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Do different chemical and rheological properties act as effective and critical indicators for efficiency evaluation of rejuvenated bitumen?

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A R T I C L E I N F O

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A B S T R A C T

This study investigates the impacts of rejuvenator type/dosage and the aging degree of bitumen on the chemical and rheological properties of rejuvenated bitumen, and propose critical chemo-rheological indicators for evaluating rejuvenation efficiency. Moreover, the potential connections between essential chemical and rheological indices of rejuvenator-aged bitumen blends are explored. Results indicate that chemical indices show linear relationships with rejuvenator dosage and vary depending on the rejuvenator type and aging level of bitumen. All rejuvenators can regenerate certain rheological parameters of aged bitumen to varying degrees, but cannot restore the crossover modulus (G_c). Various rheological indices exhibit different correlations with rejuvenator dosage and sensitivity degrees to the discrepancy in rejuvenator type and aging degree of bitumen. Critical chemical and rheological indicators are proposed based on their sensitivity levels to influence factors, with the aromaticity index (AI), carbonyl index (CI), and sulfoxide index (SI) as effective chemical indices and the complex modulus (G*), crossover frequency (f_c), and high-temperature master curve area (A_{MH}) as critical rheological indices for rejuvenation efficiency evaluation. The study finds that the magnitude of rejuvenation efficiency for four rejuvenators is Bio-oil > Engine-oil > Naphthenic-oil > Aromatic-oil, and the linear correlations between the critical chemical and rheological indices, together with their rejuvenation percentages, are significantly affected by the rejuvenator type and aging level of bitumen.

1. Introduction

At present, there is a growing push for the adoption of circular economy and sustainability concepts, with a notable focus on the utilization of reclaimed asphalt pavement (RAP) materials in maintenance and reconstruction projects, capturing the interest of experts in the field of pavement [1,2]. Maximizing the recycling ratio of RAP is appreciated, but it would deteriorate the mechanical performance of asphalt pavement, particularly the resistance to cracking and moisture damage [3–5]. The aging-induced increase in stiffness contributes a lot to the insufficient low-temperature, fatigue, and adhesion performance of RAP binder due to the evaporation and conversion from lightweight molecules (saturate and aromatic) to resin and asphaltene fractions with large polarity and heavy molecular weight [6,7].

The primary approach for achieving a balance among components and improving the viscoelastic properties of aged bitumen is through the integration of lightweight additives known as rejuvenators. Over time, a variety of rejuvenators have been created, and researchers have extensively examined their impact on the chemical [8], physical [9], microstructural [10], and rheological [11] characteristics of the aged binder. It is widely reported that the rejuvenator type plays a decisive role in rejuvenation efficiency [12]. According to the recommendation from the America National Center for Asphalt Technology (NCAT), the rejuvenators are classified into five groups (fatty acids, aromatic extract, naphthenic oils, paraffinic oils, and tall oils) [13]. Zhou et al. concluded that a rejuvenator with high aromaticity diluted the asphaltene clusters and stabilized the colloidal structure of aged bitumen [14]. Meanwhile, it was reported that bio-oils could regenerate the chemical and mechanical properties but increase the risk of moisture damage of rejuvenated bitumen [15]. Nonetheless, the variety of rejuvenator types and the varying degrees of bitumen aging significantly complicate the task of devising a standardized evaluation criterion for assessing the efficiency of different rejuvenators in aging bitumen blends [16–18].

Furthermore, a range of material characteristics are assessed quantitatively to evaluate how effectively rejuvenators contribute to restoring the performance of aged bitumen [19]. Some commonly-used chemical and rheological indices for rejuvenation efficiency evaluation are listed in Table 1. The complex modulus (G*), phase angle (δ), G-R

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Table 1
The summary of commonly-used rheological and chemical indices.

Case	Rejuvenator type	Rheological parameters	Chemical indices	Ref
1	OP900;	G* , δ, m, S	CI, SI	[22]
2	Tall-oil, aromatic extract	G* ,δ, G′, G", ΔTc	SARA fraction, CI,	[23]
3	Naphthenic-, paraffinic-, and biomaterial-based rejuvenators	G* , δ, m, S	CI, SI	[24]
4	Bio-oils (caster oil, gutter oil, soybean oil, straw oil, vegetable oil)	G* /δ, Jnr, G′, G", S, m-value	$I_{S=O}$, $I_{C=C}$	[25]
5	Paraffinic oil, aromatic extract, naphthenic oil, triglycerides/fatty acids, tall oil	G* , δ, G-R, S, m	SARA fraction, colloidal index	[26]
6	80/100 soft bitumen, warm mix surfactant (Evotherm-DAT), vegetable oil-derived rejuvenator	Jnr, N_f	CI, SI, M_n, M_w	[27]
7	Aromatic extract, waste vegetable oil	G* , δ, S, m-value	SARA fraction,	[28]
8	Different components from waste cooking oil	G* , δ, S, m	FTIR cluster analysis	[29]
9	V12000 soft bitumen, aromatic nitrogen oil	G* , δ, G-R, $T_{\delta=45}$	I_{AR}, I_{AL}, I_{CO}, I_{SO}	[30]
10	Waste cooking oil, waste bio-oil, waste engine-oil	PG, η, σ, T_{CR}	SARA components, Instability index Ic,	[31]
11	Agriculture-based rejuvenator, petroleum-based rejuvenator	G* , δ	SARA fraction, colloidal index CI, $I_{C=O}$, $I_{S=O}$, I_{Al}, I_{Ar}	[20]
12	Soy-based bio-oils (SB1, SB2, SB3)	Jnr, N_f	CI, SI	[32]
13	Vegetable oil, tall oil, bio oil, aromatic extract, paraffinic oil	G* , G-R,	AFM microstructure, T_g	[33]
14	Waste cooking oil, chemically modified WCO, and hydrolene	G* , δ, G-R, f_c, Jnr	-	[21]
15	Plant extract, vegetable oil, soy-based oil, petroleum-based, waste vegetable oil, rubberwood oil, swine manure, algae	G* , δ, f_c, G_c,	Colloidal stability index CI	[34]

Note: G* -complex modulus, δ-phase angle, m value-creep rate, S-stiffness, G′-storage modulus, G″-loss modulus, ΔTc-critical temperature difference, G* /sinδ-rutting parameter, Jnr-creep compliance, N_f-fatigue life, η-viscosity, f_C-crossover frequency, G_C-crossover modulus, CI-carbonyl index, SI-sulfoxide index, SARA-saturate, aromatic, resin, asphaltene, $I_{S=O}$-sulfoxide index, $I_{C=C}$-aromaticity index, M_n-number average molecular weight, Mw-weight average molecular weight, AFM-atom force microscopic, T_g-glass transition temperature.

parameters from DSR tests, stiffness (S), and creep rate m-value from BBR tests are always adopted to assess the rejuvenation efficiency of various rejuvenators. Meanwhile, the preferred chemical indices are the SARA fractions (Saturate, Aromatic, Resin, Asphaltene), colloidal index CI, carbonyl index $I_{C=O}$, and sulfoxide index $I_{S=O}$. In addition to material characteristics, the fluctuation in rejuvenation effectiveness is also influenced by the arbitrary choice of these chemical and rheological parameters as evaluation criteria in various studies. However, limited research focuses on comparing and determining the critical chemo-rheological parameters for the rejuvenation efficiency evaluation of variable rejuvenator-aged bitumen blends [20,21]. Further standardizing the evaluation criteria is essential to better comprehend the variations in the efficiency of rejuvenators, investigate the underlying mechanisms, and create new rejuvenators with desired characteristics.

The intimate relationships between chemical and rheological indices of bituminous materials were studied, and it was believed that the changes in chemical compositions contributed to mechanical performance variations [28]. For example, Fan et al. found that the SI/CI values of aged and rejuvenated binders exhibited correlations with various rheological parameters (G*, δ, G*/sinδ, and S) [22]. However, the chemo-rheological links established and discussed were specific to limited rejuvenation scenarios. Furthermore, the introduction of a new material (rejuvenator) into aged bitumen significantly deviates from the chemo-rheological relationships derived from the bitumen system [35–37]. However, the effects of rejuvenator type/dosage and aging level of bitumen on these chemo-rheological connections of rejuvenated binders have not been studied systematically. Based on the literature review, some challenges are still existed, for instance:

(a) A standardized chemo-rheological measure for evaluating the effectiveness of various rejuvenation scenarios is lacking.

(b) Chemical and rheological parameters of rejuvenated bitumen are notably influenced by both the composition of the rejuvenator and the degree of bitumen aging, yet their combined impact on rejuvenation efficiency has not been systematically examined.

(c) There is limited research available that investigates and establishes chemo-rheological performance connections for diverse blends of rejuvenated bitumen.

2. Research objective and parameters

The aim of this study is to comprehensively comprehend the trends in typical chemical parameters and rheological characteristics of rejuvenated bitumen. This investigation will account for the combined effects of factors such as the type and dosage of the rejuvenator and the degree of bitumen aging. Subsequently, the study intends to propose reliable chemo-rheological indicators for the simultaneous evaluation and differentiation of the rejuvenation efficiency of different rejuvenators applied to aged binders. Lastly, it will explore potential correlations between the chemical and rheological properties of rejuvenated bitumen, as such correlations are commonly observed in bitumen and aged bitumen systems.

Table 2 lists the chemical and rheological indices selected in this study for virgin, aged, and rejuvenated bitumen. One type of virgin bitumen was selected for preparing long-term aged bitumen with variable aging levels. Afterward, several rejuvenated binders with different rejuvenator types and dosages are manufactured for further chemo-

Table 2
Chemical and rheological indices involved in this study.

Samples	Chemical indices	Rheological indices
Virgin bitumen	Aromaticity index AI	Complex modulus G*
Aged bitumen	Aliphatic index AlI	Phase angle δ
(LAB20, LAB40, LAB80)	Branched aliphatic index BAlI	Crossover modulus Gc
BO rejuvenated bitumen	Long chains index LCI	Crossover frequency fc
EO rejuvenated bitumen	Carbonyl index CI	G* master curves area (A_{ML}, A_{MH}, and A_{MT})
NO rejuvenated bitumen	Sulfoxide index SI	
AO rejuvenated bitumen		

Note: BO-bio oil, EO-engine oil, NO-naphthenic oil, AO-aromatic oil.

Table 3
The chemo-physical properties of Total 70/100 bitumen.

Items	Properties	Value	Test standard
Physical indicators	Density (25 ℃, g/cm^3)	1.017	EN 15326
	Penetration (25 ℃, 1/10 mm)	91	ASTM D35
	Softening point (℃)	48.0	ASTM D36
	Viscosity (135 ℃, Pa·s)	0.80	AASHTO T316
Element analysis	Carbon C (wt%)	84.06	ASTM D7343
	Hydrogen H (wt%)	10.91	
	Nitrogen N (wt%)	0.90	
	Oxygen O (wt%)	0.62	
	Sulfur S (wt%)	3.52	
SARA fractions	Asphaltene As (wt%)	12.8	ASTM D4124
	Resin R (wt%)	30.3	
	Aromatic A (%)	53.3	
	Saturate S (%)	3.6	
Mechanical properties (60 ℃, 1.6 Hz)	Complex modulus G* (kPa)	2.4	AASHTO M320
	Phase angle δ (°)	84.5	

rheological analysis. The Attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy and dynamic shear rheometer (DSR) are utilized to detect the combined effects of rejuvenator type/dosage and aging grade of bitumen on the chemical and rheological properties of rejuvenated binders, respectively.

3. Materials and evaluation methods

3.1. Bitumen and rejuvenators

This study utilized a Total 70/100 bitumen due to its extensive application in Europe. The chemo-physical properties of the virgin bitumen are summarized in Table 3. In addition, four rejuvenators from different categories were selected following the rejuvenator classifications made by NCAT [13], including bio-oil (BO), engine-oil (EO), naphthenic-oil (NO), and aromatic-oil (AO). Different basic indicators of these rejuvenators are listed in Table 4.

3.2. Preparation of aged and rejuvenated bitumen

The initial bitumen underwent aging treatments of both short-term and long-term durations, utilizing the Thin Film Oven test (TFOT) and Pressure Aging Vessel (PAV). In the TFOT test, the temperature and aging time were set at 163 ℃ and 5 h, respectively, while the PAV test was conducted at a temperature of 100 ℃ and a pressure of 2.1 MPa. To achieve varying degrees of long-term aging for the bitumen, aging times were adjusted to 20, 40, and 80 h. The corresponding aged bitumen samples were labelled as LAB20, LAB40, and LAB80 (1 P, 2 P, and 4 P for the rejuvenated bitumen abbreviations).

