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Research Article

Study on the Properties of Waste Oil-Activated Crumb Rubber-Modified Asphalt Based on Molecular Dynamics Simulation and Rheology

Qi Liu ,[1] Jinzhou Liu ,[1] Bin Yu ,[1] Jiupeng Zhang ,[2] Yuchen Wang ,[1] and Chuanyu Xiao [1]

[1]*School of Transportation, Southeast University, Nanjing 211196, Jiangsu, China*
[2]*Highway School, Chang'an University, Xi'an 710064, Shaanxi, China*

Correspondence should be addressed to Bin Yu; yb@seu.edu.cn

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To investigate the effect of activated crumb rubber content on the molecular interactions and the properties of crumb rubber-modified asphalt (CRMA) with waste oil, six models of asphalt with various rubber dosages were developed using Materials Studio software, and the molecular dynamics performance of the system was further examined. Then, the fatigue and high- and low-temperature performances of the CRMA binders were characterized by dynamic mechanical experiments in the laboratory. The mean square displacement and diffusion coefficient were used to quantify the migration of molecules. The aggregation state of the components was evaluated using a radial distribution function. The bulk modulus of the CRMA models was calculated to study the mechanical properties. Dynamic shear and bending beam rheometer tests were implemented to evaluate the road performances of the CRMA binders. The results show that increasing the amount of powder could improve the mechanical properties of the asphalt, that is, the modulus of 70% of the asphalt was improved by 57.5%. The rubber and waste oil were evenly dispersed in the system, and the distribution of asphalt components was in accordance with the colloid theory. The temperature-sensitive properties of the rubber led to the improvement of road properties of the CRMA binders with the increase of the admixture. Combined with the distribution of molecules in the asphalt model, the results of rheological indexes show that the waste oil could improve the rheology and stability of binders. This will provide theoretical support for upgrading the content of crumb rubber in CRMA binders.

1. Introduction

With the growing number of automobiles, a significant number of used rubber tires are produced each year [1]. The improper disposal of which is bound to cause environmental pollution and waste resources [2]. The use of crumb rubber-modified asphalt (CRMA) produced from waste tires for road construction has the potential to effectively alleviate the above issues [3]. However, the low superficial activity of untreated rubber powder and its low dispersion and compatibility with asphalt result in inferior stability and ease of construction of the CRMA [4–6]. In contrast, it has been reported that, after the activation modification treatment, the surface activity of the rubber powder increases, the internal structure changes, and the chemical bonds break and reorganize, which facilitate the degradation and depolymerization of the powder in asphalt and improve the compatibility with asphalt [7–10].

In the production of activated CRMA binders, choosing an appropriate method of activation of rubber powder is one of the key factors to improve the compatibility of asphalt with rubber particles. It has been reported that the storage stability and high- and low-temperature properties of CRMA prepared using desulfurized activated rubber powder were improved [11, 12]. It was found that the treatment of crumb rubber using the grafting method could improve the surface activity of powder and make it significantly more compatible with asphalt [13]. The method of activation by microwave treatment may induce the cleavage of the rubber powder, which reduces the macromolecular content in the CRMA and viscosity as a result. Further studies found that microwave activation enhances the surface activity of the powder and makes it more stable in combination with asphalt. In terms of performance, the complex shear modulus G^* of the CRMA increases and the phase angle δ decreases. The above indicates that the activation by microwave treatment can improve the compatibility of the powder with the asphalt, so that the rheological properties of the CRMA can be effectively improved, while the size effect of the mastic powder particles could be eliminated to some extent [14, 15]. Despite the benefits of using activated rubber powders to modify asphalt binders, susceptibility to dosing restrictions is the critical issue preventing the wider use of rubberized asphalt as a pavement material.

Several attempts have been made to pre-treat rubber particles with bio-oil or motor oil, manifesting great potential for improving the performance deteriorated after increased dosage of rubber powder. Yang et al. found that bio-oil may promote the swelling of the rubber powder in asphalt. Meanwhile, with the increase of shear temperature, the viscosity and high- and low-temperature properties of bio-oil-modified asphalt were enhanced and then weakened [16]. Furthermore, diesel and vegetable oils have also been used for the activation of rubber powder, and the performance of the corresponding crumb rubber-modified asphalt has been investigated [17]. This provides an effective means of enhancing the amount of rubber powder, but the stability of asphalt performance after the addition of waste oil is still seriously doubted. Hence, researchers have also tried to find ways to increase the amount of rubber powder from the preparation process of the CRMA [2].

To date, more studies focus on the method of increasing the amount of crumb rubber and the change of macroscopic properties of asphalt after increasing the amount, but they ignore the modification mechanism [18, 19]. Analyzing the interaction between asphalt components and rubber at the molecular scale will help the development of methods to increase the amount of rubber [20]. In recent years, with the development of molecular simulation methods and the improvement of computer arithmetic power, there are more and more studies on the molecular dynamics simulation of CRMA binders [21, 22]. For example, the diffusion and aggregation behaviors of rubber molecules in the asphalt were studied by calculating radial distribution functions (RDFs), mean square displacement (MSD), and diffusion coefficients. Solubility parameters and binding energies are calculated to characterize the compatibility between rubber molecules and asphalt components. In addition, the modulus and viscosity could be calculated to evaluate the performance of the asphalt. Guo et al. used molecular simulation to study the effect of different rubbers and different dosages on the compatibility of CRMA and found that polybutadiene rubber has the best compatibility with asphalt and the optimal amount of the rubber should be controlled in the range of 25%–30% [23]. The positive contribution of desulfurized rubber to the compatibility was also confirmed from the molecular scale by molecular dynamics simulations.

Table 1: Technical properties of SK-90# asphalt.

Technical indexes		Requirement	SK-90#
Penetration (25°C, 100 g, 5 s)/0.1 mm		80–100	89.9
Softening point (°C)		≤42	45.5
Ductility (5 cm/min, 10°C) (cm)		≤100	>100
RTFOT (163°C, 85 min)	Mass loss (%)	≥±0.8	0.07
	Penetration ratio (%)	≤54	70
	Ductility of 10°C (cm)	≤6	9.0

In summary, a CRMA binder with waste oil has great potential to enhance the amount of rubber powder, studying on the modification mechanism and performance of which is of extreme significance to promotion of application and material design. In this study, molecular dynamics simulations and experimental validation were combined to investigate the effect of activated rubber powder dosage on the component behavior and performance evolution rules of waste oil-pretreated CRMA binders. First, a CRMA model, including asphalt components and rubber and waste oil molecules, was developed using Materials Studio software. To obtain a reasonable CRMA model, the density, energy variation, and molecular order of the model were further checked. Then, molecular dynamics analysis was performed using Forcite module to quantitatively investigate the molecular interactions and the mechanical properties of the CRMA systems. Finally, to characterize the fatigue properties and high- and low-temperature of the CRMA binders with various content of rubber powder, the dynamic mechanical experiments were carried out using a dynamic shear rheometer (DSR) and a bending beam rheometer (BBR), and the experimental results are further compared and analyzed with the molecular simulation results.

2. Materials and Methods

2.1. Raw Materials. The base asphalt is SK-90#, which is produced in Korea; the technical index of SK-90# is shown in Table 1. The crumb rubber is 60-mesh waste radial tire powder produced by a company in Tianjin, as shown in Figure 1(a); the technical index of the crumb rubber is shown in Table 2. Waste oil is utilized engine oil purchased from 4S stores of automobile sales and service, and the solids in it are filtered, as shown in Figure 1(b).

2.2. Preparation of Activated Crumb Rubber and Modified Asphalts. The crumb rubber was activated for the preparation of CRMA binder. First, the rubber powder was mixed with waste oil (20%, by weight of crumb rubber) and put into a 100°C thermostat for half an hour for pre-swelling. Then, it was placed in a microwave oven with a microwave frequency of 2450 MHz for two minutes. Finally, the activated crumb rubber was prepared.

