Investigating the long-term aging behaviors of bitumen based on experimental chemo-physical characterizations

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Abstract

The aging resistance performance of bitumen binder is of importance to the durability of asphalt pavement, especially for long-term aging behavior. To further observe the long-term aging influence on the chemical properties of bitumen. To that end, laboratory aging methods were employed to obtain the aged binders with different long-term aging degrees. The chemical properties of virgin and various aged bitumen were evaluated using the thin-layer chromatography with flame ionization detection (TCL-FID), attenuated total reflectance-Fourier-transform infrared (ATR-FTIR) spectroscopy, elemental analyzer and gel permeation chromatography (GPC) tests.

Keywords: Long-term aging, Bitumen, Molecular models, MD simulation, Experimental characterization

1. Introduction

It is estimated that the current world use of bitumen is approximately 102 million tons per year due to its superior engineering performance and comfortability. The asphalt mixture is composed of the aggregate, sand, filler, bitumen binder and air voids. The asphalt pavement is exposed to the complex environment with the various atmosphere temperature and loading conditions, which accelerates the diseases occurrence of asphalt roads, such as the rutting deformation, thermal and fatigue cracking [1-3]. Lots of modifiers have been proposed to improve the rutting, adhesion, and cracking resistance of bitumen binder, such as the styrene-butadiene-styrene copolymer, nano-caly, carbon black, nano-silica and other nanomaterials [4-9]. Apart from the external factors, the aging phenomenon of bitumen is the internal cause of performance deterioration [10]. These diseases greatly shorten the service life of asphalt pavement, and increase the cost of reconstruction and maintenance. Moreover, a large amount of reclaimed asphalt pavement (RAP) waste material are produced [11, 12]. It was reported that the aging degree of bitumen significantly affected the selection of rejuvenators type and dosage, which was directly associated with the performance restoration of RAP material [13]. Therefore, before rejuvenation, the fully understanding on the aging mechanism of bitumen is of the significance, especially for its long-term aging behaviors.

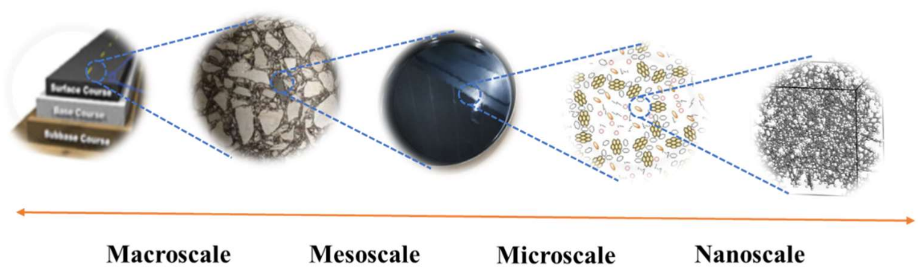

Fig.1. Multiscale illustration of asphalt materials

Currently, the multiscale methods for exploring the effects of modification, aging or rejuvenation on the properties of bitumen [14] or bitumen-aggregate interfacial systems [15, 16] from the microscale to macroscale perspectives have attracted researchers' attentions dramatically. As shown in **Fig.1**, asphalt binder and mixture are both composed of different types of molecules, and their macroscale performance is essentially determined by the microstructure, thermodynamics properties and nanoscale molecular interactions [22]. Molecular dynamics (MD) simulation is an effective method to predict the physical and mechanical properties of complex molecular systems, design proper advanced materials, as well as verify and explain the experimental results from the viewpoints of molecular interaction and microstructure. Previous studies demonstrated that MD simulation was a useful supplementary technology to further insight into the modification and rejuvenation mechanisms of bitumen binder [17-19]. Zhang et al. employed the MD simulation to verify the positive role of an anti-stripping agent on improving the cohesive, adhesive and moisture damage resistance of bitumen, and found that the improvement mechanism was due to the formation of hydrogen bonding between bitumen and anti-stripping agent molecules [20]. Su et al. investigated the effects of Nano-ZnO/SBS modifiers on the physical properties and molecular structure of bitumen through the MD simulation method, and clarified that the adequate high-temperature properties and durability of the modified binder was associated with the facilitation function of these two materials on accumulating the molecular interaction and structural tightness in bitumen system [21]. Besides, Cui et al. implemented the MD simulation to evaluate the rejuvenator effects on aged bitumen at the microscale level, and concluded that the addition of rejuvenator would not only be beneficial to improve the cracking and moisture resistance, but also mitigate the self-aggregation of asphaltene molecules and restore the colloidal structure of aged binder [22].