There aged bitumen (LAB20, LAB40, and LAB80) was firstly preheated to 160 ℃, and rejuvenator was blended for 10 min to manufacture the rejuvenated bitumen. To the LAB20, the rejuvenator dosages are 1.25%, 2.5%, 3.75%, 5%, 7.5%, and 10%, which increases from 2.5% to 15% with an interval of 2.5% to both LAB40 and LAB80 binders considering their high aging degrees. It should be noted that the abbreviation of one

Table 4
The basic indicators of four rejuvenators.

	Rejuvenators	BO	EO	NO	AO
Physical	Density (25 ℃, g/cm^3)	0.911	0.833	0.875	0.994
	Viscosity (25 ℃, cP)	50	60	130	63100
	Flash point (℃)	265-305	> 225	> 230	> 210
Chemical	Nitrogen N (%)	0.15	0.23	0.12	0.55
	Carbon C (%)	76.47	85.16	86.24	88.01
	Hydrogen H (%)	11.96	14.36	13.62	10.56
	Sulfur S (%)	0.06	0.13	0.10	0.48
	Oxygen O (%)	11.36	0.12	0.10	0.40
	Mn (g/mol)	286.43	316.48	357.06	409.99

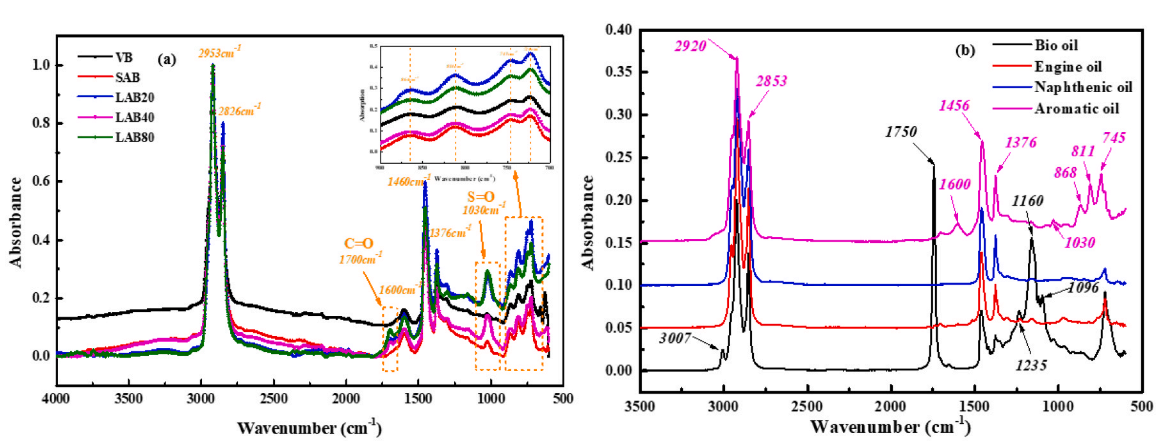

Fig. 1. FTIR curves of virgin/aged bitumen (a) and rejuvenators (b).

specific rejuvenated bitumen is composed of the aging level of bitumen and rejuvenator type/dosage. The 2P10B rejuvenated bitumen is prepared by adding a 10% bio-oil rejuvenator to LAB20-aged bitumen.

3.3. ATR-FTIR measurements

This study performs the ATR-FTIR test to detect the impact of rejuvenation treatment on the chemical properties of aged bitumen. During the FTIR measurement, the scan number and wavenumber region are set as 12 and 600–4000 cm^{-1} [38]. It should be mentioned that three same specimens were tested to ensure data reasonability.

The FTIR results are shown in Fig. 1. During the long-term aging procedure, the FTIR peaks of C═O and S═O peaks enlarge gradually, and the peak intensity of 1600 cm^{-1} also increases, representing the C═C feature in aromatic molecules. All FTIR peaks of engine-oil, naphthenic-oil, and aromatic-oil rejuvenators can be found in bitumen. The reason is that the origin of these three rejuvenators is similar to bituminous materials derived from the crude oil refinery. However, the aromatic-oil shows an additional peak at 1600 cm^{-1} resulting from aromatic molecules, while the EO and NO rejuvenators mainly comprise alkanes. In addition, two peaks at 868 and 811 cm^{-1} are also found, which refer to the Meta and Para C-H bend. For bio-oil rejuvenator, several supernumerary peaks at 1750, 1235, and 1160 cm^{-1} are observed, related to the ester functional group inside. Thus, these specific extra functional groups in AO and BO rejuvenators should be discussed separately, failing to evaluating rejuvenation efficiency due to their particularity.

In this study, these commonly-used chemical indices of bitumen with different aging and rejuvenation conditions will be calculated: aromaticity index (AI), aliphatic index (AlI), branched aliphatic index (BAII), long chains index (LCI), carbonyl index (CI), and sulfoxide index (SI).

$$\text{Aromaticity index AI} = \frac{A_{1600}}{\sum A} \tag{1}$$

$$\text{Aliphatic index AlI} = \frac{A_{1460} + A_{1375}}{\sum A} \tag{2}$$

$$\text{Branched aliphatic index BAII} = \frac{A_{1375}}{A_{1460} + A_{1375}}4 \tag{3}$$

$$\text{Long chains index LCI} = \frac{A_{725}}{A_{1460} + A_{1375}} \tag{4}$$

$$\text{Carbonyl index CI} = \frac{A_{1700}}{\sum A} \tag{5}$$

$$\text{Sulfoxide index SI} = \frac{A_{1030}}{\sum A} \tag{6}$$

$$\sum A = A_{(2952,2862)} + A_{1700} + A_{1600} + A_{1460} + A_{1375} + A_{1030} + A_{864} + A_{814} + A_{743} + A_{725} \tag{7}$$

where A_{1700}, A_{1600}, A_{1460}, A_{1375}, A_{1030}, and A_{725} represent the absorption peak area in FTIR curves at 1700, 1600, 1460, 1375, 1030, and 725 cm^{-1}, respectively. Besides, $\sum A$ refers to the whole area of all detected absorption peaks.

3.4. Dynamic shear rheometer tests

The frequency sweep tests were conducted on bitumen specimens with a frequency region of 0.1–100 rad/s. To build the complex modulus (G*) and phase angle (δ) master curves, the temperature varies from 0 °C to 70 °C with an interval of 10 °C. The reference temperature is 20 °C. This study involved seven rheological indices to estimate the rejuvenation efficiency of various rejuvenator-aged bitumen blends.

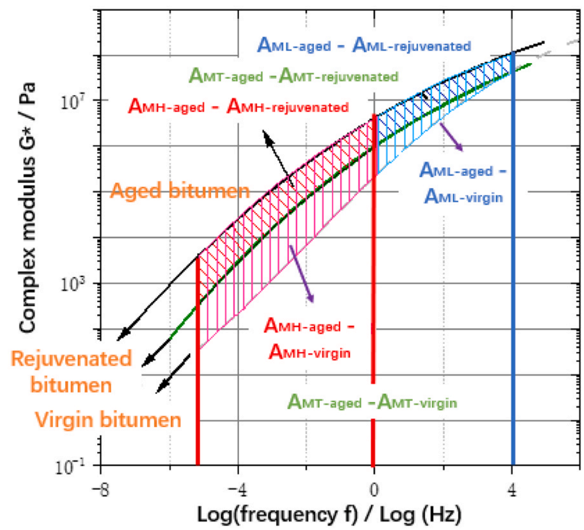

Fig. 2. Graph illustration of A_{ML}, A_{MH}, and A_{MT} parameters derived from G* master curve.

- G* and δ parameters at 60 °C and 10 rad/s, obtained directly from master curves.
- Crossover modulus (G_c) and crossover frequency (f_c) are the complex modulus and frequency values when the phase angle is equal to 45°.
- A_{ML}, A_{MH}, and A_{MT}: The area under the G* -master curve (shown in Fig. 2) at low temperatures (high frequency of 1-10^4 Hz), high temperatures (low frequency of 10^{-5}-1 Hz), and whole temperature region (frequency rises from 10^{-5} to 10^4 Hz) [39]. These three parameters are to evaluate the rejuvenation efficiency on the rheological properties within the whole temperature-frequency region, calculated as follows:

$$A_{MH} = \int_{-5}^{0} \log(G^*) \bullet \log(f)df \tag{8}$$

$$A_{ML} = \int_{0}^{4} \log(G^*) \bullet \log(f)df \tag{9}$$

$$A_{MT} = A_{MH} + A_{ML} = \int_{-5}^{0} \log(G^*) \bullet \log(f)df + \int_{0}^{4} \log(G^*) \bullet \log(f)df \tag{10}$$

where G* and f are the complex modulus and reduced frequency values of bituminous material, respectively.

4. Results and discussion

4.1. Chemical indices of aged bitumen

Fig. 3 illustrates the changes in chemical indices of bitumen due to the long-term aging process. As the long-term aging time prolongs, the aromaticity index (AI), long chains index (LCI), carbonyl index (CI), and sulfoxide index (SI) increase, while the aliphatic index (AI) and branched aliphatic index (BAII) decreases. This suggests that the level of aromaticity and quantity of C═O and S═O functional groups in bitumen increase as a result of the thermal reactions between bitumen and oxygen molecules [40,41]. The reduction of AI and BAII indices mainly resulted from the chain scission reactions. It is interesting to note that the ratio of long-chain molecules shows a certain increase. This occurs because the thermal oxidation mechanism of bitumen molecules involves a free radical reaction. The steps of chain growth and chain termination encourage the creation of long-chain molecules, which also incorporate aromatic compounds with extended side-chain groups.

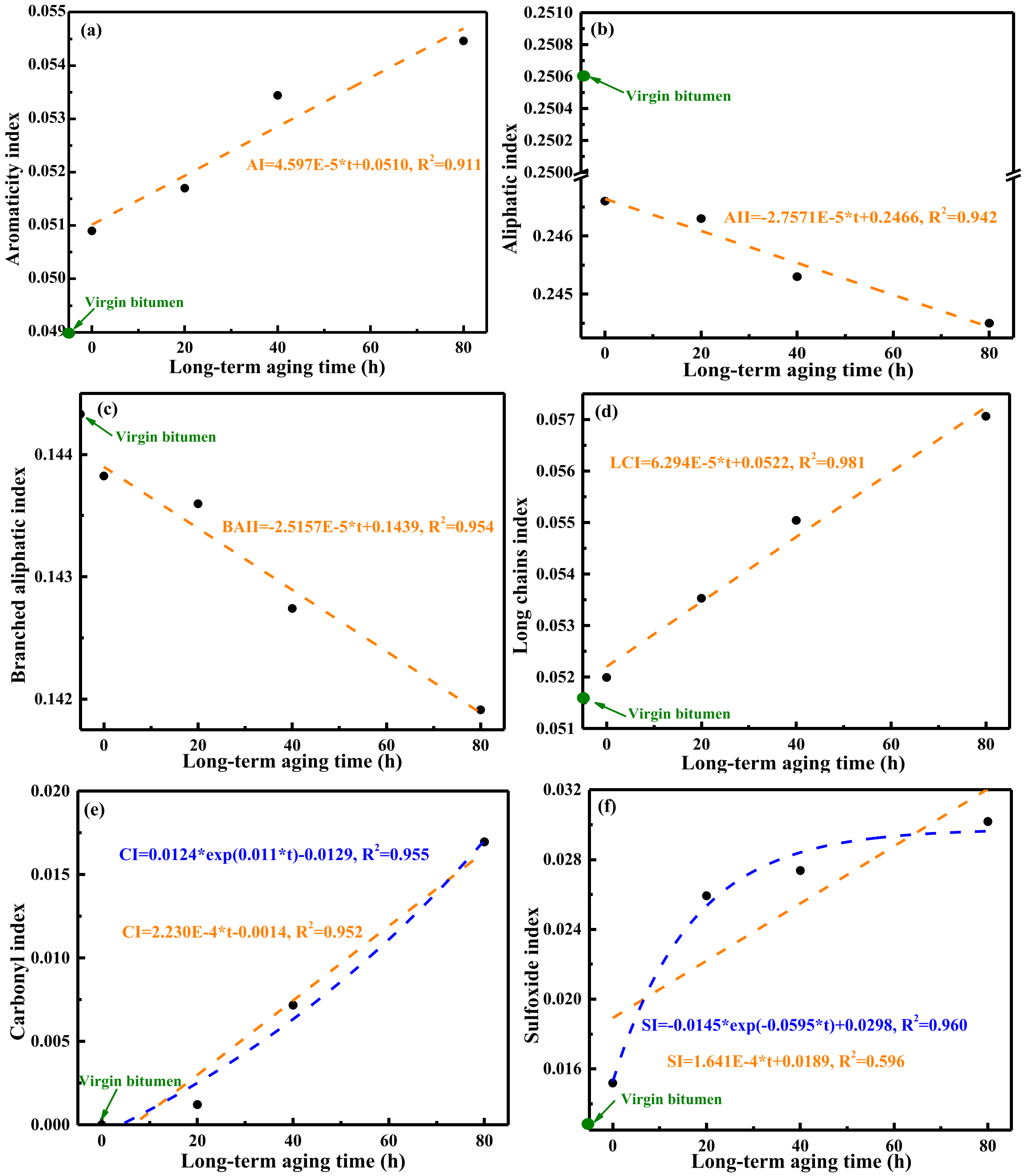

Fig. 3. Influence of aging on the chemical indices of bitumen.