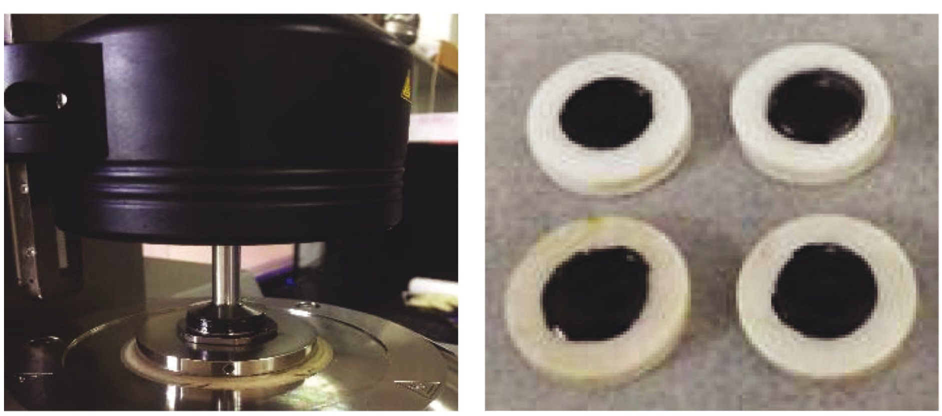
The wet process was employed to prepare the CRMA binders. The base asphalt is heated to flow in an oven at 160°C, and the shear is set to 500 r/min at slow speed. When

(a) (b)

Figure 1: Raw materials: (a) crumb rubber and (b) waste oil.

Table 2: Technical index of crumb rubber.

Technical indexes		American	Chinese	Test results
Physical indexes	Relative density (g/cm^3)	1.1–1.2	1.1–1.3	1.26
	Moisture (%)	≤0.75	<1	0.83
	Metal content (%)	<0.1	<0.03	0.02
	Fiber content (%)	—	<1	0.8
Chemical indexes	Acetone extract (%)	≤25	≤22	10.55
	Ash (%)	≤8	≤8	2.43
	Carbon black content (%)	20–40	≥28	34.78
	Rubber hydrocarbon content (%)	40–55	≥42	52.24
	Natural rubber content (%)	16–24	≥30	30.56



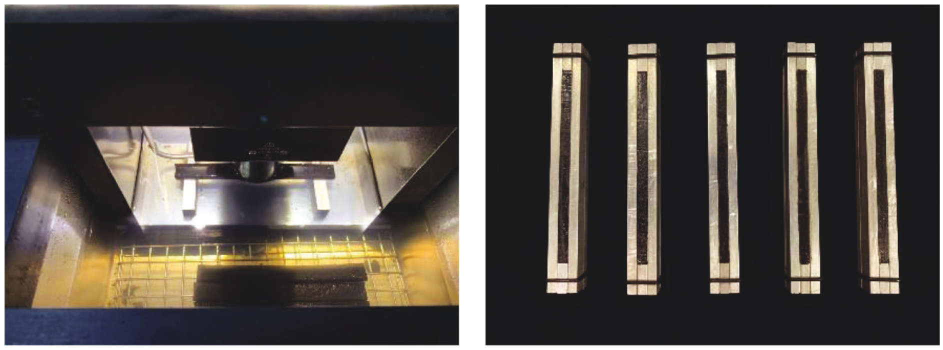
(a) (b)

Figure 2: Frequency sweep test: (a) DSR equipment and (b) asphalt specimens.

the temperature of asphalt reaches 180°C, the rubber powder is added (20%, 30%, 40%, 50%, 60%, and 70%, according to the mass of the base asphalt). After all the powder is added, the temperature rises to 230°C and the shear rate is set to 3000 r/min. During the shearing process, the temperature is measured with an infrared thermometer and the furnace power is adjusted to control the temperature between 230°C and 240°C for three hours.

2.3. Experimental Methods

2.3.1. Frequency Sweep Test. The frequency sweep test was performed to test the rheological parameters of CRMA binders including complex shear modulus G^*, phase angle δ, storage modulus G', loss modulus G'', and complex viscosity η' in the linear viscoelastic range. The rutting factor $G^*/\sin\delta$ and fatigue factor $G^*\cdot\sin\delta$ were further calculated for



(a) (b)

Figure 3: BBR test: (a) BBR loading module and (b) rectangular asphalt samples.

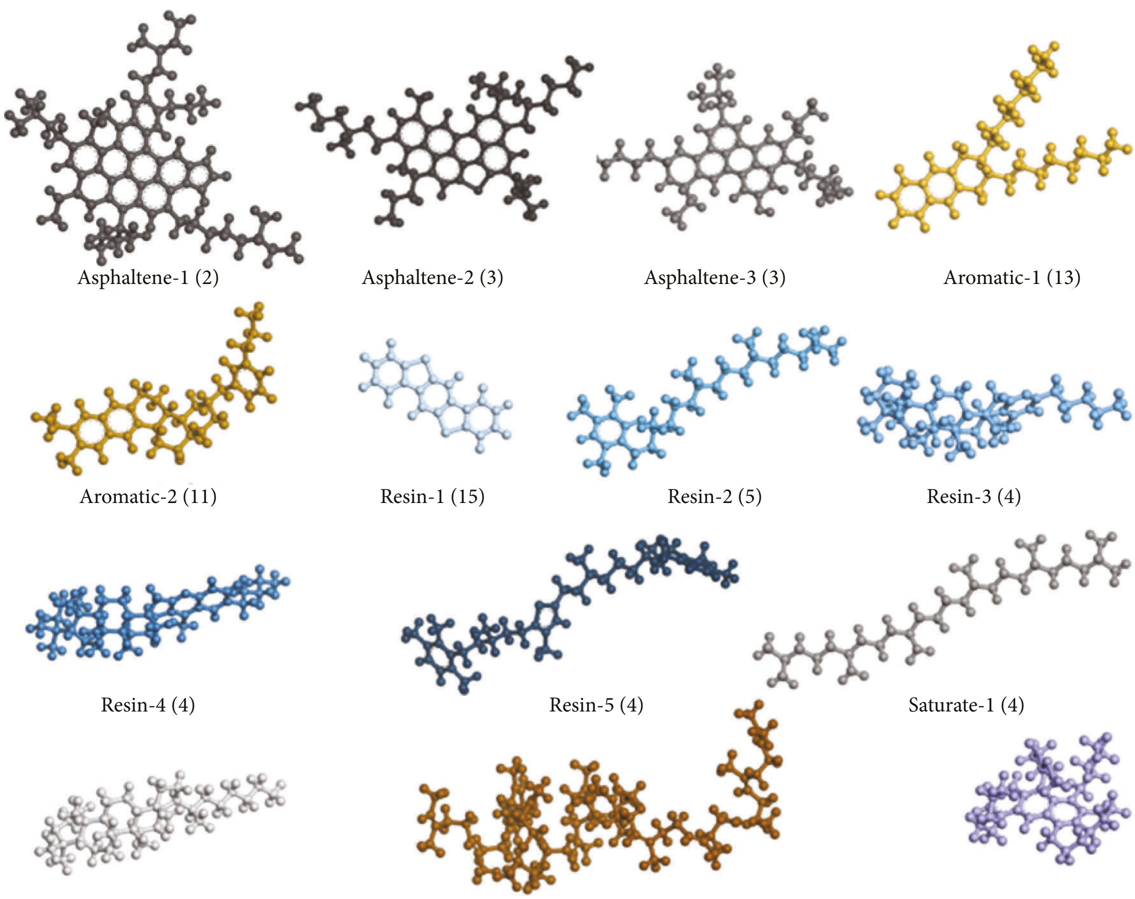


Figure 4: Molecular models of CRMA.

evaluating high-temperature and fatigue performance of binders, respectively. The frequency sweep test conditions are as follows: temperature 64°C, 25 mm parallel plate, frequency range 0.1–10 Hz, control strain 8%, and gap 1.5 mm. Figure 2 shows the dynamic shear rheometer and the asphalt specimens to be tested.

2.3.2. Multiple Stress Creep Recover (MSCR) Test. The MSCR test proposed by the AASHTO is exerted to evaluate the high-temperature performance of asphalt binders using the DSR at PG temperature. The specimens are RTFOT-aged asphalts. The recovered creep compliance R, nonrecoverable creep compliance Jnr, and stress sensitivity R_{diff} and Jnr_{diff} are applied as evaluation indexes.

2.3.3. Linear Amplitude Sweep (LAS) Test. To evaluate the fatigue performance of asphalt, LAS experiments were conducted on PAV-aged asphalt using DSR-controlled strain loading mode. Based on the obtained stress-strain

Table 3: Technical parameters of CRMA models.