However, the research gaps regarding the application of MD simulation technology in the field of bituminous binder still exist. The common-used 12-components molecular models of SARA fractions for virgin bitumen did not consider the existence of the sulfoxide groups [23-27]. Moreover, the current molecular models for aged bitumen were established empirically, and the effects of long-term aging degree on the molecular structure and material composition have not been studied yet. Further, the MD simulation method is essential to explain the long-term aging mechanism and predict the important thermodynamics properties of aged bitumen from the viewpoint of nanoscale, which is difficult to implement with the macroscale experiments. Therefore, this study will develop the molecular models of virgin and different aged bitumen according to their chemical characteristics. Additionally, the molecular models will be verified with the physical properties from laboratory experiments. Further, other important parameters regarding the bulk, interfacial and dynamic properties will be predicted to further fundamentally understand the long-term aging behaviors of bitumen from the perspective of thermodynamics performance.

2. Research objective and protocol

This study aims to investigate the influence of long-term aging of bitumen on the chemical and thermodynamic properties, which are strongly related to the rheological and mechanical performance. To achieve the objective, the experimental and molecular dynamics simulation methods are incorporated. The detailed research protocols are shown in **Fig.2.** The molecular models of virgin and different aged bitumen will be established in line with their corresponding chemical properties, including the SARA group constituents, chemical functional groups, elementary composition and molecular weight distribution. Afterwards, MD simulation will be employed to study the effects of long-term aging on the thermodynamic properties of bitumen, which will be further compared with the experimental results. Lastly, the long-term aging mechanism at the molecular level will be explored.

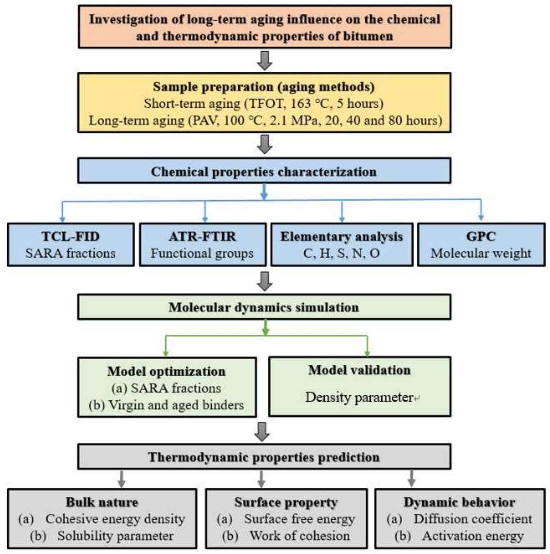

Fig.2. The research protocol of this study

3. Materials

In this study, one PEN70/100 bitumen from Total Nederland N.V. was used. The physical properties and chemical components of virgin bitumen are shown in **Table 1**.

Table 1 The physical properties and chemical components of PEN 70/100 bitumen

Properties	value	Test standard
25℃ Penetration (1/10 mm)	91	ASTM D5 [40]
Softening point (℃)	48	ASTM D36 [41]
135℃ Dynamic viscosity (Pa•s)	0.8	AASHTO T316 [42]
25℃ Density (g/cm^3)	1.017	EN 15326 [43]
60℃ Density (g/cm^3)	0.996	

In order to understand the fundamental aging influence on the chemical and thermodynamic properties of bitumen binder, the fresh bitumen binder was subjected to short-term and long-term aging processes with different aging durations. In the study, the short-term aged bitumen binder was prepared by using the thin film oven test (TFOT) at 163℃ for 5 hours, which were further transferred to the pressure aging vessel (PAV) device to manufacture the long-term aged binders under the temperature and pressure condition of 100℃ and 2.1MPa, respectively. The long-term aging durations for different aged bitumen binders were selected as 20, 40 and 80 hours according to the aging degree of bitumen in the realistic reclaimed asphalt pavement [47, 48]. For convenience, the virgin binder, short-aged and long-aged bitumen binders for 20, 40 and 80 hours are abbreviated as the VB, SAB, LAB20, LAB40 and LAB80.