There are linear relationships between long-term aging time and AI, AII, BAII, and LCI indices. The absolute slope values of these equations (magnitude in 10^{-5}) indicate these four chemical indices exhibit similar sensitivity to the aging degree of bitumen, and the exact sensitivity order follows LCI > AI > AII > BAII. Moreover, the CI parameter shows a linear increment as the aging time rises, and the corresponding sensitivity is lower than other parameters. The correlation equations can be used to forecast the chemical properties of aged bitumen with different levels of aging. Conversely, the rate of increase in sulfoxide content diminishes gradually, a trend attributed to the limited presence of sulfur atoms within bitumen molecules. The chemical indices of aged bitumen with different aging levels are important to detect the influence of rejuvenator type/dosage and evaluate their rejuvenation efficiency on restoring the chemical properties.

4.2. *Chemical indices of rejuvenated bitumen*

This section focuses on the effects of rejuvenator type and content on different chemical indices of rejuvenated bitumen according to variation trends and calculated rejuvenation percentages with the below equation:

$$XR = \frac{(X_{aged} - X_{rejuvenated}) \times 100}{X_{aged} - X_{virgin}} \quad (11)$$

where XR represents the chemical index-based rejuvenation percentage, and X herein is aromatic index (AI), aliphatic index (AII), branched aliphatic index (BAII), long chains index (LCI), carbonyl index (CI), or sulfoxide index (SI).

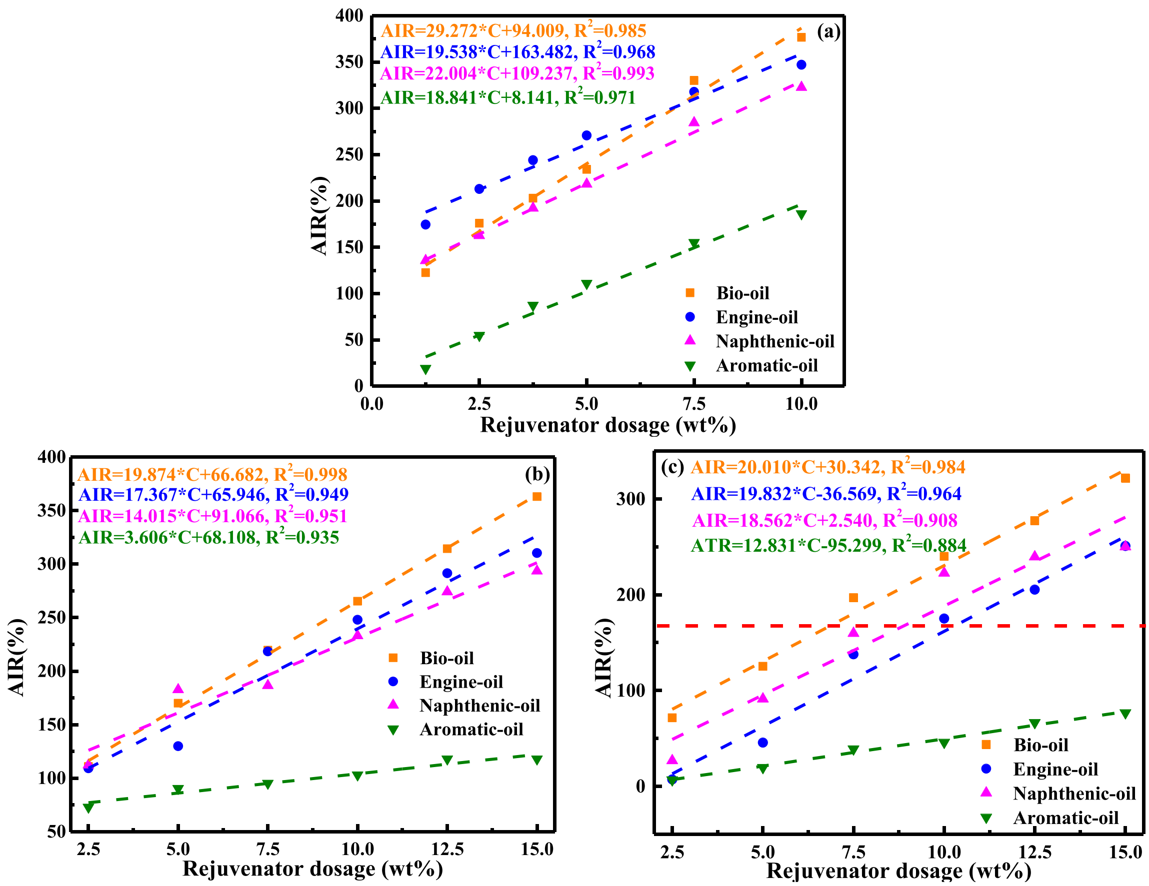

Fig. 4. AI-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 bitumen.

4.2.1. *Aromaticity index AI-based rejuvenation percentage AIR*

The AIR results are displayed in Fig. 4. All rejuvenators show positive effects on restoring the AI parameter of aged bitumen, which enlarges linearly as the rejuvenator dosage rises. Regardless of the aging grade of bitumen, the ATR values of aromatic-oil rejuvenated bitumen are much lower than the other three groups of rejuvenated binders. It is related to more aromatic molecules in aromatic-oil than other rejuvenators, which are still lower than aged binders. Interestingly, a small amount (less than 5%) of rejuvenators could restore the AI parameter by more than 100% for BO, EO, and NO rejuvenators. In addition, the bio-oil rejuvenator exhibits the most significate reduction effect on the ATR parameter. Thus, the ATR indicator provides an effective means to differentiate the rejuvenation efficiency of the AO rejuvenator from the other three rejuvenators. Nevertheless, there is minimal disparity in ATR values among the BO, EO, and NO-rejuvenated binders, with certain points of overlap, particularly in LAB20 and LAB40. The order of dosage sensitivity for these four rejuvenated binders is the same as BO > EO > NO > AO, while the sensitivity level is affected by the aging status of bitumen. The ATR parameter can distinguish the aromatic-oil from the other three rejuvenators.

4.2.2. *Aliphatic index AII-based rejuvenation percentage AIIR*

The characteristic peaks of 1375 and 1460 cm^{-1} in FTIR curves are related to the number of aliphatic groups in bitumen. The AIIR of rejuvenated binders are summarized in Fig. 5. The negative AIIR values manifest the impossibility of adding rejuvenators to restore the aliphatic index of aged binders. Regardless of aging level, the magnitude of AIIR values of four rejuvenated binders follows BORB < EORB < NORB < AORB. This indicates that the bio-oil rejuvenator has the most pronounced impact on reducing the AII value of aged bitumen, whereas the aromatic-oil has the least significant effect. The distinct molecular compositions and functional group distributions in four rejuvenators pose a challenge when it comes to making quantitative comparisons regarding influence on the specific chemical properties of aged bitumen. The linear correlations between the AIIR and rejuvenator dosage are observed in all rejuvenation cases. Furthermore, the aging level of bitumen and rejuvenator type show a coupling effect on the AIIR values of rejuvenated bitumen, and the influence of the rejuvenator type is more prominent. It should be mentioned that the AII value of rejuvenated bitumen with various rejuvenator dosages can be predicted with these correlation equations. Overall, the AII parameter is not appropriate to be an effective chemical index for rejuvenation efficiency evaluation.

4.2.3. *Branched aliphatic index BAII-based rejuvenation percentage BAIIR*

The BAIIR values of various rejuvenated binders are presented in Fig. 6 to assess the possibility of using the BAII parameter to evaluate and distinguish the rejuvenation efficiency of different rejuvenation cases effectively. The BAIIR values of rejuvenated bitumen have a linear relationship with the rejuvenator concentration, independent on the rejuvenator type and the aging degree of bitumen. The positive BAIIR values for EO, NO, and AO suggest that these three petroleum-based rejuvenators can partially rejuvenate the BAII value of aged bitumen. Conversely, the bio-oil rejuvenator's involvement results in negative BAIIR values, indicating that it doesn't effectively restore the BAII parameter. Consequently, the BAII parameter, serving as an evaluation indicator for rejuvenation efficiency, is not suitable for all types of rejuvenators. However, the difference in BAIIR values of EO, NO, and AO rejuvenated binders are significant, implying that the rejuvenation efficiency of these petroleum-based rejuvenators can be identified using BAII parameter. Regardless of rejuvenator dosage and aging stage, the BAIIR values of rejuvenated bitumen with three petroleum-based

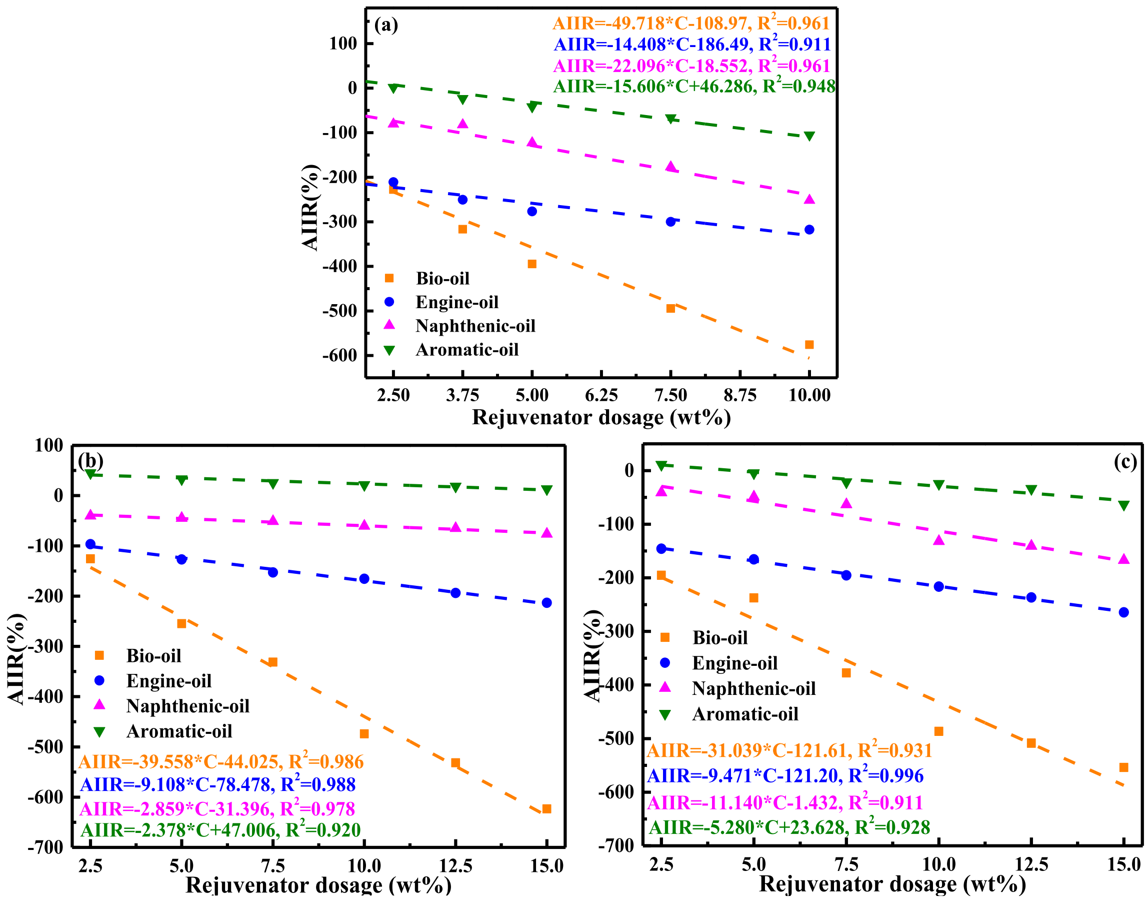

Fig. 5. AII-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 bitumen.

rejuvenators are NORB > EORB > AORB. The scope of BAIIR values are much higher than rejuvenation percentages based on rheological properties. Further, the aging status of bitumen has a distinct impact on both BAIIR values and its sensitivity level to rejuvenator dosage. For three petroleum-based rejuvenators, a high aging degree of bitumen reduces the BAIIR values and weakens the influence of rejuvenator dosage. The rejuvenator concentration has the greatest effect on the BAIIR values of NORB, followed by EORB and AORB binders.