Parameters		CRMA-20	CRMA-30	CRMA-40	CRMA-50	CRMA-60	CRMA-70
Number	Rubber	6	9	12	15	18	21
	Waste oil	3	4	5	6	8	9
Density (g/cm^3)		0.933	0.979	0.948	0.959	0.950	0.959

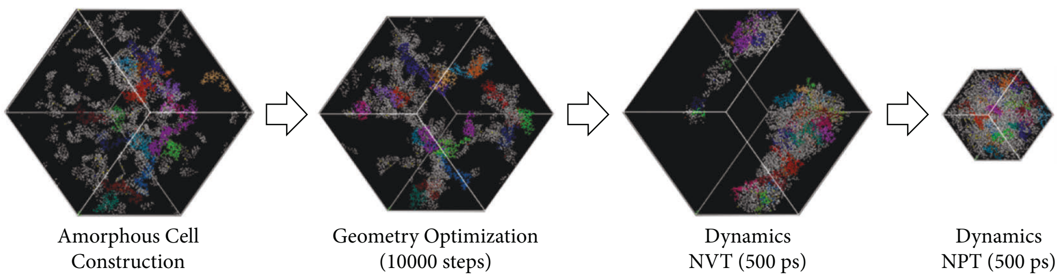


Figure 5: Simulation process of CRMA models.

data, a nonlinear fit was performed using the S-VECD model to obtain the damage characteristic curve of the material, which the fatigue life N_f was calculated.

2.3.4. Bending Beam Rheometer (BBR) Test. The BBR test could be employed to study the low-temperature performance of asphalt binder, and the PAV-aged asphalt binder is selected as the test specimen, as shown in Figure 3. The SHRP stipulates that the BBR test results should meet the following criteria: $S \leq 300$ MPa, $m \geq 0.3$, and the breaking strain of asphalt greater than or equal to 1%. If $S \geq 600$ MPa or $m \leq 0.3$, then the asphalt binder should not be used for low-temperature pavement design.

3. Molecular Dynamics Simulation

3.1. Molecular Modeling. Asphalt is a highly complex material consisting of various alkanes, cycloalkanes, and aromatic hydrocarbon molecules and comes mainly from the byproduct of crude oil distillation [24]. The molecules in asphalt are divided into four components—saturates, aromatics, resins, and asphaltenes (SARA)—based on similar physical and chemical properties according to ASTM D4124-09 [25]. Various molecular models of asphalt have been determined to implement molecular dynamics simulation studies of asphalt based on SARA [26, 27]. To enable molecular simulations that can further an understanding of asphalt's physical, rheological, and mechanical properties, Li and Greenfield proposed a model to represent the AAA-1, AAK-1, and AAM-1 asphalts of the Strategic Highway Research Program (SHRP) [28, 29]. The AAA-1 asphalt fraction was utilized in the simulation of the CRMA model. The molecular compositions of the model are shown in Figure 4, with the number of molecules in parentheses. The solubility parameters of SARA components are 16.6, 19.8, 19.1, and 20.8, respectively. Natural rubber molecules with a degree of polymerization of 16 were selected as the main components of crumb rubber [20]. The main components of used engine oil were similar lighter components of asphalt.

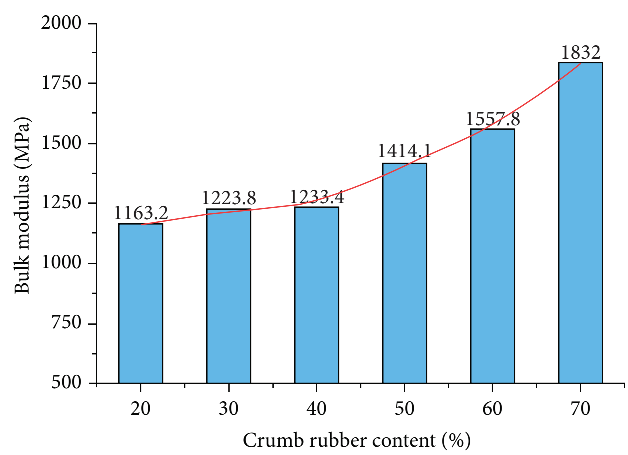


Figure 6: Bulk modulus of six CRMA models.

Hence, the aromatic molecules in petroleum are used as the main component [30, 31]. The number of rubber and waste oil molecules is shown in Table 3. The percentages of rubber molecules to asphalt molecular mass are 20.1%, 30.1%, 40.2%, 50.2%, 60.3%, and 70.3%, respectively. The waste oil molecules represent about 20% of the rubber molecules.

In this study, the molecular model of CRMA was developed using Materials Studio 2020 software. First, the amorphous cell module was utilized to introduce the component molecules into the cubic box. The initial density was set to 0.1 g/cm^3, and the system was geometrically optimized to minimize the system energy. Then, the optimized model was run for 500 ps NVT at 298.15 K. Finally, 500 ps of NPT simulation was performed to obtain the CRMA model, as shown in Figure 5. COMPASS II force field was selected during the simulation.

3.2. Dynamic Parameters

3.2.1. Mean Square Displacement. There is a correlation between the mean square displacement (MSD) and the diffusion coefficient, as in equations (1) and (2). The MSD

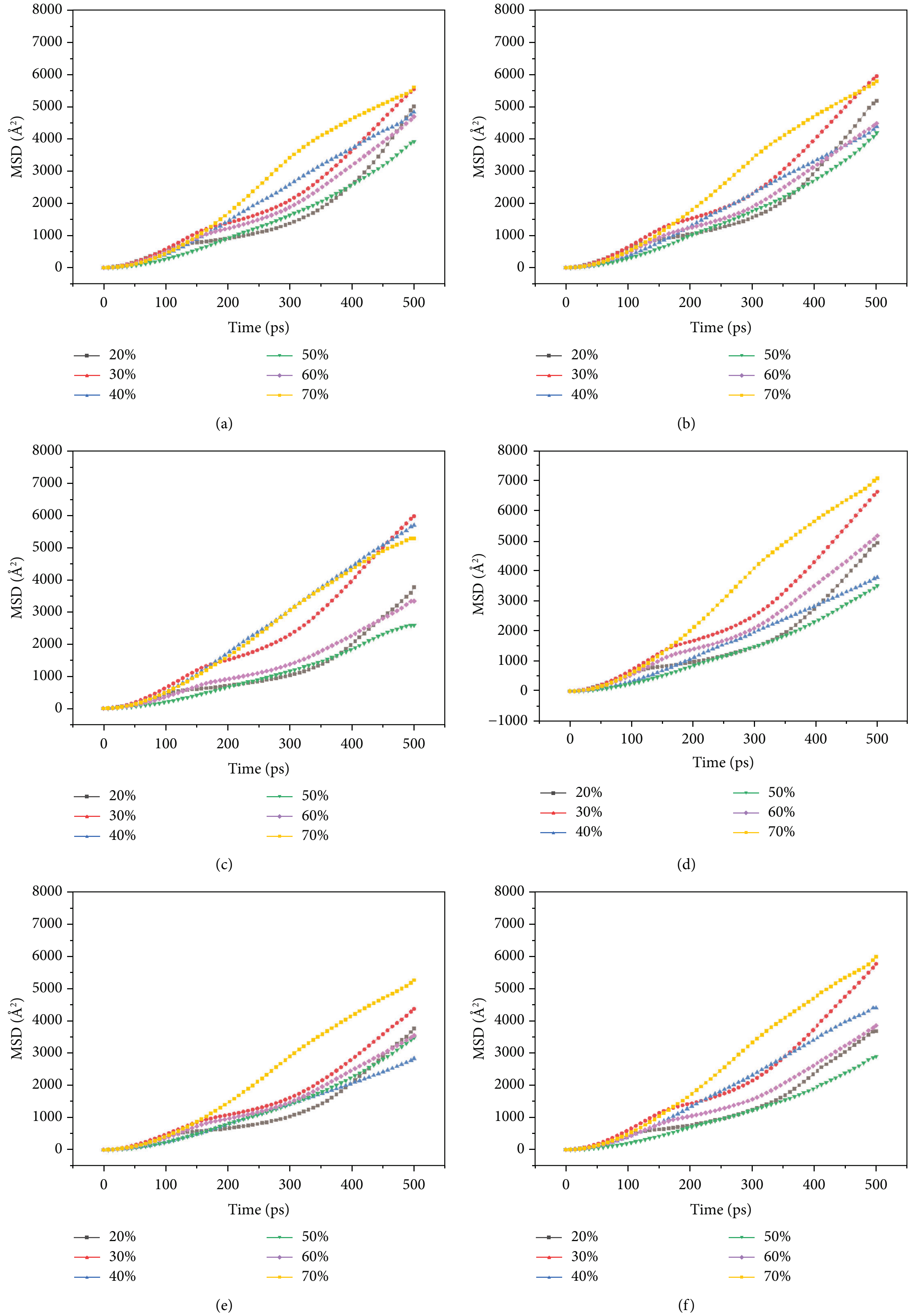


FIGURE 7: MSD curves of different components in CRMAs: (a) asphaltene; (b) aromatic; (c) saturate; (d) resin; (e) rubber; and (f) waste oil.

