

Research Article

Micro/Nanoscale Study on the Effect of Aging on the Performance of Crumb Rubber Modified Asphalt

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In order to explore the variation of crumb rubber modified asphalt (CR) before and after aging at the micro- and nanoscales, the molecular dynamics simulation and atomic force microscopy (AFM) asphalt microstructure scanning were carried out on CR before and after aging. The molecular dynamics energy, radial distribution function (RDF), surface microstructure image, roughness, and other test results of the CR before and after aging were compared and analyzed. The results show that the molecular dynamic energy of the CR after aging increases; the asphaltene-asphaltene RDF is decreased after aging; and the AFM surface microstructure images of asphalt before and after aging do not change significantly, but quantitative analysis by roughness theory shows that aging makes the microscopic surface of the CR more uniform and gentle. By analyzing the changes of asphalt properties before and after aging at two scales, it can be found that there is a certain relationship between the properties obtained at different scales. The reasons and mechanisms for the influence of microstructure on aging are obtained by analyzing this relationship.

1. Introduction

There is a certain relationship between the properties of the same material observed at different scales. For asphalt materials, exploring the relationship between the properties at different scales can further obtain the mechanism of the formation and change of macroperformance, which can provide a theoretical basis and feasible ways for further improving the macroperformance of asphalt. The molecular dynamics simulation method is introduced into the field of asphalt research to provide a new scale and method for the research of asphalt materials.

Tang et al. established a three-component model of asphalt through molecular simulation to study the aggregation state of asphalt molecules, but a simple three-component model is difficult to establish a relationship with the macroperformance, which can only be used as a simple analysis [1]. Yue and Ding et al. calculated the interaction between asphalt mixture and rejuvenator by molecular dynamics. The results show that the effect of rejuvenator on

long-term aging asphalt is obvious, and the effect on short-term aging asphalt is limited [2, 3]. Xu and Wang et al. applied molecular dynamics simulation to study the effect of thermal oxygen aging on asphalt and the water damage of asphalt mixture. It is found that aging would reduce the nanoaggregation behavior of asphaltene molecules, reduce the migration rate, and reduce the asphalt and aggregate interface bonding effect [4, 5]. Pan and Tarefder studied the effects of asphalt chemical composition and molecular structure on macroscopic rheological properties and mechanical properties after aging by molecular dynamics. The oxidation functional groups' viscosity of asphalt increases after aging, which leads to the increase of asphalt viscosity, indicating that the hardness of asphalt increases [6]. Yao et al. studied the effect of asphalt aging on the water damage resistance of asphalt mixture by molecular dynamics simulation. The results show that the aging component in asphalt helps to reduce the damage of water to asphalt mixture [7]. Xu et al. established a molecular interface model of asphalt samples and AFM siliceous needle tips. The adhesion

of asphalt at different aging degrees was obtained by AFM test and molecular dynamics simulation. It is found that the adhesion results obtained by the two methods are consistent [8]. Yi used molecular dynamics simulation to study the effect of asphalt aging on the adhesion work of asphalt-aggregate interface. It is believed that aging weakened the aggregation effect of asphalt. When moisture enters the interface between asphalt and aggregate, the interfacial adhesion work decreases, and the decline of aging asphalt is more obvious than that of matrix asphalt [9]. Yongjie Ding et al. studied the changes of functional groups and molecular weight of asphalt before and after aging by liquid chromatography. It is found that the aging effect has little effect on the aliphatic chain, but the aging effect increases the molecular weight of asphalt [10]. Fardin studied the glass transition temperature of SBR modified asphalt by molecular dynamics. It is found that the addition of SBR increases the aggregation of asphaltene molecules and reduces the migration of other component molecules. The macroscopic performance is that the addition of SBR improves the glass transition temperature of asphalt. At the same time, it is observed that the selected molecules follow the relationship between melt viscosity and temperature (VFT) as the behavior function of temperature [11]. Amit Bhasin studied the relationship between the chain length, chain branching, and self-diffusion of asphalt molecules through molecular simulation, so as to understand the nanoscale mechanism of molecular diffusion and self-healing ability [12]. Wang Lan et al. carried out molecular dynamics research on the compatibility between SBR molecules and asphalt, found that the best compatibility state was achieved between asphalt molecules and SBR molecules at 160°C, and studied the compatibility between asphalt molecules and SBR molecules before and after aging under the best compatibility state [13, 14].