4.2.4. *Long chains index LCI-based rejuvenation percentage LCIR*

The LCIR values of various rejuvenators are displayed in Fig. 7. The linear relationship between the LCIR parameter and rejuvenator concentration is detected. The addition of bio-oil and engine-oil results in negative LCIR values of BORB and EORB, while the positive LCIR values of NORB and AORB validate the rejuvenation potential of naphthenic-oil and aromatic-oil rejuvenators. Irrespective of the level of aging and the quantity of rejuvenator used, the LCIR values for AORB consistently exceed those of NORB. In addition, EORB exhibits a lower LCIR parameter than BORB. This implies that the rejuvenation effectiveness of aromatic oil is greater than that of naphthenic oil. At the same time, engine-oil involvement introduces more long-chain molecules to aged bitumen than bio-oil rejuvenator. Additionally, the sensitivity of the LCIR parameter to rejuvenator dosage is strongly dependent on the rejuvenator type, and a high aging degree would weaken the sensitivity, especially from LAB20 to LAB40. Hence, the LCI parameter is a suitable tool for assessing and differentiating rejuvenation effectiveness of naphthenic-oil and aromatic-oil rejuvenators, but it may not be applicable to cases involving bio-oil and engine-oil rejuvenators.

4.2.5. *Carbonyl index CI-based rejuvenation percentage CIR*

The CIR-C curves of various rejuvenated bitumen are drawn in Fig. 8. The CIR values for all rejuvenated binders are positive but remain below 100%. This suggests that the rejuvenation conditions employed in this study are insufficient to completely restore the chemical properties of aged bitumen to those of fresh bitumen. The CIR values of all rejuvenated binders present a linear intensifying tendency as the rejuvenator content enlarges. This mechanism primarily arises from the amalgamation of functional groups in the rejuvenators, which results in the dilution of the CI concentration throughout the entire rejuvenated bitumen. Regardless of rejuvenator dosage and aging level, the CIR values of bio-oil rejuvenated bitumen are the highest, indicating it presents the greatest rejuvenation efficiency on the CI parameter of aged bitumen. Moreover, the CIR parameter exhibits the greatest sensitivity to an increased dosage of rejuvenator in the BORB binder. In general, the CI parameter proves effective in evaluating and distinguishing the rejuvenation effectiveness of various rejuvenators.

4.2.6. *Sulfoxide index SI-based rejuvenation percentage SIR*

The SIR values of all rejuvenated bitumen systems are calculated following Eq.11 and drawn in Fig. 9. The SIR values of all rejuvenated bitumen are positive, indicating that all rejuvenators exhibit restoration functions on the SI parameter of aged bitumen. However, these SIR values fall below 100%, indicating that the rejuvenation conditions chosen in this study do not entirely restore the SI parameter of aged bitumen to the level of a pristine binder. Moreover, the SIR parameter of the AORB binder is much lower than the other three kinds of rejuvenated binders. Hence, the aromatic-oil rejuvenator shows the weakest rejuvenation efficiency on the SI parameter of aged bitumen.

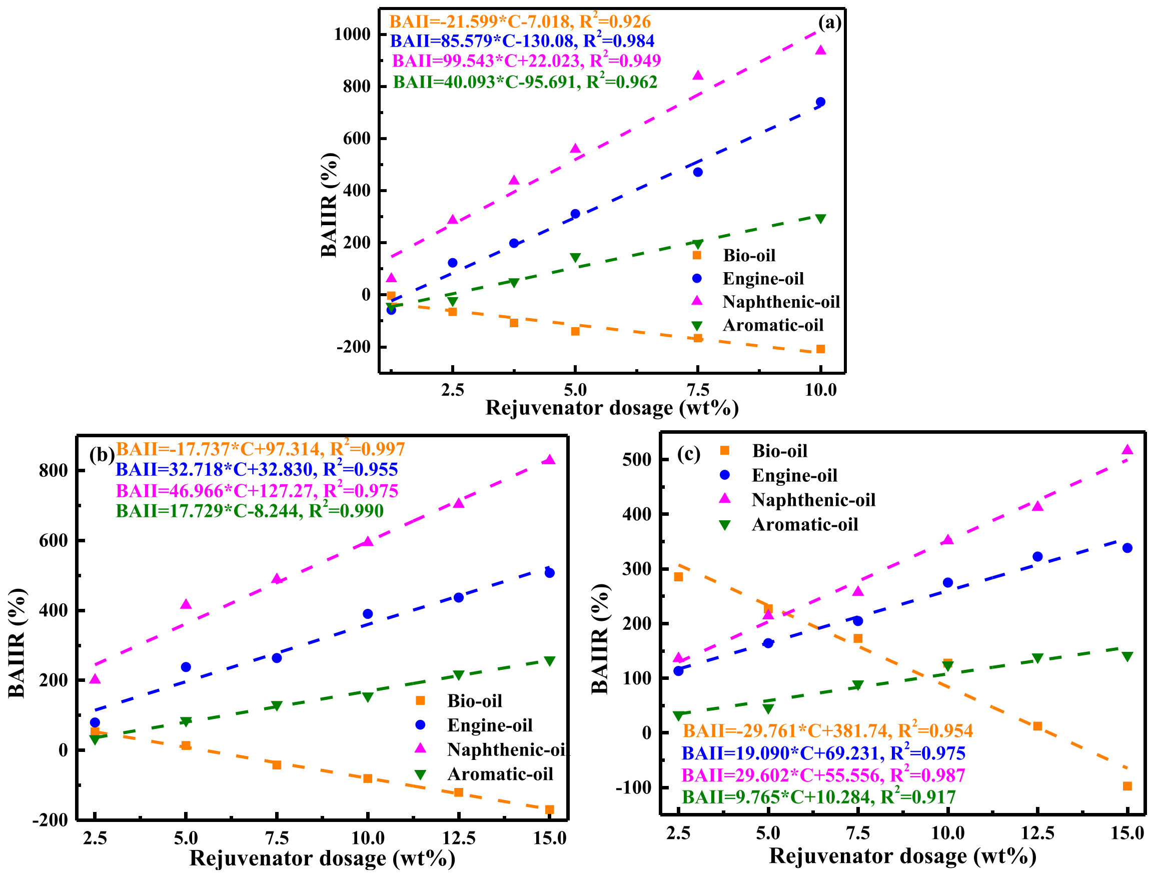

Fig. 6. BAII-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 bitumen.

The magnitude of SIR values of BO, EO, and NO rejuvenated binders strongly depends on the rejuvenator dosage and aging level of bitumen, especially the NORB binder. The difference in SIR parameters of BORB and EORB samples is also limited. Distinguishing the rejuvenation efficiency of bio-oil, engine-oil, and naphthenic-oil rejuvenators is difficult using the SIR parameter. However, it proves valuable in understanding the impact of the aromatic-oil rejuvenator. Interestingly, the finding is the opposite of the CIR case. Thus, it is recommended to amalgamate the SIR and CIR parameters for fully evaluating the rejuvenation efficiency of various rejuvenators on the chemical properties of aged bitumen with influence factors of rejuvenator type/dosage and aging grade of bitumen.

4.2.7. Further discussion on vital chemical indices for rejuvenation efficiency evaluation

Fig. 10 illustrates a screening program for effective and critical evaluation indicators. Initially, it's essential to identify potential indicators for evaluating rejuvenation efficiency and corroborate their effectiveness through experimental results. Subsequently, the viability of these chosen potential indicators for rejuvenation should be verified, and any unsuitable indices, characterized by negative rejuvenation percentages, should be eliminated. To develop and optimize the rejuvenator components, the chosen evaluation indices in Step iii must be sensitive to rejuvenator type. The role of rejuvenator dosage will also be considered when selecting indicators. Any indicators that fail to assess the effects of both rejuvenator type and dosage on rejuvenation efficiency will be eliminated. In Step v, the sensitivity of potential evaluation indicators to bitumen aging levels will be examined. The goal is to ensure that the influence of rejuvenator type and dosage on rejuvenation percentages remains consistent across different aging stages of bitumen, while the effect of aging level should be evident. Indicators that don't meet these requirements will be categorized as sensitive indices. In Step vi, the remaining indicators will be assessed in terms of their range of rejuvenation percentages. Since the primary objective of adding rejuvenators is typically to restore aged bitumen properties to those of virgin bitumen, the expected range of rejuvenation percentages is generally 0–100%, or slightly more in cases of high rejuvenation efficiency. This step will filter out any indicators with abnormal rejuvenation percentage results, ultimately identifying effective evaluation indicators.

Table 5 lists the sensitivity levels of all chemical indices to different influence factors for estimating their potential to be effective rejuvenation efficiency evaluation indicators. While the AII, BAII, and LCI parameters exhibit good sensitivity to rejuvenator type, dosage, and bitumen aging, they are eliminated due to their inability to demonstrate the potential for recovery. The other three chemical indices (AI, CI, and SI) of aged bitumen can be regenerated by incorporating all rejuvenators. Nevertheless, some limitations are also observed. For example, the magnitude of rejuvenation efficiency on AI and SI parameters of rejuvenated bitumen is affected by the aging status of bitumen, especially for BORB, EORB, and NORB. Moreover, the impacts of adding BO, EO, and NO rejuvenators with the same rejuvenator concentration on SI-based rejuvenation percentages are similar. As a result, the CI parameter is better suited as a crucial chemical indicator for assessing the rejuvenation efficiency in various rejuvenation systems compared to the AI and SI parameters. Moreover, these potentially valuable chemical indices will be linked to essential rheological parameters, along with their associated rejuvenation percentages. This will facilitate the exploration of chemo-rheological relationships in rejuvenated bitumen, considering varying rejuvenator type, dosage, and bitumen aging level.

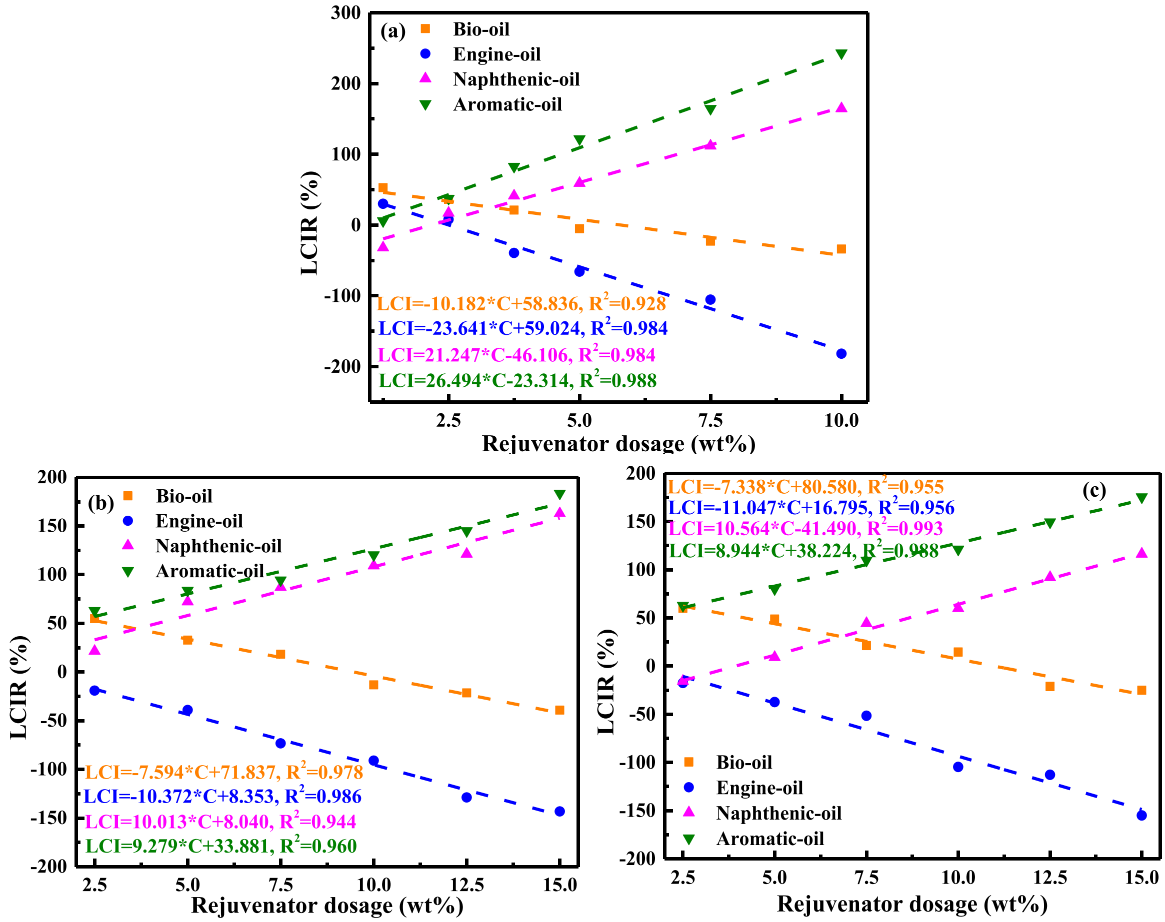

Fig. 7. LCI-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 bitumen.