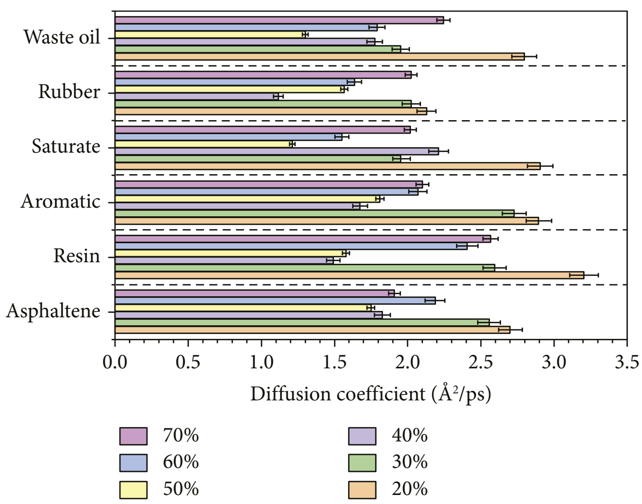


FIGURE 8: Diffusion coefficients of components in CRMAs.

and diffusion coefficient were used to investigate the effect of the number of rubber molecules on the diffusion behavior of component molecules.

$$MSD(t) = \langle [r_i(t) - r_i(0)]^2 \rangle, \quad (1)$$

$$D = a \cdot \lim_{t \longrightarrow \infty} \frac{1}{t} \cdot MSD(t), \quad (2)$$

where $r_i(t)$ is the position vector of particle i at time t and $r_i(0)$ is the position vector of atom i at the initial moment.

3.2.2. Radial Distribution Function. The radial distribution function (RDF) was applied to characterize the distribution of molecules in a characterized asphalt system over a certain distance, reflecting the adsorption behavior of the molecules present in the system. Therefore, the RDF could be used to evaluate the distribution of bitumen components and rubber and waste oil molecules. The RDF is calculated using the following equation:

$$g_{A-B}(\mathrm{r}) = \frac{dN/4\pi r^2 dr}{N/v}, \quad (3)$$

where dN is the number of B atoms at a distance r from A atoms; N is the total number of B atoms; and v is the volume of the periodic model.

3.2.3. Bulk Modulus. The bulk modulus K is a macroscopic property of a material that reflects its resistance to external homogeneous compression in the elastic regime. The K is calculated by

$$K = -V\left(\frac{\partial P}{\partial V}\right)_T = \frac{\langle V \rangle K_B T}{\left(\langle V^2 \rangle - \langle V \rangle^2\right)}, \quad (4)$$

where P denotes the system pressure; T denotes the system temperature; V denotes the system volume; and K_B is the Boltzmann constant.

4. Results and Discussion

4.1. Effect of Rubber Content on the Properties of CRMA

4.1.1. Modulus Variation. Figure 6 shows the bulk modulus of asphalts with different rubber contents at 298.15 K obtained from the Forcite module. It can be seen from Figure 6 that the modulus of CRMA models gradually rises with the increase of the number of rubber molecules. This indicates that increasing the amount of rubber could improve the strength of the asphalt [32]. The modulus growth rates of 30–70% asphalts compared to 20% are 5.2%, 6%, 21.6%, 33.9%, and 57.5%, respectively. The advantages of high-temperature strength and low-temperature toughness of rubber fully illustrate the great potential of the high-content CRMA in terms of mechanical properties.

4.1.2. Component Diffusion. To study the effect of crumb rubber admixture on the diffusion of different components, the MSD of each component centroid in CRMA was calculated, and the diffusion coefficients were further obtained, as shown in Figures 7 and 8.

It can be seen from Figure 7 that the MSD curves of each component have significant differences as the number of rubber molecules increases. But the MSD data at the initial and final stages of the curves are not stable. Thus, the middle part (200–400 ps) of the curves is analyzed. As shown in Figures 7(a)–7(d), the changes in the diffusion pattern of asphalt molecules are essentially the same as those which are influenced by the increase in the proportion of rubber. When the simulation time is 300 ps, the approximate ranking of the MSD values of SARA molecules is as follows: 70%, 40%, 30%, 60%, 50%, and 20%. This indicates that the effect of the number of rubber molecules on the diffusion of asphalt molecules is nonlinear. Factors such as waste oil molecules, the free volume of the system, and the total number of molecules may have an impact on the diffusion of asphalt molecules, and hence they need to be considered. It is noteworthy that, in Figure 7(e), the MSD results for the other four models are extraordinary close except for the 20% and 70% systems. This further indicates that the diffusion of rubber molecules is weakened by its own quantity, which is the significance of introducing waste oil in the rubber asphalt system. In Figure 7(f), the variation of the diffusion behavior of the waste oil molecules is similar to that of the SARA molecules. The diffusion coefficients were obtained by fitting the MSD curves. In Figure 8, the diffusion coefficients of the CRMA models show a decreasing and then increasing trend as the number of rubber molecules rises. Among them, the dropping trend of the diffusion coefficient is obvious from 20% to 40% content. The trend of change begins to shift at 40% and 50% of rubber molecules. The diffusion coefficients of the component molecules begin to increase again when the content increases to 60% and 70%. The above results indicate that the increase in the number of rubber molecules limits the diffusion of molecules to some extent. Moreover, the value of diffusion coefficient is influenced by the total number of molecules of the system, which may be