For the relationship between multiscale performances, scholars at home and abroad use different test methods to study and analyze asphalt materials. Loeber et al. first discovered the asphaltene molecular gel network using AFM and named it “bee structure” [15]. Alireza Samieadel et al. carried out a multiscale analysis on the behavior of paraffin in asphalt as a warm mixing agent and studied the influence of paraffin on the thermodynamic properties of asphalt by DSC. It is found that the glass transition temperature and viscosity of asphalt decrease with the increase of paraffin concentration. The interaction between asphaltene and paraffin is studied by molecular dynamics simulation, and it is found that the increase of paraffin content leads to the decrease of viscosity. The average size of the asphaltene phase decreases but the number increases, and the diffusion coefficient of paraffin increases about 7 times [16]. Chen Jianhua carried out macroscale analysis on the thixotropic zone of the casting asphalt mixture, such as DSR and rutting test. The aging zone was analyzed by spectroscopy and performance of casting asphalt concrete by using infrared spectroscopy, AFM, and other microscale analysis methods. The thixotropy and serviceability of the casting asphalt concrete were systematically studied. After that, the research results were verified by a full-scale accelerated loading test,

and the correlation among micro-, meso-, macro-, and full-scale indexes was established [17]. Cao Peng and Gong Xiangbing carried out multiscale mechanical analysis and model study on the road surface. The results show that the simulation results of the multiscale unified model have a corroboration relationship with the simulation results of the mesoelement equivalent model, and the mechanism of the micro-meso-macro scale is preliminarily established [18, 19]. Wang Peng et al. explored the genesis of asphalt bee structure through molecular dynamics simulation, studied the contribution of each component to the formation of asphalt colloid, and analyzed the correlation between chemical composition and road performance [20].

Aging is accompanied by the full-life cycle of crumb rubber modified asphalt (CR), so it is necessary to study the aging of CR. Domestic and foreign scholars have used molecular dynamics simulation and cross-scale analysis methods to conduct a variety of research on asphalt materials, but there are few cross-scale studies on crumb rubber modified asphalt aging. Atomic force microscope quantitative nanomechanical model (AFM-QNM) is used to scan the microscale of CR before and after aging, and the change of molecular dynamics energy and radial distribution function of crumb rubber modified asphalt before and after aging are calculated by molecular dynamics simulation. The effects of aging on CR at two scales are analyzed to study the reasons and mechanism of performance change of crumb rubber modified asphalt before and after aging.

2. Test Material

Panjin 90# matrix asphalt and 60 mesh waste crumb rubber were used to prepare crumb rubber modified asphalt by the wet process. The crumb rubber content was 20%, and the aging crumb rubber modified asphalt was prepared by pressure aging test. Three main indexes were tested on the original crumb rubber modified asphalt (O-CR) and aging crumb rubber modified asphalt (A-CR). The results are shown in Table 1. The temperature sensitivity parameters were calculated according to formulas (1) and (2), and the calculation results are shown in Table 2.

$$\lg P = K + AT, \quad (1)$$

$$PI = \frac{20 - 500A}{1 + 50A}, \quad (2)$$

where P is penetration (0.1 mm), K is regression constant, A is temperature sensing coefficient, T is test temperature (°C), and PI is penetration index.

As the temperature coefficient A decreased, the penetration index PI increased, the asphalt temperature sensitivity decreased, and the high-temperature performance of the asphalt improved. The softening point and penetration index of A-CR increased, indicating that its temperature sensitivity was reduced and its high-temperature stability was improved to a certain extent. However, the decrease of ductility and penetration indicated that the main effect of aging was the deterioration of the low-temperature performance of asphalt.

TABLE 1: Three main indexes test of O-CR and A-CR.

Item	O-CR	A-CR
Penetration (5°C, 100 g, 5 s; 0.1 mm)	11.6	6.3
Penetration (15°C, 100 g, 5 s; 0.1 mm)	30.2	15.6
Penetration (25°C, 100 g, 5 s; 0.1 mm)	73.8	37.5
Ductility (5°C; cm)	16.5	12.3
Softening (°C)	57.3	63.4

TABLE 2: Temperature sensitivity parameters.

Parameters	A	K	PI	Correlation coefficient R
O-CR	0.0402	0.8681	1.91	0.9998
A-CR	0.0387	0.6078	2.585	0.9999

3. Analysis of Nanometer Energy of CR before and after Aging

In this paper, Panjin 90# matrix asphalt was used to prepare long-term aging asphalt (hereinafter referred to as aging asphalt) according to T0630-2011 pressure aging vessel accelerated asphalt aging test regulations in JTJG E20-2011 “Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering.” The four components of asphalt were analyzed according to the regulations of T0618-1993 asphalt chemical component test (four-component method) in JTJG E20—2011 “Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering.” During the thermal aging process of asphalt, due to the influence of high-temperature oxidation, the change of its components was mainly manifested as the conversion of the resins in the light components into asphaltenes, which resulted in the decrease of the plasticity and increase of the brittleness of the asphalt in the macroperformance, while the saturate and aromatic content of the asphalt had little change, which made little contribution to the change of the asphalt performance. It can be seen from Table 3 that the mass ratio of four components (m (asphaltene): m (saturate): m (aromatic): m (resin)) before aging was about 1.0 : 2.7 : 3.1 : 3.9, and the content of resin was the largest in matrix asphalt. Therefore, matrix asphalt showed obvious viscosity, high penetration degree, and low softening point. After aging, the mass ratio of four components (m (asphaltene): m (saturate): m (aromatic): m (resin)) of aging asphalt was about 1.0:1.0:1.0:1.1. The asphaltene content increased significantly while the resin content decreased, and the brittleness of asphaltene became the main factor affecting the macroproperties of asphalt.