4. Experimental results and discussion

4.1. Bitumen components

Fig.3 shows the mass weight of the saturate, aromatic, resin and asphaltene fractions in virgin and aged bitumen. The total weight of aromatic and resin fractions in the virgin binder is more than 80wt%, indicating that the internal colloidal structure of the bitumen shows more stable. Moreover, it can be found that the aging process has no obvious influence on the saturate content of bitumen, which is due to the saturated hydrocarbon structure of saturate with no heteroatom and less reaction potential to oxygen molecules. Thus, the conversion ratio of saturate constitution of bitumen is close to zero during the oxidation aging process. Meanwhile, as the aging degree increasing, the aromatic dosage of bitumen declines dramatically, while the concentration of both resin and asphaltene fractions increases gradually, which results in the physical hardening and colloidal structure instability of bitumen. In other words, the agglomeration of asphaltene nanoparticle in aged bitumen tends to increase, which contributes to the stiffness improvement and the crack phenomenon occurring easily for the aged binder.

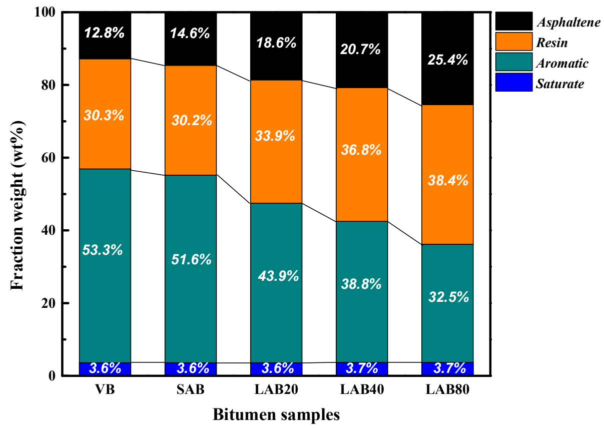

Fig.3. SARA fractions of virgin and aged bitumen

In addition, the aging degree of bitumen has a remarkable influence on the mass weight of aromatic, resin and asphaltene fractions. In detail, the aromatic concentration decreases by 1.7%, 9.4%, 14.5% and 20.8%, when the bitumen is subjected to short-term aging and long-term aging 20, 40 and 80 hours, respectively. Hence, the compensation of aromatic composition is the main way during the rejuvenation technology of aged binder in reclaimed asphalt pavement (RAP) materials. On the contrary, the corresponding asphaltene content increases by 1.8%, 5.8%, 7.9% and 12.6%, respectively. The increase of asphaltene fraction would reduce the relative distance of the asphaltene molecules and strengthen their molecular interaction, which accelerates the aggravation of the agglomeration phenomenon of asphaltene nanoparticle. It is well-recognized that the bitumen has the typical colloidal structure, where the asphaltene nanoparticle is regarded as the colloidal nucleus.

4.2. Chemical characteristics

The FTIR spectra curves of virgin and aged bitumen samples are displayed in **Fig.4.** Regarding the functional groups' characteristics of fresh bitumen, the strong peak at 1030 cm^{-1} can be observed, which shows that the sulfoxide functional groups exist in the fresh bitumen. This finding will be considered during the establishment of molecular models for virgin bitumen. Compared to the virgin binder, aged bitumen shows the strong absorption peaks at 1030 cm^{-1} and 1700 cm^{-1}, which refers to the sulfoxide and carbonyl functional groups.

To quantitatively evaluate the influence of long-term aging on the functional groups' distribution of bitumen, the carbonyl index CI and sulfoxide index SI are calculated as follows [16].

$$\text{Carbonyl index CI} = \frac{A_{1700}}{\sum A} \tag{1}$$

$$\text{Sulfoxide index SI} = \frac{A_{1030}}{\sum A} \tag{2}$$

$$\sum A = A_{(2952,2862)} + A_{1700} + A_{1600} + A_{1460} + A_{1375} + A_{1030} + A_{864} + A_{814} + A_{743} + A_{724}$$

where A_{1700} and A_{1030} refer to the area value of absorption peak at the wavenumber point of 1700 cm^{-1} and 1030 cm^{-1}, respectively. Besides, $\sum A$ is the total area of absorption peaks presented in **Fig.4**.

