrix asphalt and SBS-modified asphalt, the trend of the DSC curve of silica fume/SBS composite-modified asphalt is relatively gentle in the early stage. The whole endothermic peak is relatively backward. The temperature of the heat absorption peak is larger, and the acreage of the endothermic peak is larger. On the contrary, it indicated that the addition of silica fume improved the high-temperature performance of asphalt.

To sum up, the high-temperature performance of composite-modified asphalt is better than that of SBS-modified asphalt. Therefore, the addition of silica fume and SBS has a positive effect on the high-temperature performance of asphalt. Because the thermal conductivity of silica fume is very low. Adding this material to the asphalt will definitely reduce the thermal conductivity of composite-modified asphalt, improve its thermal resistance, and make it stable at high temperatures. Moreover, being a porous filler, silica fume has a lot of excellent characteristics such as small particle size of microvoids, large dead volume, strong adsorption capacity, low packing density, high filling amount, and large specific surface area. Silica fume is homogeneously dispersed in asphalt after physical mixing, and a homogeneous suspension system is formed. Then, the effective flow volume of the system would be reduced. At the same time, the large specific surface area and absorbability give it large surface energy; therefore, it can selectively adsorb light components in asphalt. The bonding strength of the adsorption layer of the boundary is enhanced, which greatly reduces the flow characteristics of modified asphalt and increases the rheological resistance. Therefore, the composite-modified asphalt shows better high-temperature performance.

5. Conclusions

In this paper, a series of tests were performed on matrix asphalt, SBS-modified asphalt, and silica fume/SBS composite-modified asphalt to study their properties. From the cone penetration test and DSR, it was observed that the cone penetration and the phase angle (δ) decreased with increase in silica fume content. However, the softening point, Brookfield viscosity, complex shear modulus (G^*), and the rutting factor ($G^*/\sin\delta$) increased, indicating that the addition of silica fume could significantly improve the high-temperature performance of the asphalt mortar. The creep stiffness (S) decreased initially and then increased, whereas the creep rate (m) increased initially and then decreased. Therefore, the recommended percentage of silica fume was fixed at 7%. An increase in the ratio of filler asphalt increased the high-temperature performance of the asphalt mortar greatly; however, the anticracking performance of the asphalt mortar deteriorated at low temperature. The addition of silica fume and SBS could improve the initial decomposition temperature, residual rate of thermogravimetry, the temperature in which the weight loss rate reached the maximum, and the area of the absorption peak. Therefore, the high-temperature stability was enhanced.

Data Availability

All tests were carried out in the standard laboratory according to the methods listed in the paper. The data were reliable.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Acknowledgments

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