Some scholars have conducted a large number of studies on the molecular structure of asphaltenes, resins, saturates, and aromatics in asphalt. Asphaltenes are a variety of compounds with aromatic ring groups and short straight chains [21]. Therefore, the highest content of alkane molecules in asphalt is $C_{22}H_{46}$, and $C_{22}H_{46}$ is close to the thermodynamic characteristic temperature of saturation fraction. Therefore, $C_{22}H_{46}$ was selected as the representative molecule of saturation fraction [22, 23]. The resin is a layered molecule composed of various condensed aromatics and

alicyclic alkene with various alkyl groups and heteroatoms [24]. The number of aromatic rings and straight chains of the aromatic component is similar to that of 1, 7-dimethyl naphthalene molecules. Therefore 1, 7-dimethyl naphthalene molecules were selected as the representative molecules of the aromatic component [25]. Asphaltenes, saturates, aromatics, and resins in asphalt were represented by known polymer molecules as shown in Figure 1. The matrix asphalt molecular models before and after aging were constructed according to the four-component ratio as shown in Figures 2 and 3.

From the asphalt molecular model before and after aging, the volume of the asphalt molecular group was larger, the content of the benzene ring was more, and the group was more compact as a whole after aging. The relative structure of the original asphalt molecules was looser, and the loose structure led to the weak interaction within the molecular group. When the temperature increased or decreased, the motion state of molecules was more susceptible to the change of external energy, so the original asphalt had higher temperature sensitivity. The closer the molecular group structure was, the higher the energy required for the molecular movement was, so the temperature sensitivity of aging asphalt decreased.

Styrene-butadiene rubber (SBR) is the main component of crumb rubber used in crumb rubber modified asphalt (CR). SBR can significantly improve the temperature sensitivity and viscoelasticity of asphalt, so it plays a major role in asphalt modification. The tire was treated with antiaging treatment during the production process. Therefore, the aging of crumb rubber was not considered in the study of the performance of CR before and after aging. The SBR model was selected to represent the rubber model in this paper. Firstly, random copolymer SBR was constructed. After the energy optimization of SBR monomer model, 50 monomer molecules were polymerized into SBR single-chain molecule. Considering that the main component of waste crumb rubber was SBR, and it had antiaging treatment in the tire production process. Therefore, when studying the properties of CR before and after aging, the aging of crumb rubber was not considered. The molecular model of SBR is shown in Figure 4. The 3D model of SBR was obtained by combining four single-chain molecules of SBR. Then, according to the mass ratio of 20%, the asphalt molecules and SBR molecules were fused to obtain the original crumb rubber modified asphalt molecular model shown in Figure 5. All kinds of molecules were optimized, and the optimization steps and parameters are shown in Table 4.

After optimization, the density of asphalt molecules was stable between 0.95 and approximately g/cm^3 , which was close to the true density of 90# matrix asphalt at $1.02 g/cm^3$. The density of SBR molecule was stable between 0.90 and $0.95 g/cm^3$, which was consistent with the real SBR density of $0.94 g/cm^3$.

The energy of the Forcite module was calculated for the optimized crumb rubber modified asphalt molecular group, the energy data of the whole process of calculation was processed, and the molecular dynamics energy chart of

TABLE 3: Original matrix asphalt and aging matrix asphalt four components ratio $\omega/\%$.

	Original matrix asphalt	Aging matrix asphalt
Asphaltene	9.4	24.6
Saturate	25.2	25
Aromatic	28.8	23.6
Resin	36.6	26.2
Total	100	99.4