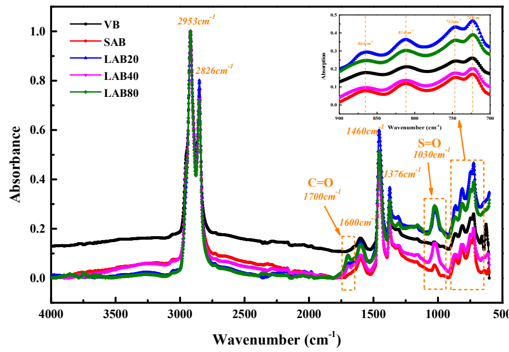

Fig.4. The detailed FTIR spectra of virgin and aged bitumen

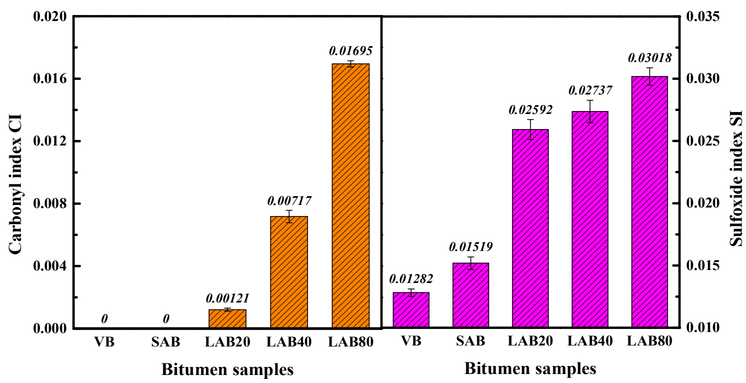

Fig.5. The carbonyl index CI and sulfoxide index SI of virgin and aged bitumen

Fig.5 shows the variations of carbonyl and sulfoxide indexes for virgin and aged bitumen with various aging conditions. It can be found that there is no carbonyl functional group observed in the fresh bitumen, which denotes that the carbonyl functional group is not sensitive to the short-term aging process. On the contrary, the sulfoxide index of virgin bitumen could be monitored, which increases after the short-term aging process. It domenstrates that the sulfoxide functional group exists in the fresh bitumen, which should be considered when the molecular models of bitumen are designed. Meanwhile, the sulfoxide index is invariably larger than the carbonyl index, regardless of the aging degree of bitumen, which may be attributed to the characteristic of the high sulfur concentration in virgin bitumen. This assumption will be validated with the elemental analysis results. In addition, both carbonyl index and sulfoxide index increase with the aging depth strengthens, which is associated with the oxidation and substitution reaction of bitumen aging.

It is interesting to note that the sensitivity of carbonyl and sulfoxide index to the aging time is significantly different. From **Fig.5**, the increasing rate of sulfoxide index is faster than carbonyl index in the short-term aging period. Moreover, the carbonyls start to form during long-term aging process and increase with aging time. In other words, the sulfoxides are generated earlier than the carbonyls, because the sulphur is more reactive than the carbon atom in bitumen. On the contrary, the carbonyls increase during long-term aging process, whereas the sulfoxide index stabilizes probably due to the full consumption of sulfur in bitumen. In conclusion, the sulfur oxidation reaction is easier to occur than the carboxylation, which agrees with the previous studies [16, 32]. **Fig.6** displays the graphical illustration of main oxidation reactions, including the generation of carbonyl and sulfoxide functional groups. From the perspective of chemical bonding theory, the bond energy of the sulfoxide group (522 kJ/mol) is lower than that of the carbonyl group (728 kJ/mol).

In addition, from the viewpoint of electronic interaction theory, the sulfur atom has two long-pair electrons, while the carbon atom has no long-pair electrons because all four electrons are occupied to form the covalent bonds with the two adjacent hydrogen and carbon atoms. Thus, it is easy for the sulfur atoms to react with the oxygen molecules and form the peroxide radical, followed by the formation of the sulfoxide functional group. That's why the sulfoxide function groups are easier to form than the carbonyl component. This result is in line with the previous literature [16, 17, 32], which revealed that the sulfoxide formation would occur earlier than the ketone group. Hence, the aging behavior and mechanism are closely related to the chemical components of virgin bitumen, especially for the sulfur element concentration.