- **XXO[1]:** The aging level shows a great influence on the magnitude of AIR for most rejuvenated binders (BORB, EORB, and NORB), except the AORB.
- **XOO[2]:** Most rejuvenators (engine-oil, naphthenic-oil, and aromatic-oil) in this study exhibit rejuvenation effects on the BAII value of aged bitumen towards virgin bitumen level, while the bio-oil shows a converse result.
- **XO[3]:** The LCI parameter can be adopted to evaluate the rejuvenation efficiency of naphthenic-oil and aromatic-oil rejuvenators while adding bio-oil and engine-oil fails to restore the LCI value of aged bitumen.
- **Θ[4]:** The magnitude for CIR values of BORB, EORB, and NORB binders is not dependent on the aging level of bitumen, while the CIR parameter of AORB varies a lot as a function of aging level.
- **XXO[5]:** The SI values of bio-oil, engine-oil, and naphthenic-oil rejuvenated binders are very similar, while only the SI parameter of aromatic-oil rejuvenated bitumen can be distinguished.
- **XXO[6]:** With the increase in bitumen's aging level, the rejuvenation efficiency of aromatic-oil on the SI parameter of aged bitumen can be recognized consistently. However, the aging degree of bitumen significantly influences the magnitude of SIR values of BORB, EORB, and NORB binders.

4.3. Rheological properties of aged bitumen

The master curves and rheological indices of virgin and aged binders are displayed in Fig. 11. As expected, aged bitumen with a higher aging level shows larger G* and lower δ values, especially at a low-frequency region, related to the increased stiffness and elastic component. Moreover, the Log(G*) and δ parameters of bitumen exhibit a linear increasing and decreasing trend as the long-term aging time prolongs. Based on the absolute slope values, the sensitivity of δ to the aging level is more significant than the Log(G*) index. Thus, the G* and δ values of aged binders with variable aging degrees can be anticipated with the correlation equations. The logarithmic values of crossover modulus G_c and crossover frequency f_c decline linearly as the aging time extends. Similarly, the aging time significantly influences the log(f_c) more than the G_c value of bitumen.

4.4. Rheological properties of rejuvenated bitumen

4.4.1. G and δ at 60* ℃

The G* and δ parameters of rejuvenated binders at 60 ℃ and 10 rad/s are selected for quantitatively assessing and distinguishing rejuvenation efficacy of various rejuvenators on rheological properties. The test results and corresponding rejuvenation percentages (G*R and δR) are displayed in Figs. 12 and 13. It is observed that the log(G*) values of rejuvenated bitumen tend to decrease linearly as the rejuvenator dosage rises. With the same rejuvenator content, the G* magnitude of rejuvenated binders is BORB < EORB < NORB < AORB. Among the rejuvenators, the bio-oil has the most pronounced impact on reducing G* in aged bitumen, followed by engine-oil and naphthenic-oil, with the aromatic-oil demonstrating the lowest rejuvenation efficiency for the G* parameter. Meanwhile, the sensitivity order of G* to rejuvenator dosage follows BO > EO > NO > AO, independent on bitumen aging degree. Positive exponential correlations between the G*R parameters of rejuvenated binders with the rejuvenator dosage show a ranking of BORB > EORB > NORB > AORB, regardless of the aging degree of

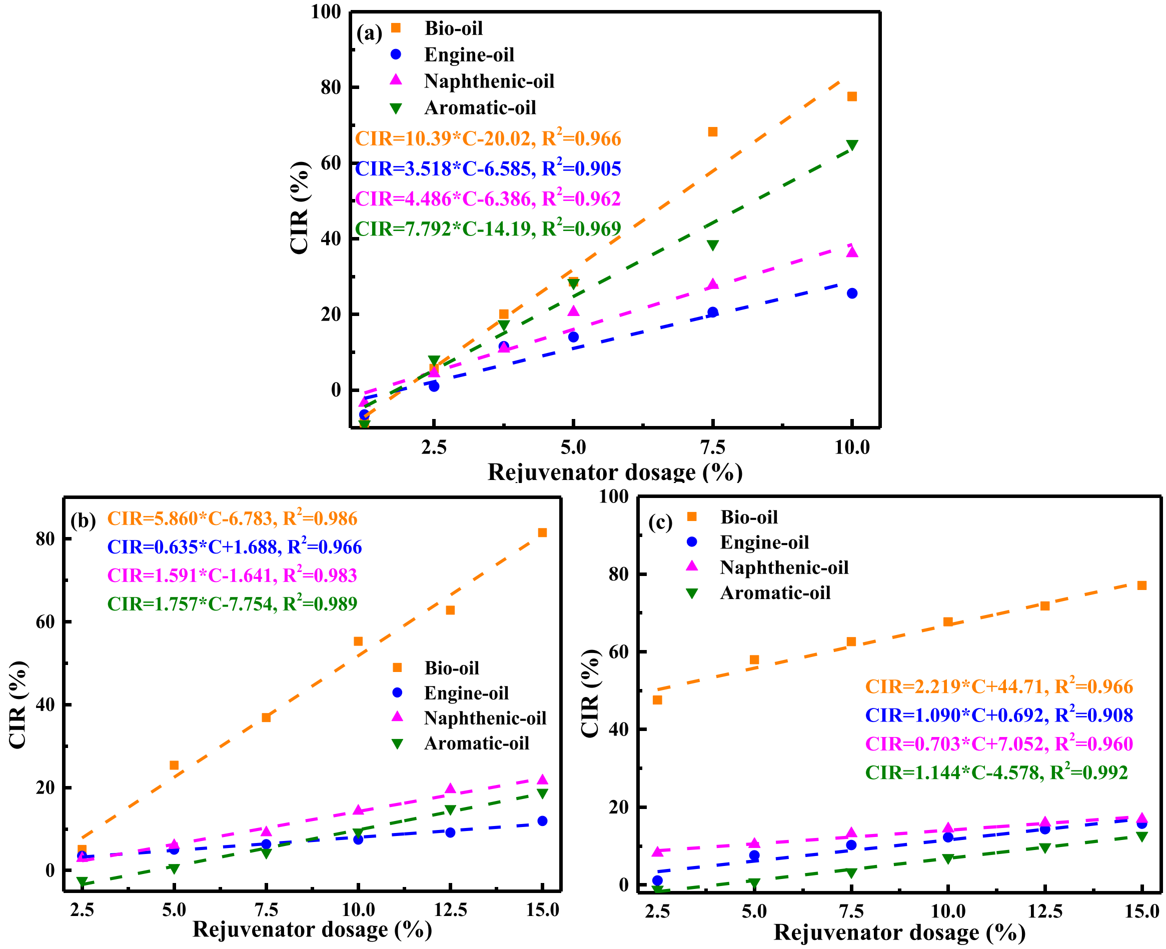

Fig. 8. CI-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 bitumen.

bitumen. As the level of aging intensifies and the amount of rejuvenator increases, the distinction in GR values among BORB, EORB, and NORB binders diminishes, while AORB consistently maintains a lower G value compared to the rest. Overall, the effects of rejuvenator type/dosage and the aging degree of bitumen on the efficiency of rejuvenation can be evaluated and distinguished effectively with the G* parameter.

Similarly, the phase angle δ values of rejuvenated binders tend to increase linearly as a function of rejuvenator content. Moreover, the δ values of bio-oil and aromatic-oil rejuvenated binders are larger than the engine-oil and naphthenic-oil rejuvenated bitumen. The presence of polar functional groups in BO (esters) and AO (aromatic rings) rejuvenators may facilitate the reestablishment of a colloidal structure by breaking apart and dispersing the asphaltene clusters within aged bitumen [27,29]. Additionally, the higher concentration of low-viscosity and lightweight components in BO enhances the dispersion and fluidity of bitumen micellar structure compared to the AO rejuvenator. On the other hand, the engine-oil and naphthenic-oil rejuvenators exhibit a less and similar enlargement function on δ value. However, the order of δ value and its sensitivity to rejuvenator dosage of rejuvenated bitumen are distinctly affected by the aging level of bitumen and rejuvenator dosage. For instance, the G* values of BORB are higher than AORB with LAB20 and LAB40, but an intersection point is observed in the LAB80 case. Meanwhile, the aging degree of bitumen leads to the sorting difference in δ values of EORB and NORB binders. All δR values of rejuvenated bitumen are lower than 100%, further illustrating the limited rejuvenation efficiency of rejuvenators on increasing δ parameter of aged bitumen. Like G*R, the δR values of rejuvenated bitumen show an exponentially growing trend versus rejuvenator content. Nonetheless, there is no consensus regarding the definitive ranking of rejuvenators in terms of their efficiency concerning the δ parameter. This lack of consensus stems from the significant influence of both the rejuvenator concentration and the level of bitumen aging. Consequently, the δ parameter is not recommended as a primary evaluation indicator for estimating and comparing the rejuvenation efficiency of various rejuvenators in relation to the rheological properties of aged bitumen.

4.4.2. G_c and f_c

The rheological crossover indices (G_c and f_c) of rejuvenated bitumen are reflected in Figs. 14 and 15. With the rejuvenator dosage increases, the G_c values of aromatic-oil rejuvenated bitumen enlarge linearly, while the other three rejuvenated binders (BORB, EORB, and NORB) show an opposite trend. For this reason, the G_cR values of AORB and the other three rejuvenated binders are positive and negative, respectively. Adding an aromatic-oil rejuvenator can restore the G_c parameter of aged bitumen, while the bio-oil, engine-oil, and naphthenic-oil have no rejuvenation effect. Additionally, the G_cR parameter of EORB is lower than BORB and NORB. The aging degree of bitumen significantly influences the sensitivity levels of G_c and G_cR parameters of rejuvenated bitumen to the rejuvenator dosage. As the aging level deepens, the absolute slope values of G_cR-C curves reduce gradually, showing reduced influence of rejuvenator dosage on the G_cR parameter. Regardless of the aging degree, the absolute slope value of AORB is the highest, implying that the impact of AO dosage on the G_cR is the largest. However, the G_cR value of AORB binder is lower than 30% and declines as the aging level of bitumen increases. Among the rejuvenators, only the aromatic-oil rejuvenator exhibits a partial restoration of the G_c parameter, albeit with limited efficiency. Consequently, the G_c index proves inadequate for evaluating the rejuvenation effectiveness of all rejuvenators.