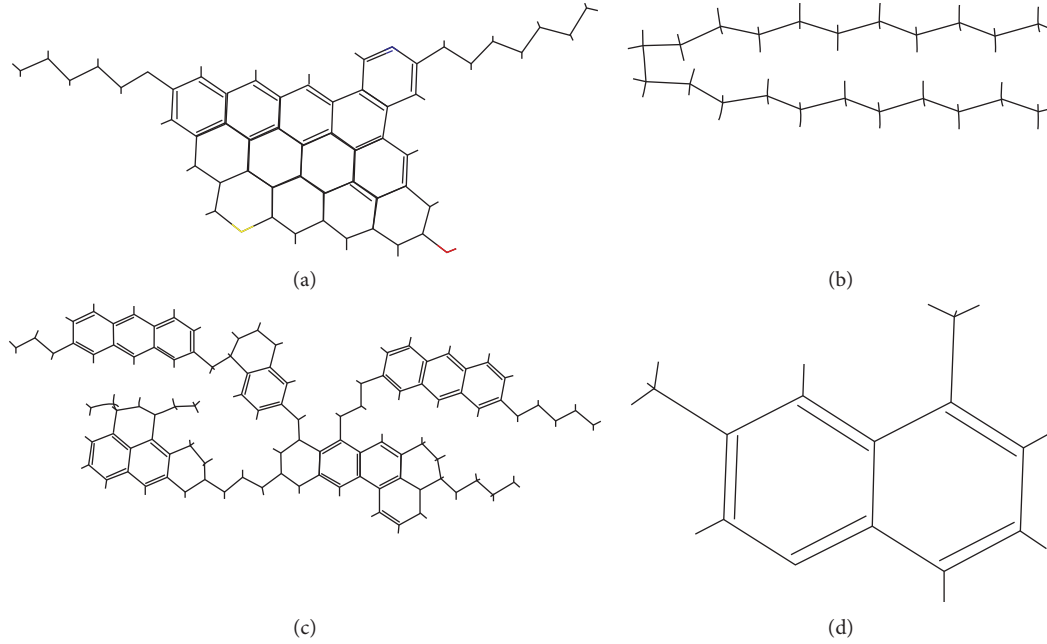


FIGURE 1: Molecular models of asphalt four components. (a) Asphaltene (As). (b) Saturated (S). (c) Resin (R). (d) Aromatic (Ar).

crumb rubber modified asphalt molecular group before and after aging was obtained as shown in Figure 6.

The molecular energy of crumb rubber modified asphalt increased after aging, which was due to the transformation of light small molecules into heavy macromolecules in crumb rubber modified asphalt, which increased the overall molecular weight and then increased the overall dynamic energy of asphalt by 17%.

4. Analysis of RDFs of CR before and after Aging

The radial distribution function $g(r)$ represented the ratio of the probability density of a molecule to the density of a random distribution probability at a distance r around a molecule. The definition of $g(r)$ is shown in the following formula:

$$g_{AB}(r) = \frac{1}{\rho_{AB} 4\pi r^2 \delta r} \frac{\sum_{i=1}^K \sum_{j=1}^{N_{AB}} \Delta N_{AB}(r \rightarrow r + \delta r)}{N_{AB} \times K}, \quad (3)$$

where N_{AB} is the number of A and B atoms in the system, K is the time step, δr is the distance interval width, ΔN_{AB} is the number of B atoms in the range of r to $r + \delta r$ from A atoms, and ρ_{AB} is the system density.

The asphaltene before and after aging was taken as the calculation center with a cut-off radius of 20 Å. The radial distribution functions of each component and crumb powder of CR before and after aging were calculated, respectively. The calculation results are shown in Figure 7.

It can be seen from Figures 7(a) and 7(c) that the aging asphaltene-SBR and asphaltene-resin RDFs are slightly higher than the original asphaltene-SBR and asphaltene-resin RDFs. It can be seen from Figure 7(b) that after aging, the RDF value of asphaltene-asphaltene is significantly lower than that of original asphaltene-asphaltene, indicating that aging weakens the stacking degree between asphaltenes, the distribution of asphaltene in the molecular model is more dispersed, and the aggregation degree is reduced. According to Figures 7(d) and 7(e), RDFs of asphaltene-saturate and asphaltene-aromatic before and after aging, asphaltene-saturate and asphaltene-aromatic basically coincide before and after aging, indicating that aging has no obvious effect on the aggregation of saturate and aromatic in the molecular model.

5. Analysis of CR Microstructure before and after Aging

AFM scanning was carried out to obtain the micrograph of CR before and after aging, as shown in Figure 8.

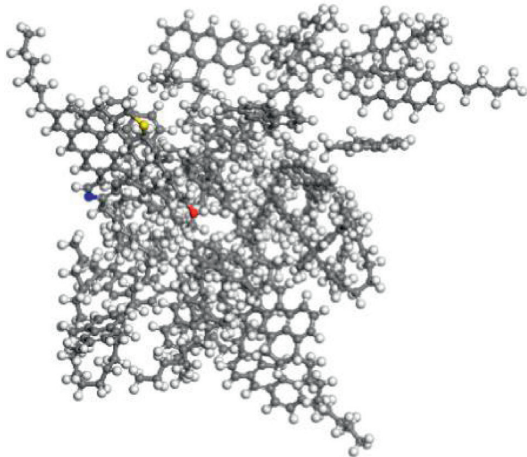


FIGURE 2: 3D model of the original matrix asphalt.

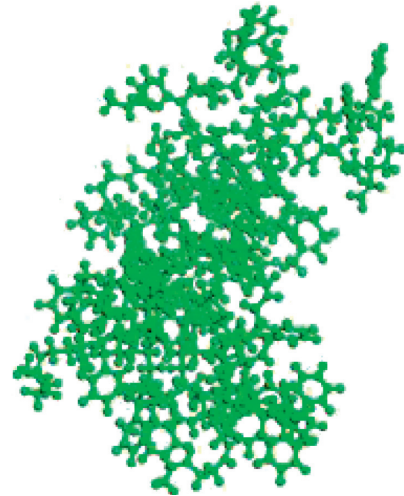


FIGURE 4: 3D model of SBR.