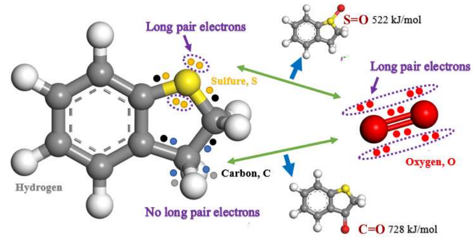

Fig.6. The graphical illustration of the main reaction during the aging of bitumen

4.3. Elemental analysis

It is well known that the bituminous material is manufactured from the crude oil refinery process, and their main elements contains the carbon C, hydrogen H and other heteroatoms, such as the sulfur S, nitrogen N and oxygen O as well as other heavy metal elements. **Table 3** displays the element compositions of virgin and aged bitumen. Overall, the carbon content C% is in the majority, which is more than 80%. Furthermore, the hydrogen concentration H% is larger than 10% for all tested samples. Importantly, it should be mentioned that the sulfur dosage S% is about 3.5%, indicating that the bitumen comes from the high-sulfur crude oil and this result is consistent with the observation of the sulfoxide functional group in the fresh bitumen from the FTIR test. Therefore, the sulfur element cannot be ignored to improve the accuracy of molecular dynamics simulation results. For virgin bitumen, both nitrogen N% and oxygen concentration O% are lower than 1%.

It can be found in **Table 3** that the aging of bitumen has no obvious influence on the nitrogen and sulfur concentration, which significantly affects the proportions of other elements. With the increase of aging degree, the carbon and hydrogen element contents both reduce, while the oxygen concentration increases dramatically. This indicates that the oxidation degree of the binder deepens gradually as the aging time increasing. To further assess the influence of aging on the molecular structure of bitumen, the ratio of H/C is calculated and shown in **Fig.7.** Here, the H/C value refers to the ratio of hydrogen atom number (N_H) to carbon atom number (N_C), which is calculated as equation (3):

$$\frac{H}{C}=\frac{N_H}{N_C}=\frac{\frac{H\%}{M_H}}{\frac{C\%}{M_C}} \tag{3}$$

where H% and C% are the mass fraction of hydrogen and carbon element in bitumen; and M_H and M_c represent the molar weight of C and H atom, which are set as 1g/mol and 12g/mol, respectively.

Table 3 The element compositions of virgin and aged bitumen

Bitumen samples	N (wt%)	C (wt%)	H (wt%)	S (wt%)	O (wt%)
VB	0.90	84.06	10.902	3.52	0.618
SAB	0.91	83.72	10.856	3.51	1.004
LAB20	0.92	83.26	10.748	3.49	1.582
LAB40	0.89	83.02	10.437	3.53	2.113
LAB80	0.91	82.14	10.098	3.54	3.312

The value of H/C ratio decreases remarkably with the increase of long-term aging time. Compared to the virgin bitumen, the H/C ratio value decreases from 1.556 to 1.475. This phenomenon reveals that the aging of bitumen would strengthen the degree of unsaturation and aromaticity. On the one hand, during the aging of bitumen, light constituents (aromatic fraction with low polarity) would transfer to the heavy components (resin and asphaltene fractions with high aromaticity). On the other hand, the oxidation reaction during bitumen aging would result in the proportion enhancement of the carbonyl and sulfoxide function groups, which increases the unsaturation degree. The phenomena have been validated by the FTIR results. Therefore, the aging mechanism of bitumen is the synergistic effect of components transforming and oxidation reaction. These chemical characterization results provide the data support and theoretical basis to the establishment of the molecular models of virgin and aged binders in following molecular dynamics simulation procedure.