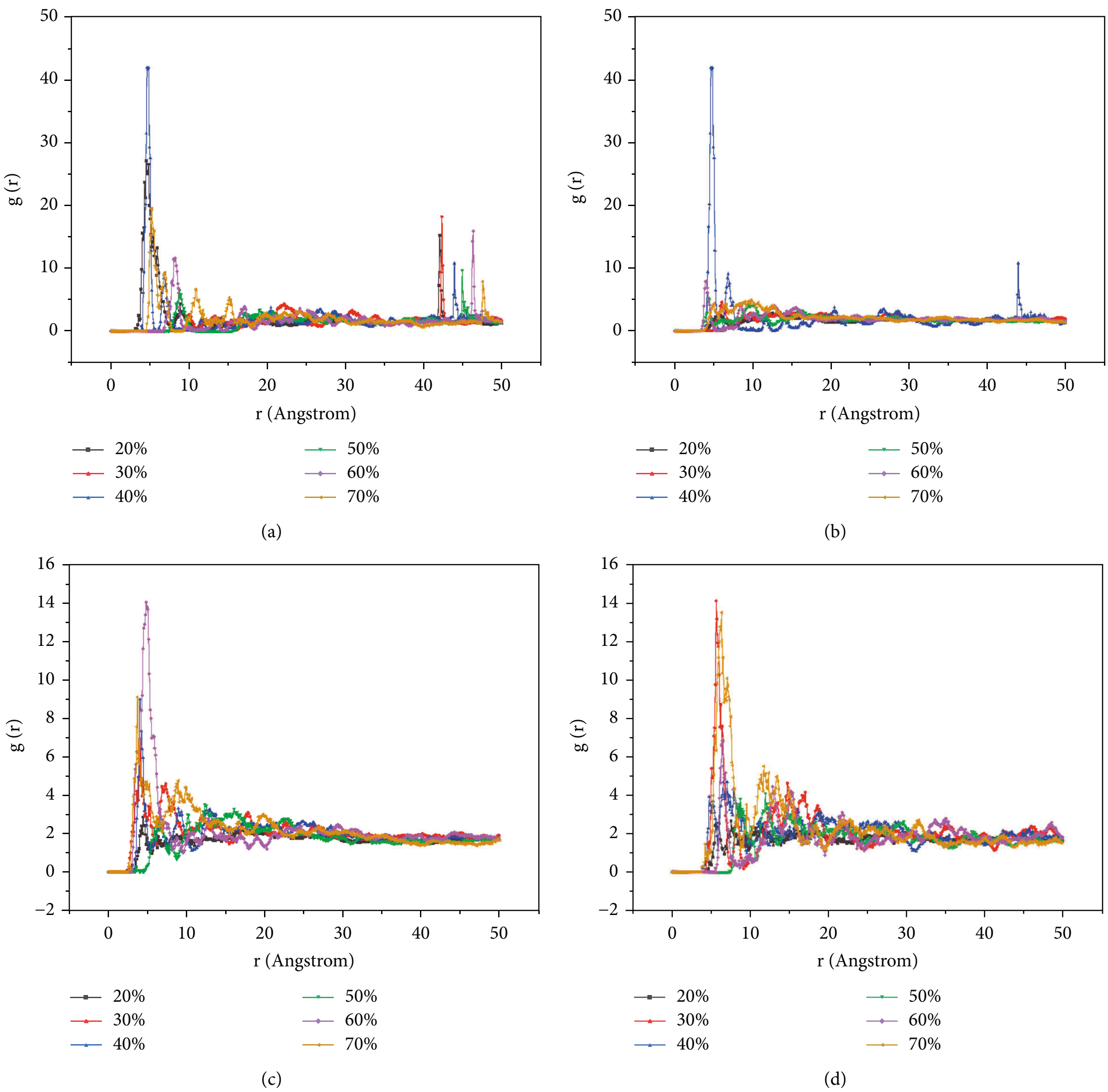


Figure 9: Continued.

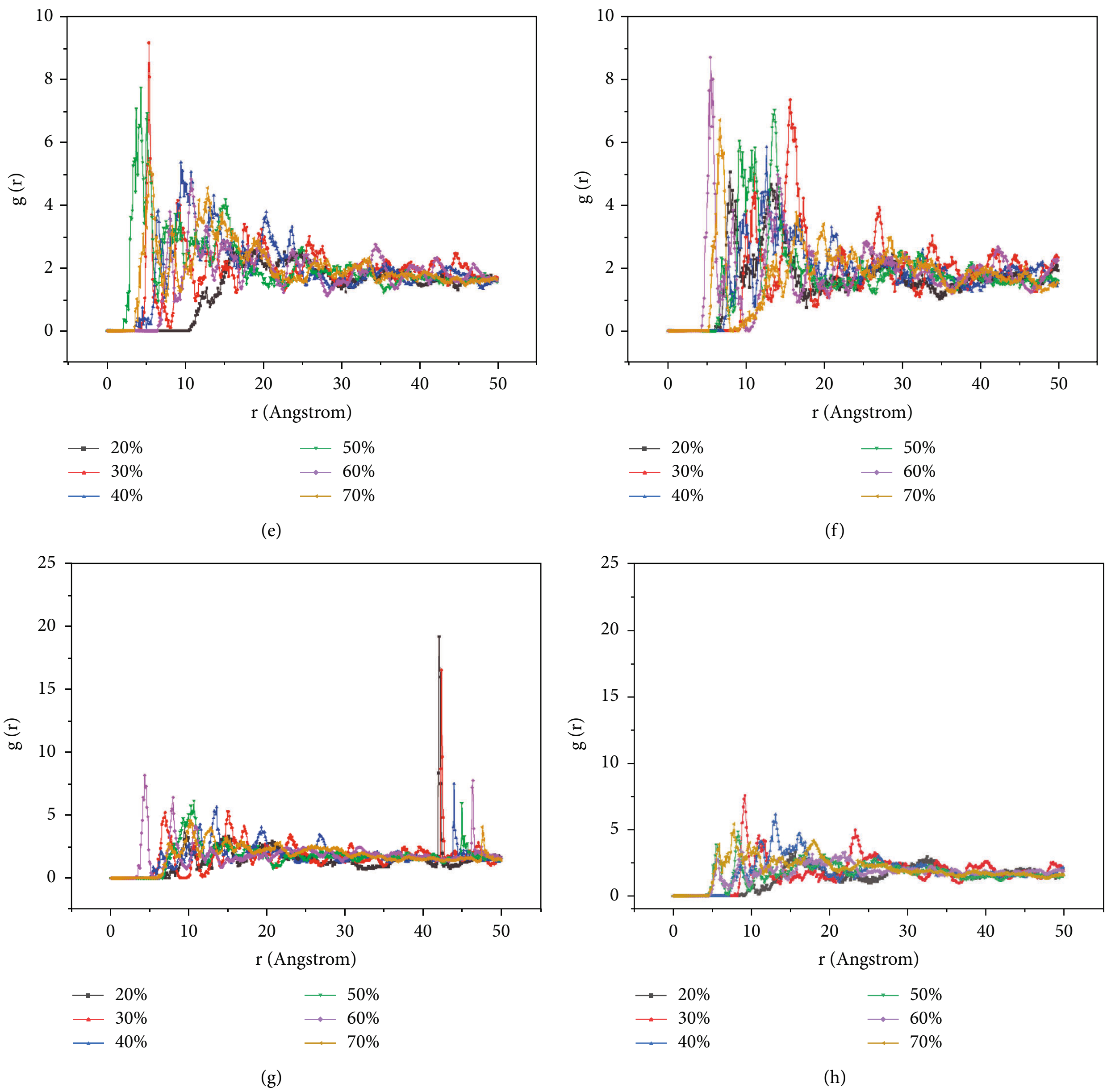


Figure 9: RDF curves of six components in CRMAs: (a) asphaltene-asphaltene; (b) asphaltene-aromatic; (c) asphaltene-resin; (d) asphaltene-saturate; (e) asphaltene-rubber; (f) asphaltene-waste oil; (g) rubber-rubber; (h) rubber-waste oil.

responsible for the increase of diffusion coefficient for 60% and 70% asphalt models [32].

4.1.3. Molecules Agglomeration. Figure 9 shows the RDF curves of asphalt models with different rubber powder dosages. In this simulation, the reference particles were set as asphaltenes in the colloidal structure, while the calculated particles were aromatics, saturates, resins, rubbers, and waste oils. It can be seen from Figure 9(a) that the strongest peak coordinate of asphaltene is (4.65, 41.95) for CRMA-40. When *r* is in the range of 0–20 Å, four peaks appear in total for CRMA-70, which are (5.15, 19.55), (6.95, 9.28), (10.85, 6.64), and (15.15, 5.32). This indicates that, as the rubber molecules increase, the aggregation of asphaltenes decreases and the probability of aggregation increases. This is mainly due to the increase in rubber molecules hindering the asphaltene movement. It is noteworthy that all six asphalts show peaks in the *r* range of 40–50. The coordinates of the peaks are 20% (42.05, 15.28), 30% (42.35, 18.23), 40% (43.95, 10.79), 50% (44.96, 9.70), 60% (46.35, 15.93), and 70% (47.55, 7.90). This also proves that asphaltene aggregation diminishes with the increase of rubber. In Figure 9(b), only the 40% asphalt model has more pronounced peaks. This indicates that the saturation fraction is more uniformly distributed in the system, which is in accordance with the colloid theory. From Figure 9(c), six asphalts show peaks in the *r* range of 0–10, indicating that colloids and asphaltenes