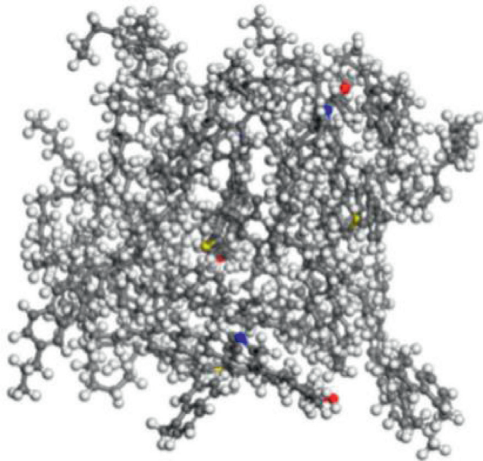


FIGURE 3: 3D model of aging matrix asphalt.

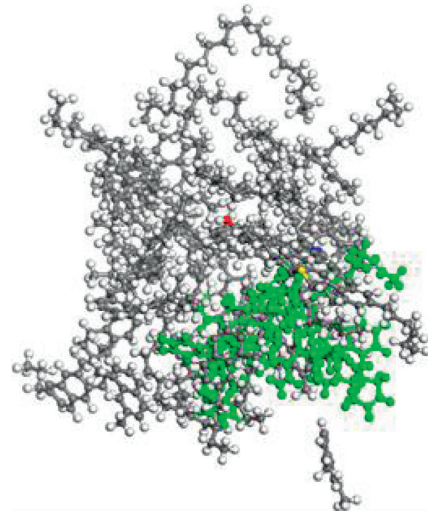


FIGURE 5: 3D model of original CR.

TABLE 4: Model optimization steps and parameters.

Steps	Number of steps or time	Forcefield or ensemble
Geometry optimization	10^5	COMPASSII
Anneal	$5 \cdot 10^4$	NPT
Dynamics	200 ps	NPT-298K
	100 ps	NVT-1500K

From the AFM scan image of crumb rubber modified asphalt before and after aging, it can be seen that, unlike matrix asphalt, crumb rubber modified asphalt has no obvious bee structure before and after aging. The addition of crumb rubber caused the microstructure of asphalt to change. Through the observation of three-dimensional images, it is found that although there is no obvious bee structure, the CR still has obvious peak and valley structures. Unlike matrix asphalt, these peaks and valleys were no longer clearly aggregated but were dispersed uniformly and

more finely. These dispersed peaks and valleys formed the black and white speckle structure of crumb rubber modified asphalt in two dimensions. These finely dispersed peaks and valleys formed the black and white speckle structure of crumb rubber modified asphalt in 2D. By analyzing the influence of crumb rubber on the microstructure of asphalt, it can be found that crumb rubber absorbed a large number of light components and wax content in asphalt, which reduced the wax content used to form a bee-shaped structure. At the same time, some long straight chain

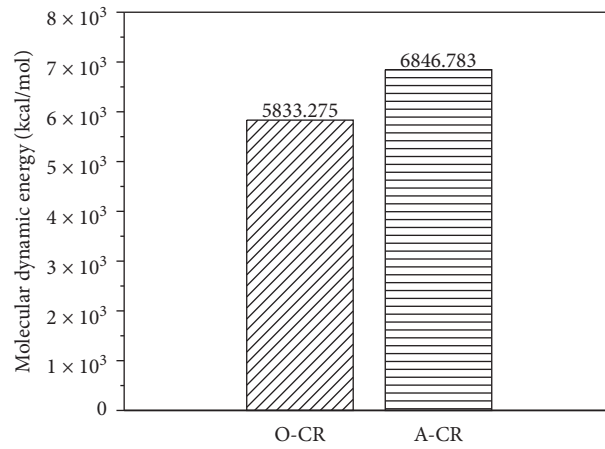


FIGURE 6: Molecular dynamics energy of O-CR and A-CR.