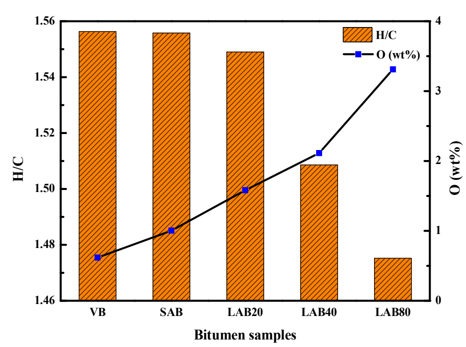

Fig.7. The aging influence on the H/C ratio and oxygen content in bitumen

4.4. Molecular weight distribution

From the stand point of polymer science, the molecular weight of material is closely associated with its thermodynamics and mechanical properties, such as the diffusion coefficient, viscosity, stiffness and etc. It is of great importance to track the influence of oxidation aging on the molecular weight distribution of bitumen, which provides the basis for determining the molecular models of aged binders with various aging levels. In this study, the weight-average molecular weight (M_w) and the number-average molecular weight (M_n) of bitumen binders were measured by using the GPC test, and the GPC curves are illustrated in **Fig.8**. Previous studies reported that in GPC curve of bitumen, the first peak (3,000-19,000 Daltons) refers to the asphaltene fraction, and the second peak (<3,000 Daltons) represents the maltene fractions [15, 18]. It is worth noting that aging has a great influence on the molecular weight distribution of bitumen, especially for the heavy fractions. With the aging degree deepens, the retention time of heavy fractions decreases, and the refractive index increases dramatically. It indicates that more heavy constituents are generated during the aging process of bitumen, which is related to the polymerization reactions of the light fractions and agglomeration of the heavy molecules. The GPC result is consistent with the SARA fractions distribution, which verifies the fraction conversion process from the light to heavy compositions and provides the essential basis for establishing of molecular models for aged bitumen with various aging degrees.

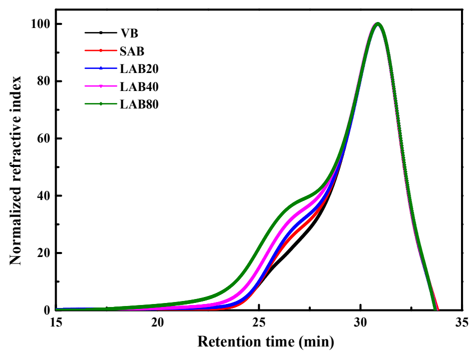

Fig.8. The GPC results of virgin and aged bitumen

Table 4 The average molecular weight (Daltons) of virgin and aged binders

Samples	Weight average molecular weight M_w	Number average molecular weight M_n	Dispersion parameter D
VB	3879	1290	3.01
SAB	4049	1310	3.09
LAB20	4773	1346	3.40
LAB40	5802	1400	4.15
LAB80	8210	1488	5.52

The number-average molecular weight M_n, weight-average molecular weight M_w, and the dispersion parameter of virgin and aged bitumen D, are calculated as follows:

$$\mathrm{M}_w = \frac{\sum_{i=1}^{n} w_i \times M_i}{\sum_{i=1}^{n} w_i} \tag{4}$$

$$\mathrm{M}_n = \frac{\sum_{i=1}^{n} N_i \times M_i}{\sum_{i=1}^{n} N_i} \tag{5}$$

$$D = \frac{\mathrm{M}_w}{\mathrm{M}_n} \tag{6}$$

where w_i and N_i refers to the weight and number of molar mass M_i, respectively.

The influence of aging on the molecular weight and dispersion value of bitumen are found in **Table 4**. With the aging degree deepening, the values of M_n and M_w of bitumen all strengthen.

It is caused by the transformation from the aromatic fractions (light molecular weight and low polarity) into the resin/asphaltenes molecules (heavy molecular weight and high polarity). It is interesting to observe that the weight-average molecular weight is higher than the number-average molecular weight, which is attributed to the large difference in molecular weight distribution, especially for the saturate and asphaltene fractions. The molecular weight distribution difference could be estimated with the dispersion parameter (D), and the aging process significantly increases the D value of bitumen. Hence, it could be summarized that the increase of aging degree would result in the more uneven molecular weight distribution because of the increase of the heavy-weight molecules and decrease of the light-weight fractions. In addition, the D parameter could also be applied to assess the dispersion ability of maltene to asphaltene, and the higher D value represents the lower dispersion ability of maltene. The aging of bitumen weakens the dispersion ability of maltene to asphaltene, while the agglomeration of asphaltene molecules and unstable colloidal structures would occur easily.

7. Conclusions

This study investigated the influence of laboratory long-term aging on the chemical and thermodynamic properties of bitumen through the combination of experimental and molecular dynamics simulation methods. The main conclusions can be drawn as follows:

(1) During the long-term aging of bitumen, the aromatic component decreased dramatically, while the resin and asphaltene fractions increased gradually. Besides, the sulfoxide and carbonyl functional groups would generate, and the sulfur oxidation reaction was easier than the carbon oxidation.

(2) The C%, H% and H/C ratio value would decrease during the long-term aging of bitumen, which markedly increased the oxygen dosage and average molecular weight due to the polymerization reaction of light fractions and agglomeration of heavy molecules.

(3) The molecular models of virgin and different aged bitumen were established and validated. The functional groups, average molecular weight and density outputs from the MD simulation agreed well with the laboratory results

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