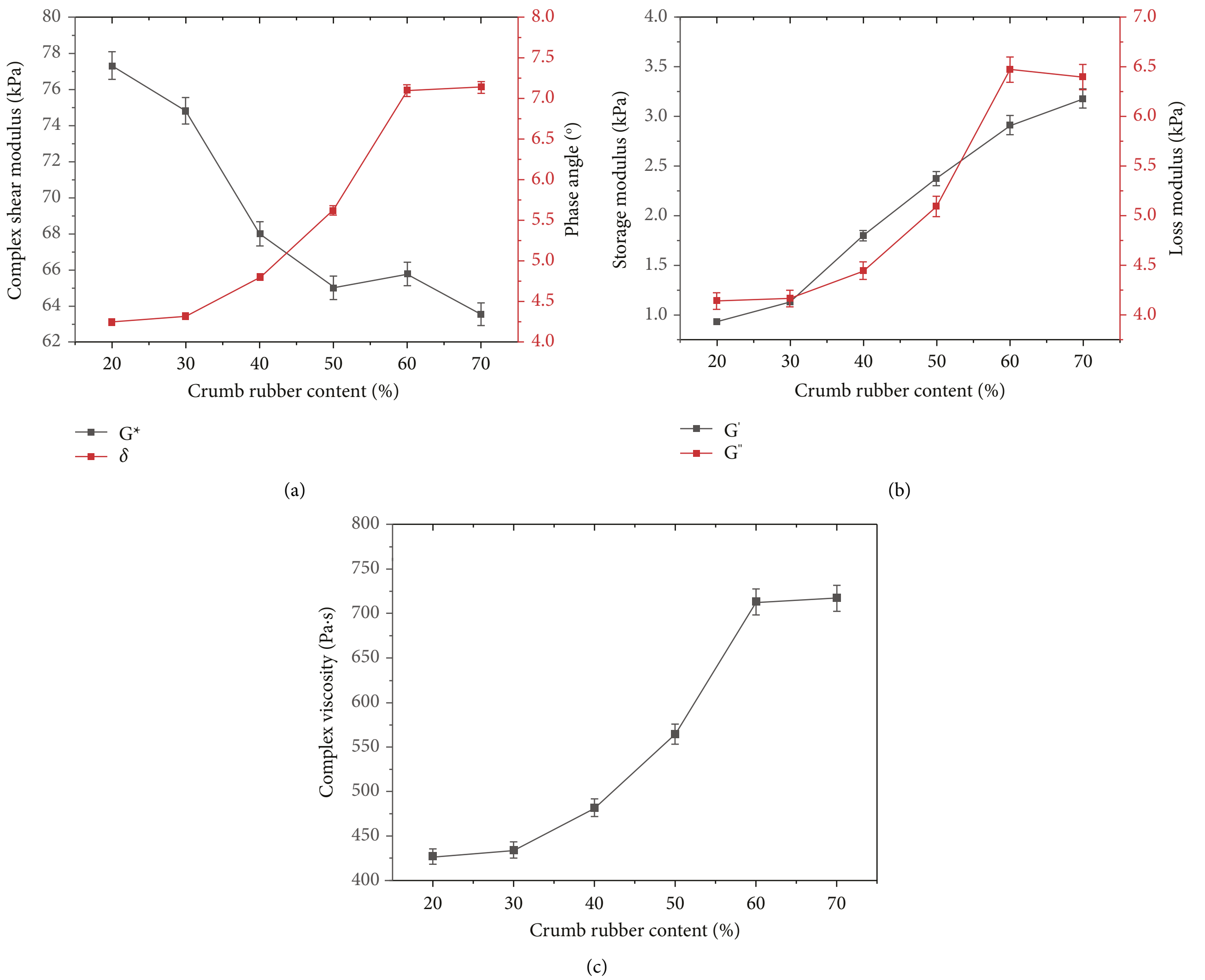


Figure 10: Rheological parameters of six CRMAs: (a) complex shear modulus and phase angle; (b) storage and loss moduli; and (c) complex viscosity.

Table 4: Test results of rutting factor (kPa).

Samples	Temperature (°C)	Rutting factor					
		20%	30%	40%	50%	60%	70%
Original	64	4.35	4.48	5.17	6.20	7.78	7.98
	76	1.14	1.29	1.86	2.50	3.09	3.40
RTFOT aged	76	2.66	2.91	2.93	5.79	6.42	6.99
	82	1.33	1.54	1.58	3.23	3.86	4.45

aggregate with each other. The saturated fraction is the same light component as the aromatic fraction, unlike the aromatic fraction, and it can be seen in Figure 9(d) that the peak appears around r of 5, indicating that the saturated fraction aggregates near asphaltenes. That is to say, asphaltene can adsorb saturated fraction. In both Figures 9(e) and 9(f), the RDF curves show more peaks. It indicates that rubber and waste oil are adsorbed around asphaltene. The number of peaks in the RDF curve in Figure 9(e) tends to rise with the increase in the number of rubber molecules. However, the

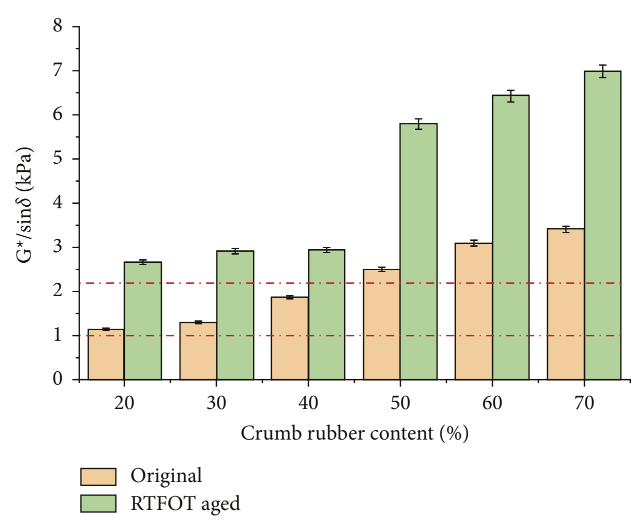


Figure 11: Effect of crumb rubber content on rutting factor (76°C).

aggregation degree of rubber and asphaltene does not rise with it. This reflects the role of waste oil in promoting rubber dispersion. From Figures 9(g) and 9(h), it can be seen that the curves have a smaller number of peaks and smaller peaks. This indicates that the rubber and waste oil could be uniformly distributed in the system. The aggregation of the rubber molecules themselves is weakened, according to Figures 9(e) and 9(g). Comparing Figures 9(f) and 9(h), the aggregation of waste oil molecules with asphaltene is stronger than that with rubber. The above results indicate that the waste oil molecules can supplement the lighter components required for the dispersion of the macromolecules in the system.

4.2. Rheology Indicators. To study the effect of crumb rubber content on the rheological properties of asphalt, the rheological parameters of six binders were tested using the DSR, as shown in Figure 10. It can be seen from Figure 10(a) that the G^* of the asphalts increases significantly with the increase of the rubber powder admixture. When the amount increases to 60%, the modulus growth slows down and the δ is in a decreasing trend. In Figure 10(b), both G' and G'' increase with the increase of crumb rubber; the difference is that the growth of storage modulus is larger than the loss modulus when the admixture reaches 60%. The trend of the composite viscosity in Figure 10(c) is similar to that of G^* and G'', and the growth of G^* is smaller at 20%, 30%, and 40% admixtures, indicating that there is a part of waste oil interacting with asphalt in addition to the part of waste oil acting on the rubber powder. The change of rheological index is larger when the content is between 50% and 60%, which indicates that, at this time, the rubber powder adsorbs a small number of light components from the asphalt in addition to the action with waste oil. When the dosage reaches 70%, the phase angle of the CRMA continues to change, while the complex shear modulus basically does not change, indicating that the reduction of light components in the asphalt leads to the properties of the CRMA binders by the rubber powder. This above results indicate that the amount of 60% has reached the maximum dosage of crumb rubber in the CRMA preparaton of this study.Exceeding this amount may have a negative effiect on the thermal stability of CRMA binders.

4.3. High-Temperature Performance

4.3.1. Rutting Factor. In the U.S. SHRP, the evaluation of the high-temperature performance of asphalt binder requires DSR tests on original and RTFOT-aged asphalt specimens, with rutting factor $G^*/\sin\delta$ as the evaluation index. The $G^*/\sin\delta$ should be greater than or equal to 1.0 kPa for the original asphalt and 2.2 kPa for the RTFOT-aged asphalt.

In this study, the rheological indexes of six types of asphalts were tested at different temperatures by the frequency sweep test and calculated to obtain the rutting factor, as shown in Table 4 and Figure 11. It can be seen from Figure 11 that the rutting factors of original and RTFO-aged asphalts increased with the increase of crumb rubber. This indicates that increasing the amount of rubber powder could increase the high-temperature rutting resistance of the CRMAs. The $G^*/\sin\delta$ of six original asphalts were greater than 1.0 kPa, and the $G^*/\sin\delta$ of six RTFOT-aged asphalts were greater than 2.2 kPa, which met the SHRP specification. This shows that the high-temperature rutting resistance of all six types of asphalts at 76°C could also meet the road requirements.