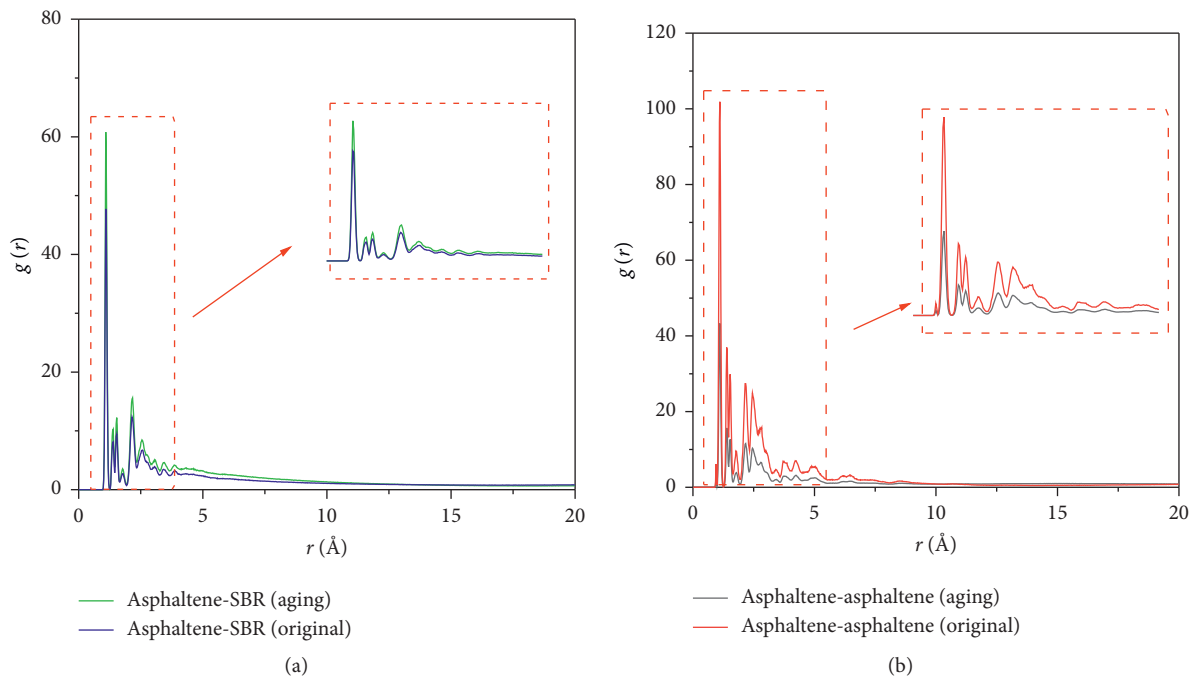


FIGURE 7: Continued.