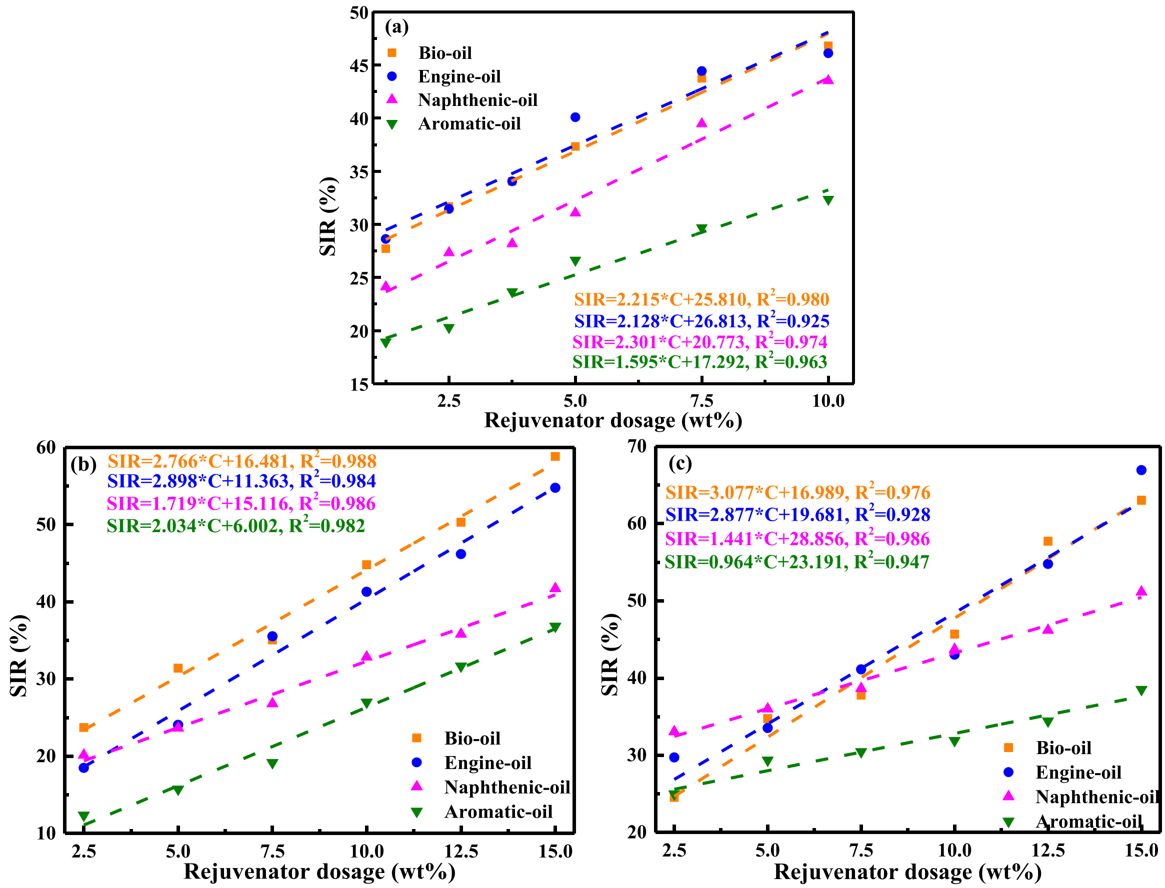

Fig. 9. SI-based rejuvenation percentages of various rejuvenated bitumen. (a) LAB20 bitumen; (b) LAB40 bitumen; (c) LAB80 aged bitumen.

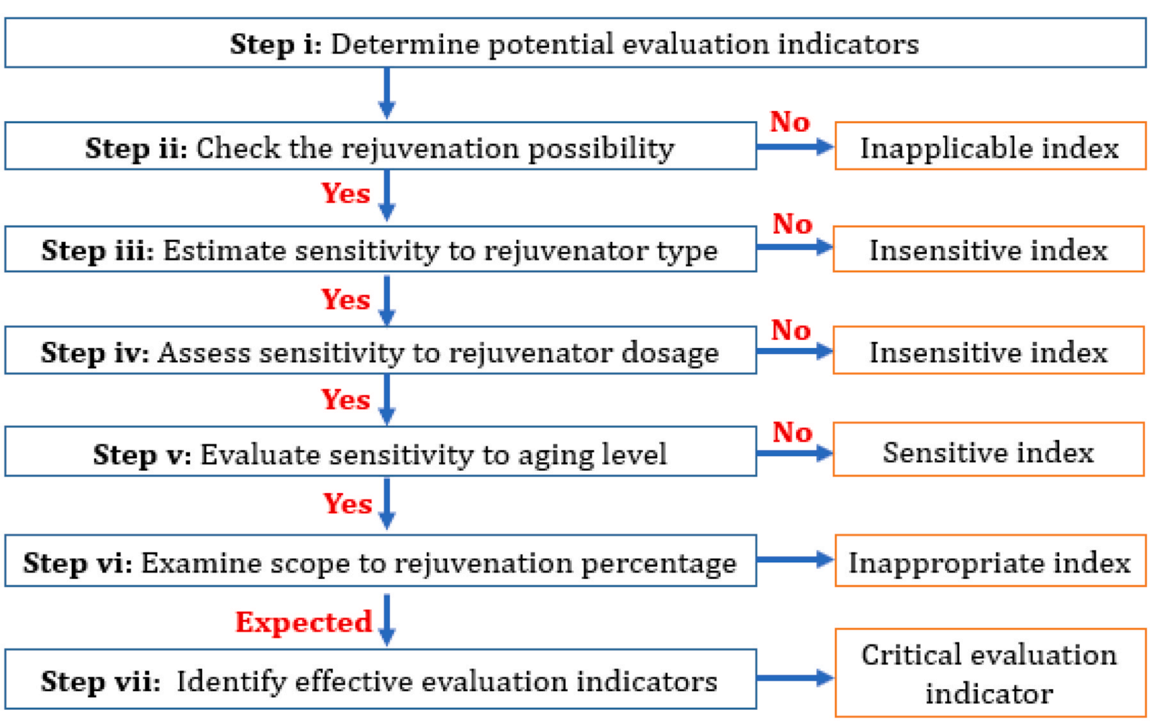

Fig. 10. The selection program for critical chemical and rheological indicators.

Table 5
Analysis of critical chemical indicators for rejuvenation efficiency evaluation.

Potential evaluation indicators	AI	AII	BAII	LCI	CI	SI
Recovery possibility	O	X	XOO[2]	XO[3]	O	O
Sensitivity to rejuvenator type	O	O	O	O	O	XXO[5]
Sensitivity to rejuvenator dosage	O	O	O	O	O	O
Sensitivity to aging level	XXO[1]	O	O	O	O[4]	XXO[6]
Rejuvenation percentage scope (%)	0-400	-700-0	-200-0 (BO) 0-1000 (Others)	-200-50 (BO, EO) 0-250 (NO, AO)	0-100	0-70

The crossover frequency f_C values of all rejuvenated binders enlarge linearly with the increase in rejuvenator dosage. It implies that adding rejuvenators can somewhat restore the fc value of aged bitumen. Based on this, the f_C index is more appropriate to assess the rejuvenation efficiency of various rejuvenators on rheological performance than G_c index. Moreover, the f_c values of BORB and AORB are higher than the EORB and NORB binders. The fc-C curves are similar to the δ-C curves. For instance, the f_c values of EORB and AORB binders in LAB20-aged bitumen are very near, and there is an intersection point between the f_c-C curves of BORB and AORB with LAB80-aged bitumen. Similar to δR, the f_cR values of all rejuvenated binders behave an exponentially increasing trend as the increase in rejuvenator content. Moreover, the increasing rate of the f_cR parameter gradually enlarges versus increased rejuvenator dosage. This indicates that the contrast in fcR values among different rejuvenated bitumen samples becomes more pronounced at higher rejuvenator concentrations. However, it proves challenging to distinguish the fcR values of various rejuvenated bitumen samples when a low dosage of rejuvenator is employed, particularly in the case of EORB, NORB, and AORB with LAB20 and LAB40 aged bitumen. When the aging level reaches LAB80, the aromatic-oil exhibits the highest rejuvenation efficiency on the f_c parameter of aged bitumen, followed by the BO, EO, and NO rejuvenators.

4.4.3. A_{ML}, A_{MH}, and A_{MT} parameters of master curves

As mentioned before, the G* parameter is an effective indicator but G* -based rejuvenation percentages depend on the corresponding frequencies (or temperatures). To eliminate this influence, the rejuvenation efficiency based on the whole G* master curve is estimated based on A_{ML}, A_{MH}, and A_{MT}, considering its sensitivity to frequency. Fig. 16 shows the A_{ML}, A_{MH}, and A_{MT} values of virgin and aged bitumen. As the aging level deepens, the G* and all area parameters enlarge dramatically due to the increment in stiffness. It is noticed that the A_{ML} of all bitumen is higher than the A_{MH}, related to the higher modulus in low temperatures (high frequencies). In addition, the influence of aging level on A_{MH} is more significant than A_{ML}.

The A_{ML} and its corresponding rejuvenation percentage A_{ML}R values of rejuvenated binders are shown in Fig. 17. As expected, adding all rejuvenators can reduce the A_{ML} values of aged bitumen, decreasing linearly as an increment in rejuvenator dosage. The AORB exhibits the largest A_{ML} values, followed by NORB and EORB, and BORB shows the lowest A_{ML} value. Meanwhile, the A_{ML} values of engine-oil and naphthenic-oil rejuvenated bitumen are similar, which agrees well with the G* result. Regarding the A_{ML}R parameter, the magnitude of rejuvenated bitumen is BORB > EORB > NORB > AORB. Moreover, the A_{ML}R tends to increase linearly with the rejuvenator content rising. The aging degree of bitumen significantly influences the A_{ML}R values of rejuvenated bitumen, within the scope of 0–1200%, 0–600%, and 0–250% when the aged bitumen is LAB20, LAB40 and LAB80, respectively. Interestingly, the A_{ML}R values of rejuvenated bitumen are much larger than the G*R parameter (0–135%, 0–100%, and 0–100%). Fig. 19 shows the rejuvenation efficiency of rejuvenators on the G* values at high frequencies (> 100 Hz)—that's why the calculated A_{ML}R values of rejuvenated bitumen are extremely high.

Fig. 18 illustrates the A_{MH} and A_{MH}R values of rejuvenated binders. The increment in rejuvenator dosage leads to a linear decrease of A_{MH} parameters of all rejuvenated bitumen. It shows that all rejuvenators exhibit the rejuvenation function of restoring aged bitumen's AMH value. However, the rejuvenation level is affected by the rejuvenator type. The aromatic-oil rejuvenated bitumen has the largest A_{MH} parameter, followed by NORB and EORB, while the BORB shows the lowest A_{MH} value. It agrees well with the G* results, and the ranking of A_{MH} sensitivity to rejuvenator dosage for rejuvenated binders is BORB > EORB > NORB > AORB. As the aging level deepens, the A_{MH} values of rejuvenated binders enlarge, while the effect of rejuvenator dosage on A_{MH} value is weakened. The rejuvenation percentage A_{MH}R has a linear increasing trend as the rejuvenator content rises. Moreover, the order of A_{MH}R values for four rejuvenators follows bio-oil > engine-oil > naphthenic-oil > aromatic-oil, independent of the aging degree of bitumen. As expected, the A_{MH}R values of rejuvenated binders reduce when the aging level changes from LAB20 to LAB40 and LAB80. Regardless of the aging level, the A_{MH}R sensitivity to the rejuvenator dosage of BORB is the strongest, followed by the EORB and NORB, while the AORB shows the least sensitivity. Further, the A_{MH}R values of rejuvenated binders with LAB20, LAB40, and LAB80 aged bitumen are 0–150%, 0–130%, and 0–85%, respectively. The range of A_{MH}R is similar to that of G*R, suggesting that the A_{MH} index can also serve as a valuable indicator for assessing and distinguishing the rejuvenation effectiveness in various rejuvenated bitumen blends.

The total area A_{MT} and its rejuvenation percentage A_{MT}R values are plotted in Fig. 19. The variation trend of A_{MT} and A_{MT}R parameters versus rejuvenator dosage is similar to A_{ML} (A_{ML}R) and A_{MH} (A_{MH}R). The A_{MT} values and their sensitivity to rejuvenator dosage (absolute slope values of correlation equations) are the sum of A_{ML} and A_{MH} values, as well as their sensitivities. Meanwhile, the A_{MT}R values of rejuvenated bitumen are between A_{ML}R and A_{MH}R values but closer to the A_{MH}R parameter. In addition, the A_{MT}R sensitivity to rejuvenator content is lower than A_{ML}R but higher than A_{MH}R. It should be noted that the difference in all A and AR values of engine-oil and naphthenic-oil rejuvenated binders is limited but distinguished. Overall, these three rheological indices (A_{ML}, A_{MH}, A_{MT}) can evaluate and differentiate the rejuvenation efficiency of various rejuvenators to G* master curves of aged bitumen. Nevertheless, there is a noticeable difference in A_{ML}R, A_{MH}R, and A_{MT}R values. The unexpectedly large and wide scope of the A_{ML}R parameter (0–1200%) indicates that the A_{ML} index fails to reasonably assess the rejuvenation efficiency on G* curves of aged bitumen. Conversely, the A_{MH}R and A_{MT}R ranges are fairly comparable, but the A_{MH}R value is smaller than that of A_{MT}R due to the inclusion of the A_{ML}R term in the latter. Given the marked resemblance between the GR and A_{MH}R profiles, it is evident that the A_{MH} parameter plays a crucial role in evaluating the rejuvenation efficiency of various rejuvenators in shaping the G master curves of aged bitumen.