Table 5: PG grade test results of RTFOT-aged asphalt.

PG grade	20%	30%	40%	50%	60%	70%
Temperature (°C)	76	76	76	82	88	88

4.3.2. Nonrecoverable Creep Compliance. The MSCR test was implemented to further investigate the effect of powder admixture on the high-temperature performance of asphalt. The test temperature was the high-temperature PG grade in Table 5, the test was loaded in controlled stress mode with stresses of 0.1 kPa and 3.2 kPa, and the test procedure was carried out in strict accordance with AASHTO T350 requirements.

Nonrecoverable creep compliance Jnr is an important indicator to evaluate the asphalt binders, and the smaller the value of Jnr, the more elastic the components in the asphalt binder at a certain temperature. The better the high-temperature elasticity of the asphalts, the better the high-temperature resistance to deformation. From Figure 12, it can be seen that the value of Jnr gradually decreases when the amount of rubber powder admixture increases at two stress levels of 0.1 kPa and 3.2 kPa. This is consistent with the results of rutting factor. Compared with 3.2 kPa, the fit of Jnr results at stress of 0.1 kPa is better. The fitted curves for both stress levels showed a decreasing trend, where the Jnr at 0.1 kPa decreased gradually with increasing admixture, while the decrease at 3.2 kPa showed a slight change, indicating that the superiority of the high-temperature performance of the CRMA binders is also significantly affected by the level of loading action. In Figure 13, the value of Jnr_{diff} increases continuously with increasing admixture, indicating that the stress sensitivity of asphalt becomes larger with increasing admixture, and the elastic component of the asphalt binders increases and is more affected by the stress magnitude.

4.4. Fatigue Performance

4.4.1. Fatigue Factor. Generally, the smaller the fatigue factor $G^*\cdot\sin\delta$ at the same temperature, the less the energy loss of the bonding material under repeated loading, and the better its fatigue performance. The SHRP specification stipulates that the fatigue factor should be less than or equal to 5000 kPa. The fatigue factor results of six CRMAs at different temperatures are shown in Table 6 and Figure 14.

It can be seen from Table 6 that the temperature has a more significant effect on the fatigue performance of CRMAs. This is due to the fact that asphalt is a viscoelastic material, and the viscoelastic nature of asphalt will be affected by the nature of the powder after adding the rubber. At 28°C, the fatigue factor

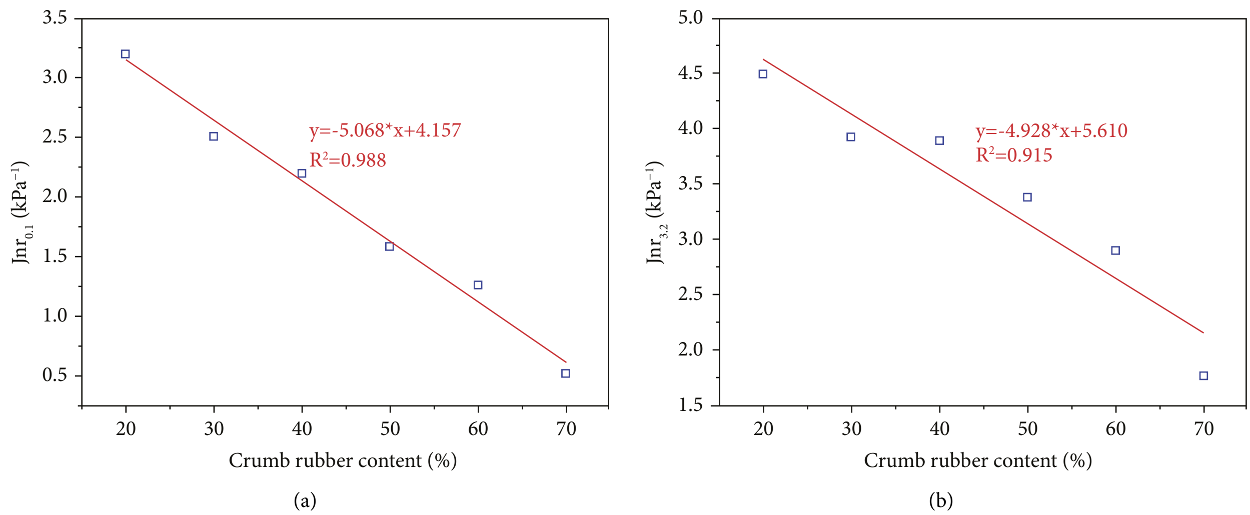


Figure 12: Effect of crumb rubber content on Jnr: (a) 0.1 kPa and (b) 3.2 kPa.

Table 6: Fatigue factor test results of original asphalts.

Test temperature (°C)	Fatigue factor					
	20%	30%	40%	50%	60%	70%
28	498.63	373.50	276.94	250.24	235.53	259.80
40	88.77	72.06	57.01	57.11	72.89	76.00

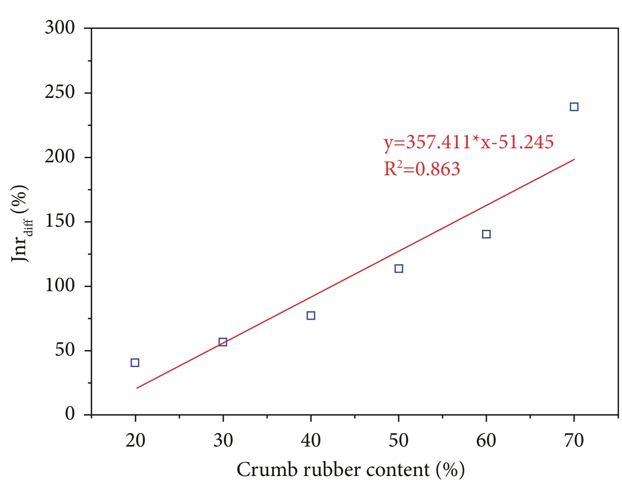


Figure 13: Effect of crumb rubber content on Jnr_{diff}.

$G^*{\cdot}\sin\delta$ gradually decreases with the increase of the amount and increases again when the doping amount increases to 70%. This indicates that the fatigue performance is influenced by the amount of admixture and the nature of the powder when the admixture is less than 60%. When the dose is greater than 60%, the fatigue performance is influenced by the content of light components in the modified asphalts, which indicates that the fatigue performance of the modified bitumen with 60% dose of powder is optimal at the test temperature of 28°C. When the temperature gradually increased, the fatigue performance of the modified asphalt is less and less influenced by the amount of the mixture.

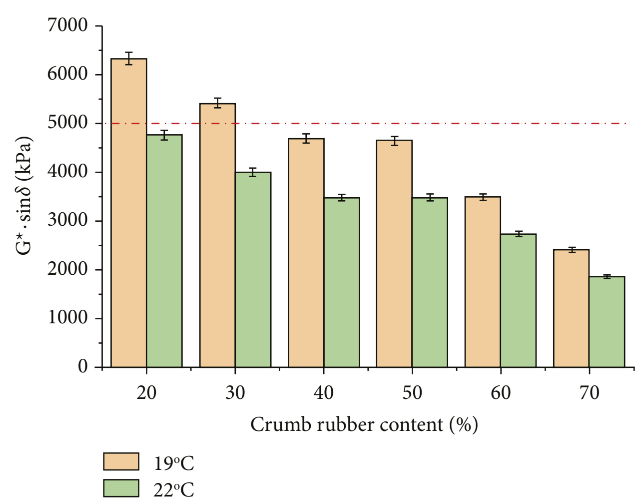


Figure 14: Fatigue factor of six CRMAs at different temperatures.

Table 7: PG grade test results of PAV-aged asphalt.

PG grade	20%	30%	40%	50%	60%	70%
Temperature (°C)	22	22	19	19	16	10

In Figure 14, the $G^*{\cdot}\sin\delta$ gradually decreases with the increase of powder admixture. This indicates that, after long-term aging, the fatigue performance of the asphalt is better when the amount of the admixture amount is larger. And after long-term aging, the durability of asphalt is greatly influenced by the crumb rubber because of the large reduction of light components in asphalt. However, the main component of the powder is rubber, rubber at lower temperatures could still maintain the elasticity and flexibility, resulting in the durability of CRMAs is improved. Six types of asphalts were able to meet the fatigue performance requirements of the AASHTO T315 standard at 22°C. In addition, four types of asphalts with 40%–70% admixture were able to meet the road requirements at 19°C.