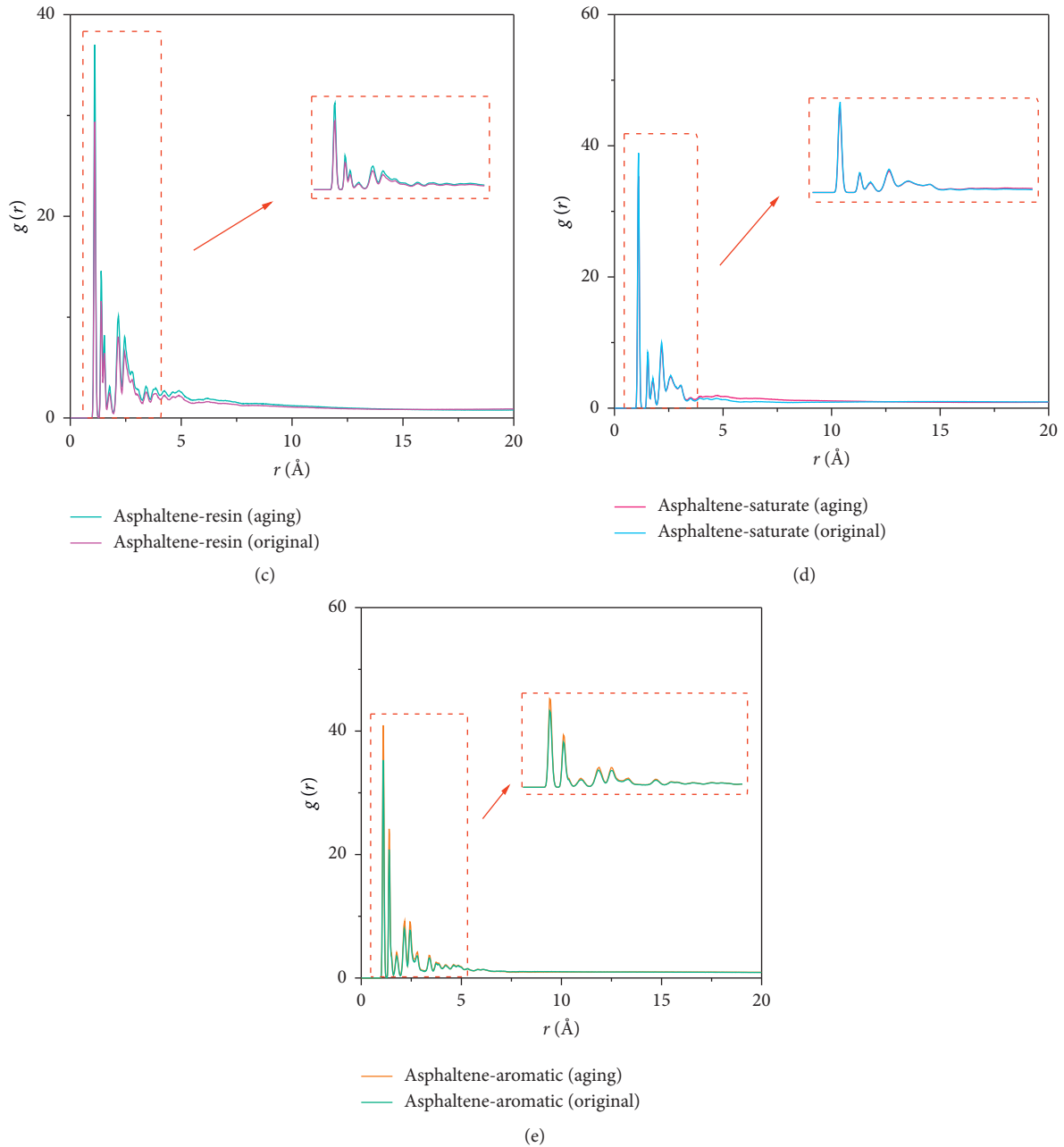


FIGURE 7: Radial distribution function for asphaltene with SBR, resin, saturate, and aromatic pairs in original and aging models. (a) Asphaltene-SBR. (b) Asphaltene-asphaltene. (c) Asphaltene-resin. (d) Asphaltene-saturate. (e) Asphaltene-aromatic.

molecules dissolved by crumb rubber also made asphaltene difficult to agglomerate on a large scale, resulting in asphaltene being more evenly dispersed in asphalt so that it was difficult to form a large bee structure. In terms of colloidal theory, the more uniform the colloidal dispersion is, the better the macroperformance will be, which should also prove the excellent macroperformance of the crumb rubber modified asphalt.

There is no obvious change in the image of the CR before and after aging. The uniformly dispersed asphaltenes and the

long-chain molecules dissolved by the crumb rubber cross-link with each other to form a more stable and uniform internal structure, so the effect of aging on it is reduced.

It was difficult to find the obvious changes of the CR before and after the aging from the morphology map, so more accurate quantitative analysis was needed to explore the microstructure changes of the CR before and after the aging. The surface roughness of the asphalt was analyzed theoretically after the first-order flatten treatment of the image with Nanoscope Analysis.

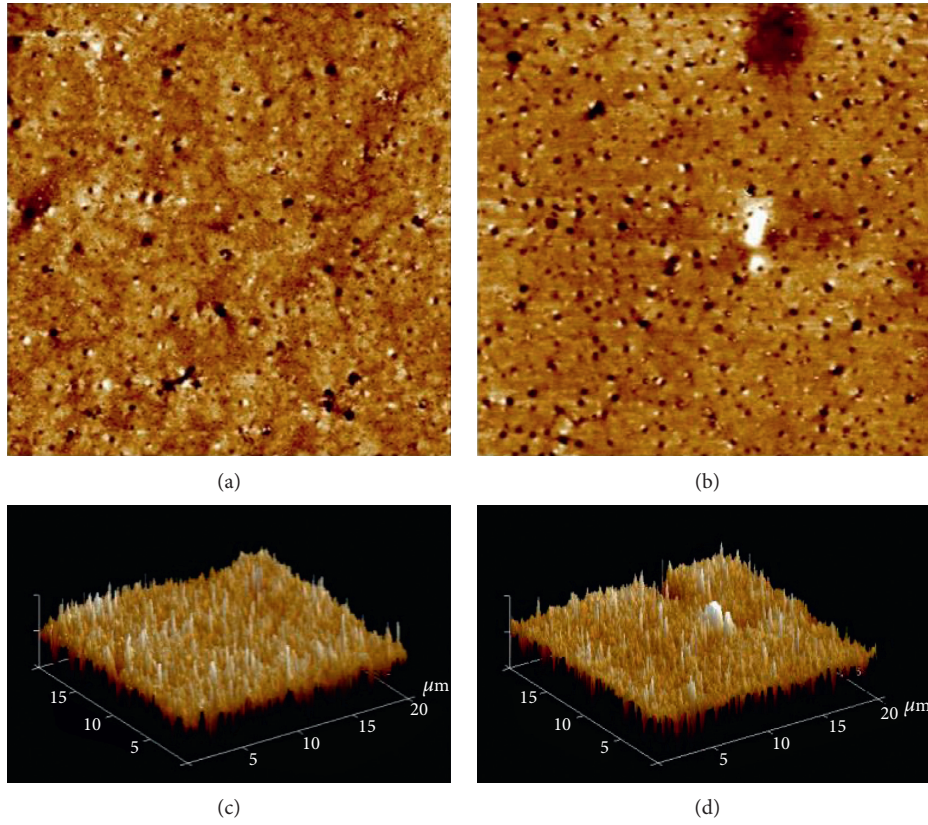


FIGURE 8: 2D and 3D microscopic morphology of O-CR and A-CR.

Root mean square roughness (R_q), surface area difference percentage (ISAD), and peak density (S_{ds}) were selected as the indexes to evaluate the micromorphology of asphalt. In a narrow sense, roughness referred to the height difference between the high and low fluctuation of the observed object surface. Due to the fact that the microstructure of the asphalt surface had slight fluctuation and peak valley intersection, R_q was selected to characterize the surface roughness of the CR before and after aging. The definition of R_q is shown in the following formula:

$$R_q = \sqrt{\frac{\iint [h(x, y) - h_0]^2 dA}{\iint dA}}, \quad (4)$$

where A is the scanning area, $h(x, y)$ is the shape height function of unit point (x, y) , h_0 is the reference height, and $h_0 = \iint h(x, y) dA / \iint dA$.