4.4.4. Further discussion on critical rheological indicators

In this section, the possibility of these rheological parameters as the critical indices for rejuvenation efficiency evaluation is discussed using a selection program shown in Fig. 10. The results are summarized in

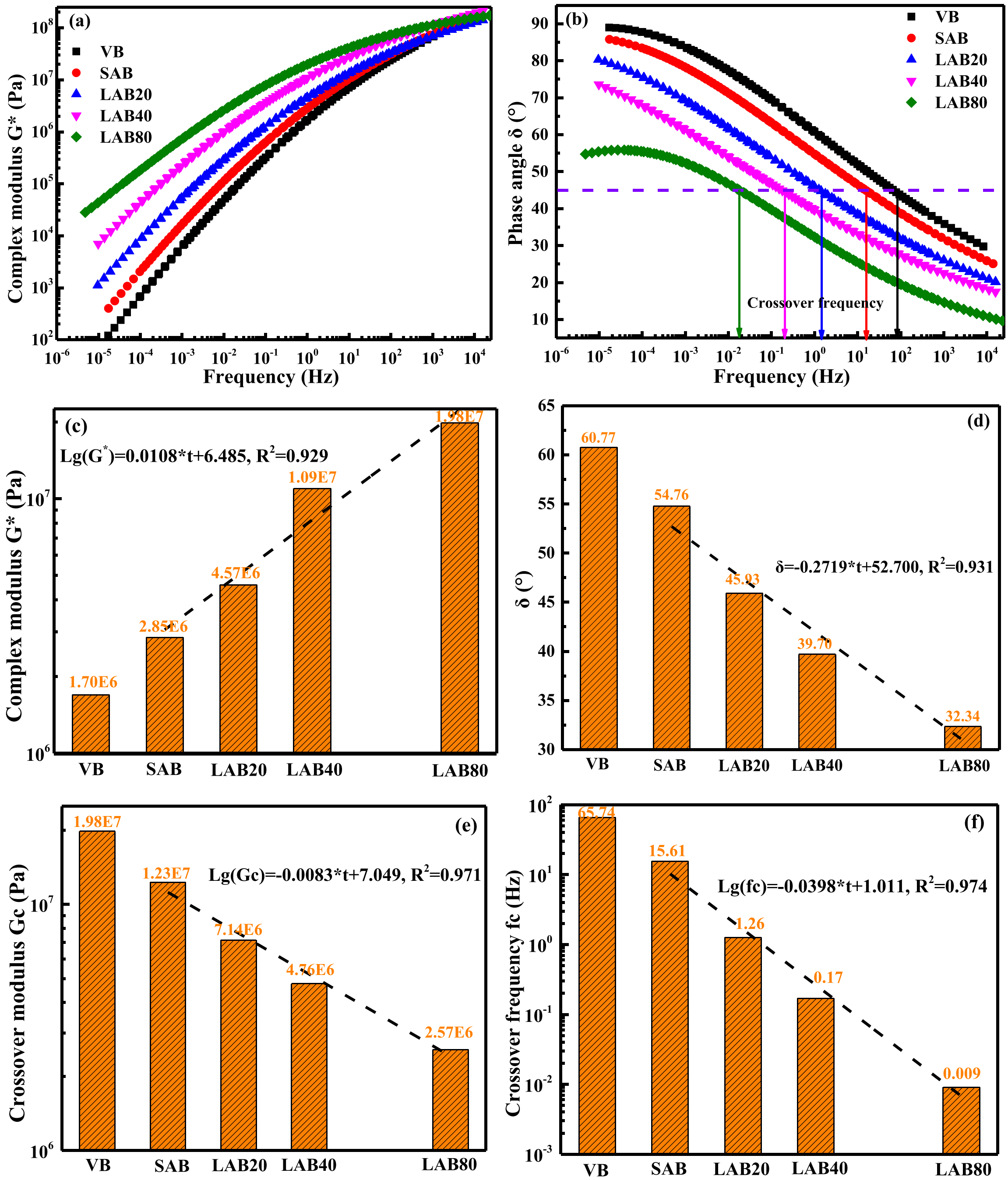

Fig. 11. Rheological properties of virgin and aged bitumen.

Table 6. It is noted that the "O" and "X" mean "Yes" and "No". The explanations for other special terms (ʘ, OXX, XXO, and XO) are attached below the table.

- **ʘ[1]:** The sensitivity of G*R to rejuvenator dosage reduces gradually as the increment in rejuvenator content.
- **OXX[2]:** The influence of bio-oil on restoring the δ value of aged bitumen can be distinguished from the other three rejuvenators. However, the aging level of bitumen and rejuvenator dosage affect the magnitude of δR values for EORB, NORB, and AORB.
- **XXO[3]:** The addition of bio-oil, engine-oil, and naphthenic-oil rejuvenators fail to rehabilitate the G_c value of aged bitumen, while the aromatic-oil shows the rejuvenating effect.
- **XO[4]:** For all aging levels of bitumen, the f_cR values of bio-oil and aromatic-oil rejuvenated bitumen are larger than engine-oil and naphthenic-oil rejuvenated binders. However, the order of the f_cR parameter of BORB and AORB or EORB and NORB distinctly depends on the aging degree of bitumen.

Following a thorough analysis and comparison of the data, it is evident that the δ and G_c parameters fall short in their capacity to effectively gauge rejuvenation efficiency. The δ parameter is insensitive to rejuvenator type, while the G_c parameter is unable to signify recovery potential. On the other hand, all the criteria for sensitivity are met by the area indicators. However, the A_{MH}R range closely aligns with the GR parameter, with unexpectedly high A_{ML}R values. Consequently, the A_{MH} parameter emerges as the preferred choice for evaluating the

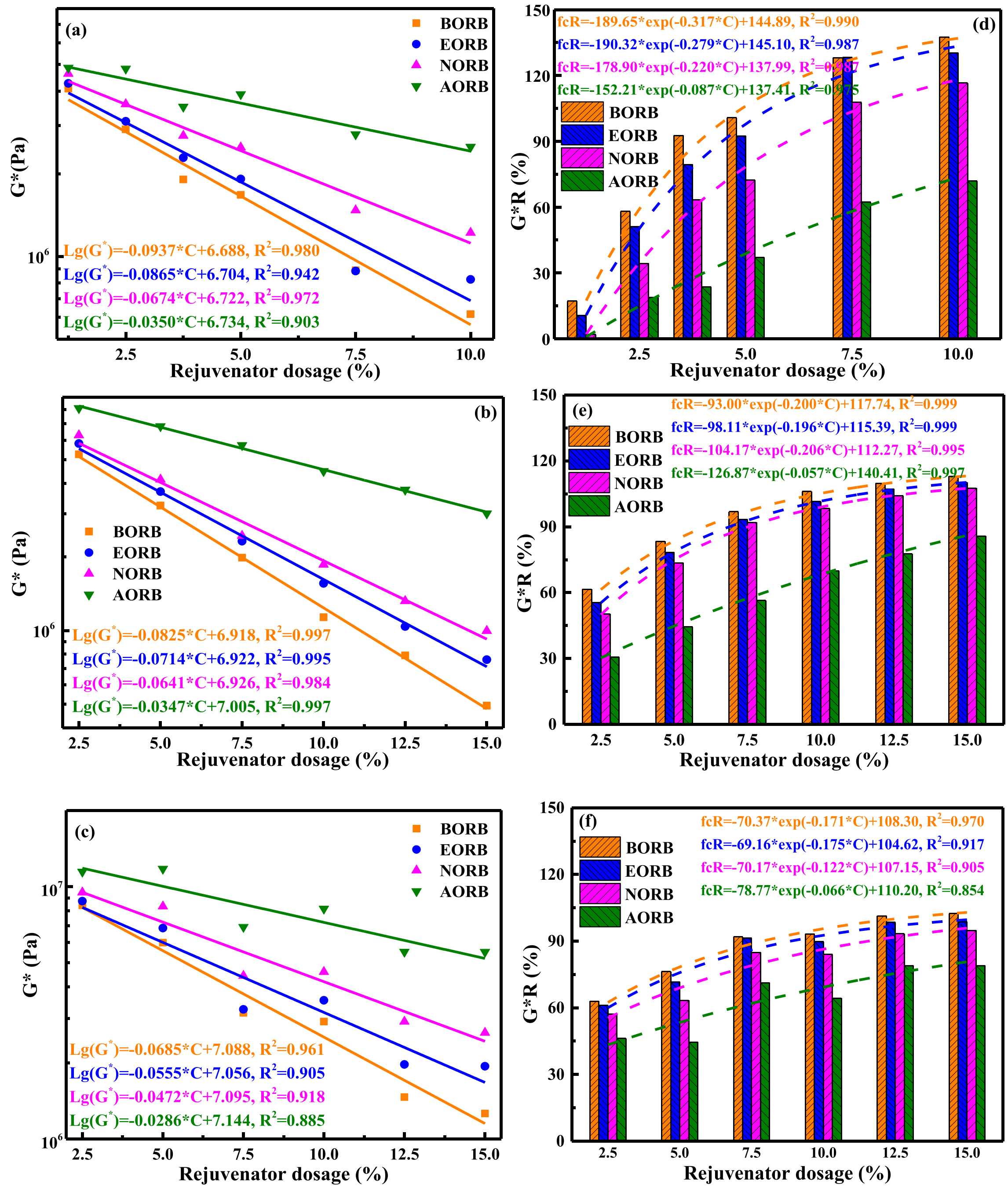

Fig. 12. The G* and G*R parameters of various rejuvenated bitumen.

rejuvenation efficiency of various rejuvenators in shaping the G master curves of aged binders. In summary, three rheological indices, G* , f_c, and A_{MH}, are recommended as essential indicators for assessing rejuvenation efficiency in terms of rheological performance.

4.5. Explanation and discussion of the underlying mechanism at the nanoscale

From the perspectives of both chemical and rheological terms, there is a considerable difference in the rejuvenation efficiency of various rejuvenators. The ranking of rejuvenation efficiency for four rejuvenators on different chemical and rheological indices is drawn in Fig. 20(a). Positive values indicate positive rejuvenation efficiency, while negative values correspond to negative rejuvenation efficiency. Additionally, the numbers "1, 2, 3, 4" represent the rejuvenation levels, where "1" signifies the lowest, and "4" denotes the highest level of rejuvenation efficiency. Bio-oil rejuvenator mostly exhibits the best rejuvenation effectiveness on chemo-rheological properties of aged bitumen, including the AIR, CIR, SIR, G*R, δR, f_cR, and AR. In addition, the aromatic-oil rejuvenator shows the lowest rejuvenation efficiency on AI, CI, SI, G* , and A, although it presents a high rejuvenation potential on BAI, LCI, δ, and f_c parameters. Regarding most of the chemo-rheological indices (AI, AII, CI, SI, G*, δ, f_c, and A), the rejuvenation percentages for engine-oil and naphthenic-oil rejuvenators fall between those of bio-oil and aromatic-oil, with engine-oil demonstrating the most substantial rejuvenation effect. Crucially, across these vital chemical and rheological indicators, the order of rejuvenation efficiency among the four rejuvenators is BO > EO > NO > AO.