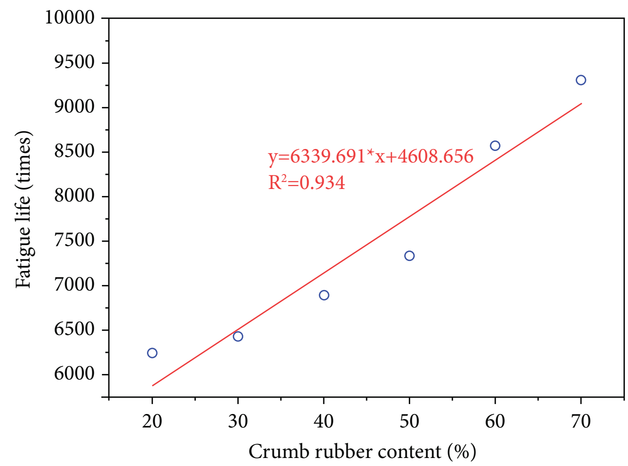


Figure 15: Effect of crumb rubber content on fatigue life.

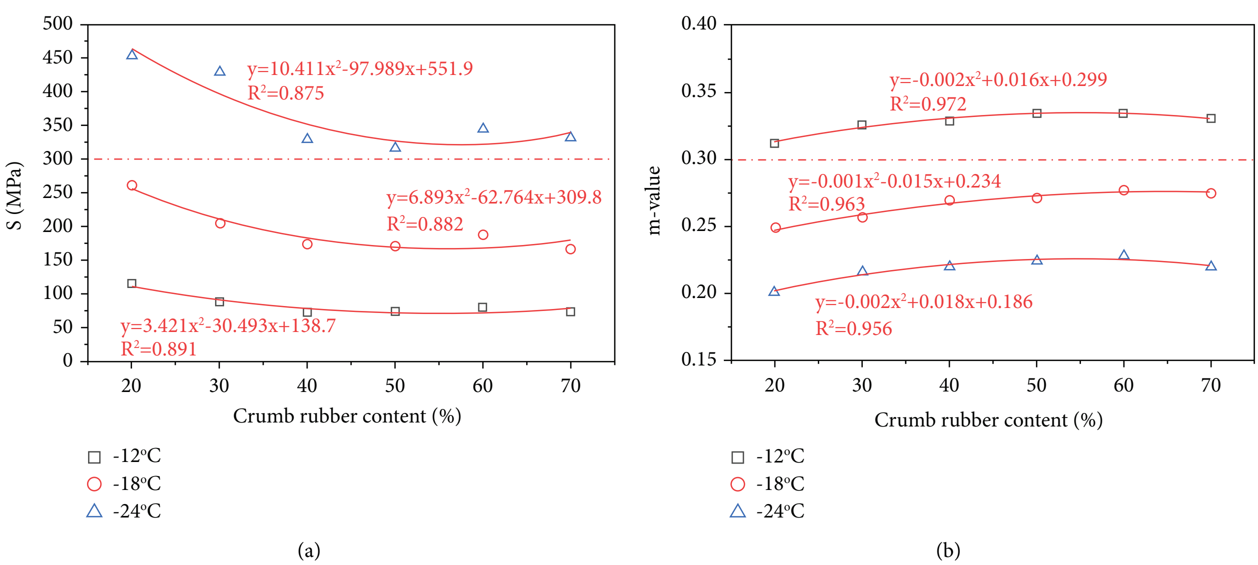


Figure 16: Results of BBR test: (a) stiffness modulus and (b) *m*-value.

4.4.2. Fatigue Life. To quantitatively describe the fatigue performance of asphalts with various crumb rubber dosages, LAS tests were conducted on six PAV-aged asphalt specimens in this study. The fatigue life of the six types of asphalts was calculated, and the variation of the asphalt fatigue life with the activated rubber dosage is shown in Figure 15. Table 7 shows the results of the PG grading of asphalt. It can be seen from Figure 15 that the fatigue life of asphalt increases significantly with the increase of crumb rubber. Combined with the results of PG grade in Table 6, the temperature through which the asphalt passes with increasing rubber powder is a decreasing trend. The results indicate that increasing the powder admixture could improve the fatigue resistance of asphalt. Meanwhile, this result is consistent with the results of fatigue factor.

4.5. Low-Temperature Performance. BBR tests were conducted on the six PAV-aged asphalts at −12°C, −18°C, and −24°C according to AASHTO T313 specifications. The test results are shown in Figure 16. It can be seen from Figure 16(a) that the stiffness was the largest at 20% of powder at three different test temperatures. As the temperature decreases, the stiffness of the six CRMAs gradually increases, and the growth rate decreases with the increase of the admixture. This indicates that the crumb rubber could improve the flexibility of asphalt at low temperature. The stiffness at both −12°C and −18°C can meet the requirement of $S \leq 300$ MPa. In Figure 16(b), with the temperature decreases, the creep rate m-value is gradually becoming smaller; at the same temperature conditions for the dose of 20% asphalt, the m-value is the smallest, and that for the dose of 60% asphalt, the *m*-value is the largest. At the dosage of 20%–60%, the *m*-value of asphalts rises with an increasing amount of powder, and at the amount of 70%, the *m*-value began to decline. This indicates that increasing the amount of powder could not improve the low-temperature performance of asphalts but rather begin to reduce. When the temperature is −12°C, the creep rate meets the requirements of $m \leq 0.3$, and the remaining two test temperatures of the six CRMAs do not meet the requirements.

Combined with the above analysis of stiffness and creep rate, it shows that the appropriate amount of powder can effectively improve the low-temperature performance of asphalt and the two indicators have a sound function with the amount. The best fitting effect was found at the temperature of −12°C. The low-temperature performance of the six asphalts at −12°C meets the requirements of $S \leq 300$ MPa and $m \geq 0.3$ at the same time. The ranking of the low-temperature performance of the six activated CRMAs according to the two indicators is as follows: 20% < 30% < 40% < 50% < 70% < 60%.

5. Conclusion

In this study, six asphalt models with different activated rubber dosages were developed and six CRMA binders were prepared accordingly. The effect of the amount of powder on the asphalt properties was investigated from both molecular simulations and laboratory experiments. The interactions between asphalt components and rubber and waste oil molecules were quantified. Meanwhile, the effects of powder dosing on the rheological, high- and low-temperature, and fatigue properties of the asphalt were characterized. The main conclusions could be drawn as follows:

(1) Increasing the number of rubber molecules leads to an increase in the bulk modulus value, indicating that an increase in rubber content enhances the mechanical strength of the asphalt system. The maximum growth rate of modulus was 57.5%.

(2) The movement of component molecules in CRMA is greatly influenced by the number of rubber molecules. Rubber molecules will hinder the diffusion of asphalt component molecules. From the results of the radial distribution function, it can be seen that, under the action of the waste oil molecules, elevated rubber content asphalt reminder can still meet the colloid theory. Meanwhile, rubber and waste oil can be uniformly dispersed in asphalt.

(3) From the results of rutting factor and non-recoverable creep flexibility, it can be seen that the high-temperature performance of the six asphalts could meet the requirements of road construction. With the increase in the amount of rubber powder, the high-temperature performance of asphalt binders gradually becomes better. The high-temperature deformation resistance of rubber itself is better than that of asphalt, so the amount of rubber should be increased as much as possible under the premise of ensuring other properties of crumb rubber-modified asphalts.

(4) The change law of fatigue factor of the original and PAV-aged asphalts is affected by temperature; the fatigue performance of asphalt with 20% and 30% of rubber powder at 19°C does not meet the requirements, while the asphalt shows elasticity at lower temperatures, indicating that the fatigue performance of asphalt is determined by the nature of the powder molecules, and the greater the amount of crumb rubber, the better the fatigue performance.

(5) The experimental results of the integrated creep stiffness and creep rate m-values show that the low-temperature performance of six types of asphalts at −12°C could meet the requirements of use and the low-temperature performance of asphalt with 60% rubber powder is the best.

Data Availability

The data used to support the findings of this study can be obtained from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest or personal relationships that could have appeared to influence the work reported in this paper.

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