Surface area difference percentage ISAD referred to the area difference percentage from 3D surface area flattening to 2D surface in the scanning area. ISAD was also a common index in the theoretical analysis of roughness. Through ISAD, the fluctuation degree of microstructure in 3D morphology can be quantitatively analyzed. The calculation formula is shown as

$$\text{ISAD} = \frac{S_{3D} - S_{2D}}{S_{2D}} \times 100\%, \quad (5)$$

where S_{3D} is 3D surface area and S_{2D} is 2D surface area.

Peak density S_{ds} referred to the number of peaks per unit area. The peak value was defined as the value higher than the adjacent 8 points in the AFM system. In addition, the peak was only used to define the point whose height was 5% higher than the maximum height of the average plane. The peak density was the density of the peaks in the region. The peak density can quantitatively represent the amount and distribution of microstructure in the scanning area. The calculation formula is

$$S_{ds} = \frac{N_{\text{peak}}}{A}, \quad (6)$$

where N_{peak} is the number of maximum height peak structures in the area and A is the area. The results of R_q , ISAD, and S_{ds} are shown in Table 5.

It can be seen from Table 5 that there is a certain change in the surface microstructure of CR before and after aging through quantitative analysis. After aging, the ISAD and S_{ds} of CR surface were decreasing, so it can be known that the number of peak structures and the height of each peak of CR were decreasing, and the comprehensive evaluation index R_q was also decreasing; aging made the microstructure of asphalt more uniform and the surface gentler. Under the influence of aging, the light components, such as aromatics and resins of CR converted to the heavy components, and the proportion of the components of asphalt

TABLE 5: Surface roughness parameter statistics of O-CR and A-CR.

	R_q (nm)	ISAD (%)	S_{ds} (μm^{-1})
O-CR	2.14	0.0214	2.20
A-CR	1.29	0.0103	1.80

TABLE 6: Microscale and nanoscale analysis on CR.

Scale	Parameter	Unit	O-CR	A-CR
Microscale	R_q	nm	2.14	1.29
	S_{ds}	μm^{-1}	2.20	1.80
Nanoscale	Energy	kcal/mol	5833.275	6846.783
	RDF(As-as)	—	101.87	43.31

changed, which showed the process of asphalt surface homogenization. The homogenization of asphalt surface resulted in the decrease of the roughness, specific surface area, and peak density of asphalt surface. After aging, the surface of CR was more uniform and flatter than that of the original CR, and the composition and structure were more uniform and stable, which led to its good high-temperature stability.

6. Correlation Analysis of Micro/Nanoproperties of CR before and after Aging

In order to explore the relationship between the structural changes of CR at the microscale and the changes of molecular dynamics energy and radial distribution function at the nanoscale before and after aging and then to explain the changes of the microstructure before and after aging at the molecular scale, the correlation analysis of the variation of nanoenergy, RDF, and microstructure changes was carried out.

It can be seen from Table 6 that the surface of A-CR at the microscale is smoother, the number of peak structures is decreasing, and the height is decreasing. However, after aging, the molecular dynamic energy of CR increased instead. This was due to the increasing proportion of asphaltenes with larger molecular weight after asphalt aging, which led to the increase of the overall molecular weight of asphalt molecules, which increased the molecular dynamic energy. When the energy was higher, the molecular agglomeration was difficult to precipitate, resulting in a reduction in the number and height of peak structures in the micromorphology. After aging, the asphaltene-asphaltene RDF was significantly lower than the original asphaltene-asphaltene RDF, indicating that aging affected the accumulation of asphaltenes and asphaltenes. After aging, the degree of asphaltene aggregation was lower and the distribution was more dispersed. As a result, the peak structure with asphaltene as the core was dispersed in the microstructure, making the asphalt surface structure gentler. Therefore, the dynamic energy and RDF of the crumb rubber modified asphalt molecular group affected the formation of the micromorphology.

7. Conclusions

- (1) Aging increases the molecular weight of the asphalt, which leads to an increase in the molecular dynamics energy of the crumb rubber modified asphalt.
- (2) After the aging, the peak value of the RDF between asphaltenes is reduced, the asphaltenes are difficult to aggregate, and the asphaltenes are more dispersed in the molecular model.
- (3) The surface microstructure of crumb rubber modified asphalt after aging is uniform and gentle. The enhanced high-temperature performance of crumb rubber modified asphalt after aging is related to surface homogenization.
- (4) The molecular dynamics energy and radial distribution function of asphalt nanostructure have a certain effect on its microstructure, and there is a certain correlation between different phenomena observed at different scales of the same substance.

Data Availability

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

Conflicts of Interest

The authors declare that they have no conflicts of interest regarding this work.

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