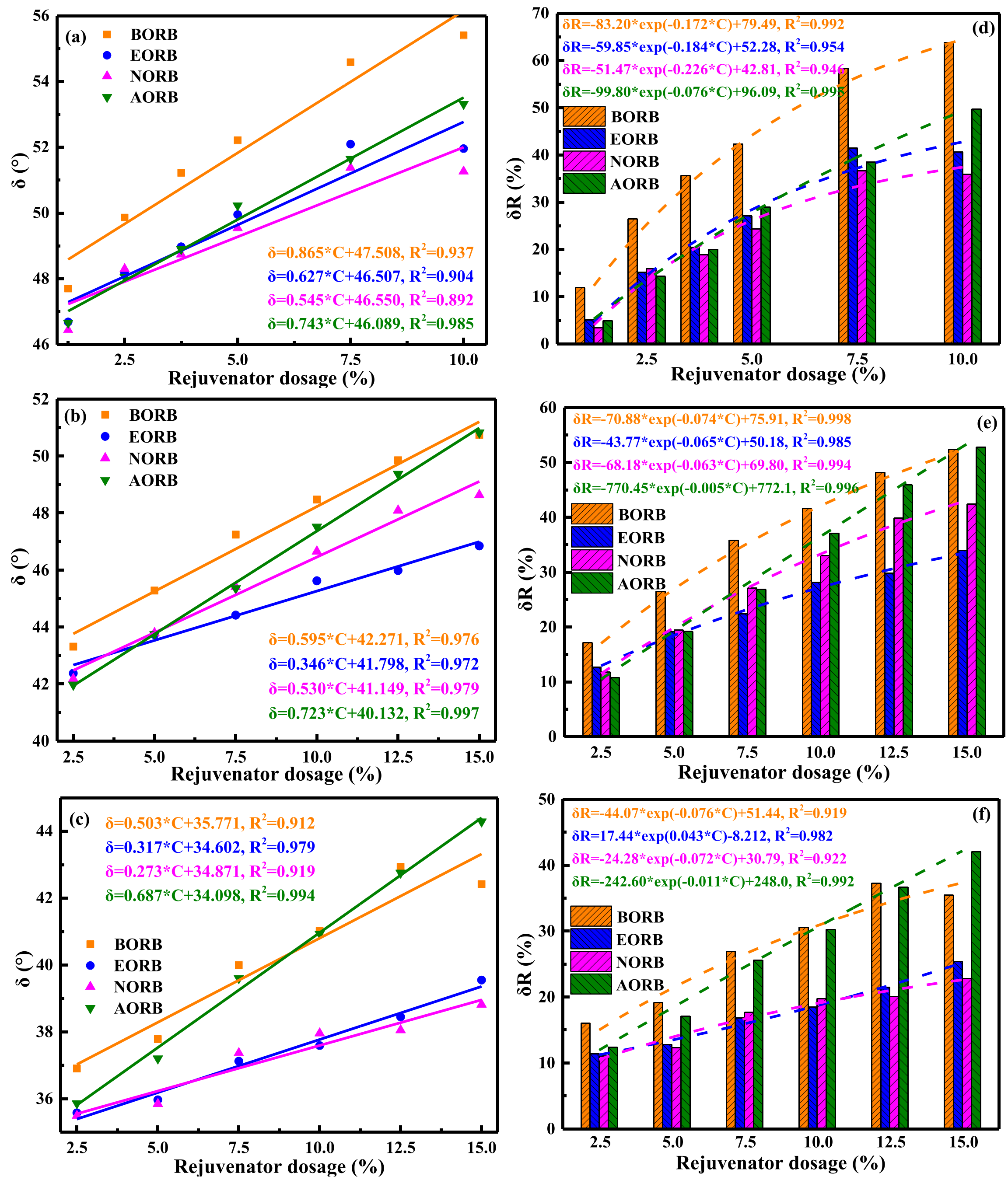

Fig. 13. The δ and δR parameters of various rejuvenated bitumen.

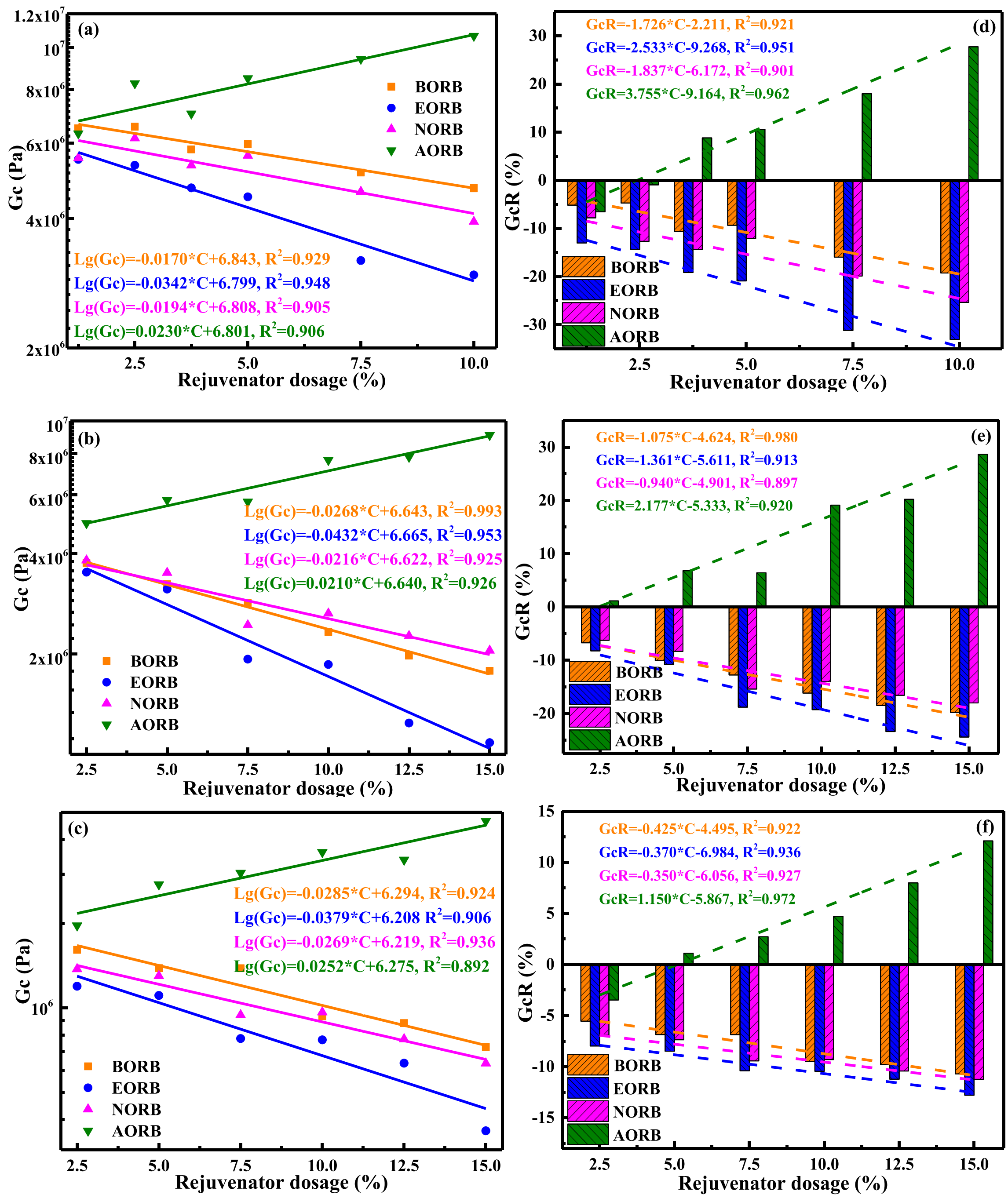

Fig. 14. The G_c and G_cR parameters of various rejuvenated bitumen.

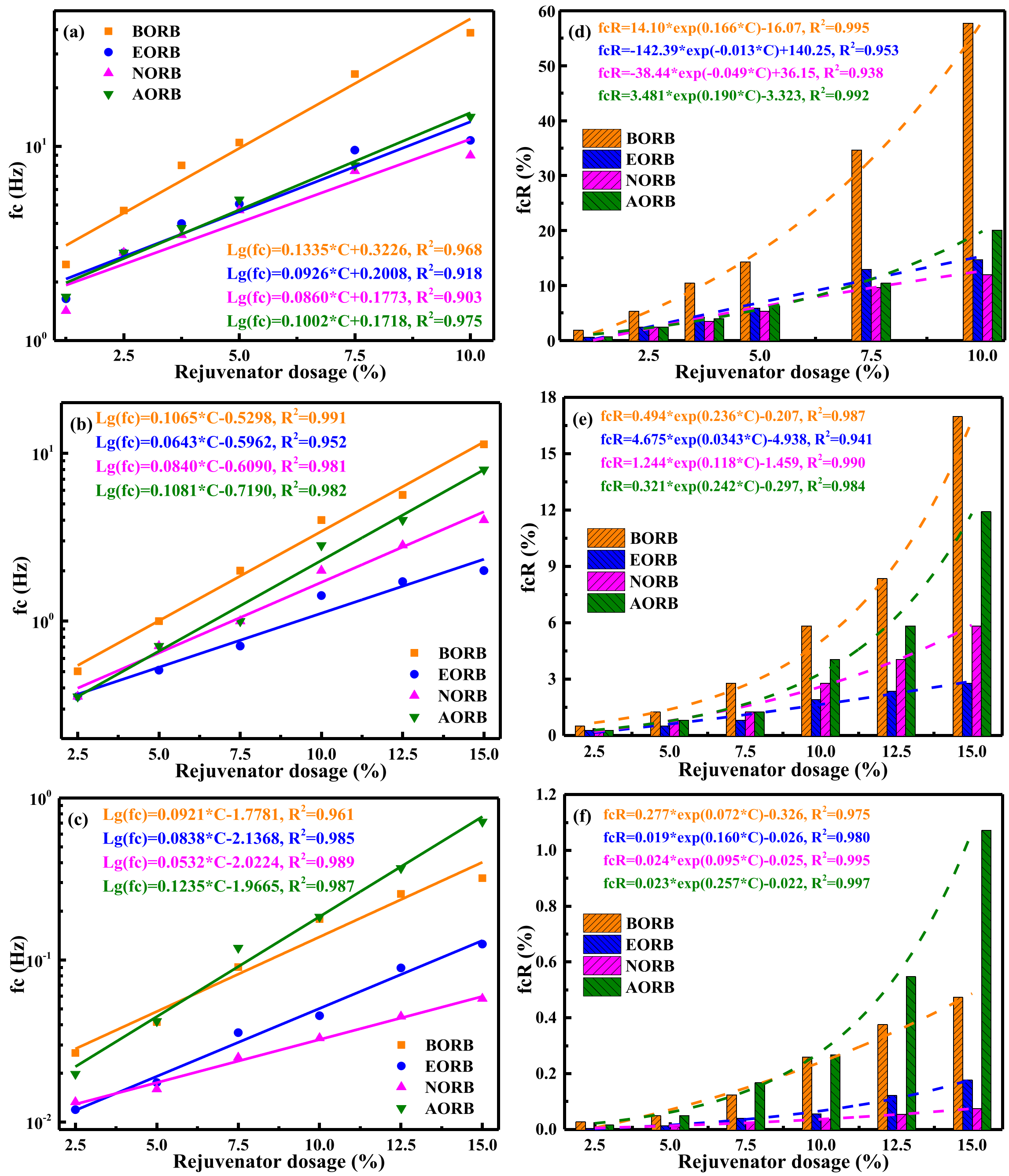

Fig. 15. The f_c and f_cR parameters of various rejuvenated bitumen.

The underlying mechanism for the difference in the rejuvenation efficiency of various rejuvenators is explained by the molecular structures and thermodynamic properties. Fig. 20**(b)** illustrates the molecular mobility, viscosity, cohesive energy density, and distribution of free volume. Based on their chemical properties, including average molecular weight, elemental compositions, and functional group distribution, as well as Gas chromatography-mass spectrometry (GC-MS) results, the average [42] and multi-component molecular models [43] were established for these four rejuvenators. It becomes evident that these four rejuvenators are comprised of chemical components with notably distinct molecular structures and characteristics. For instance, bio-oil primarily consists of long-chain unsaturated fatty acids, while engine oil contains long-chain alkanes and oxygen-containing additives. Naphthenic oil's primary component is polycyclic alkanes, accompanied by some long-chain alkanes and phenol molecules, such as 2-methoxy-4-methyl phenol. Additionally, the aromatic-oil rejuvenator comprises alkanes (like octadecane), aromatics (such as 2,7-dimethyl naphthalene), and additives (like furfural). This considerable diversity in molecular components and structures accounts for the variations in their rejuvenation efficiency concerning the chemical properties of aged bitumen.

The variances in the thermodynamic characteristics of these four rejuvenators were both forecasted and evaluated using molecular dynamics (MD) simulation techniques. The magnitude of molecular mobility for rejuvenators is BO > EO > NO > AO. In addition, the AO rejuvenator has the largest viscosity, followed by the NO and EO, while the viscosity of BO is the lowest. Regarding the free volume distribution, the bio-oil shows the most fractional free volume (FFV), followed by EO, NO, and AO. Therefore, the ranking of the thermodynamic properties of these four rejuvenators agrees well with the order of their rejuvenation

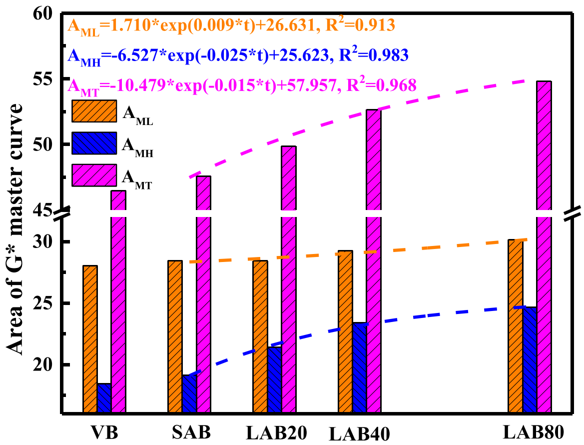

Fig. 16. The Area of G* master curves of virgin and aged bitumen.

efficiency on the rheological properties of aged bitumen (BORB > EORB > NORB > AORB).

4.6. Connections between chemical indices and rheological parameters

This study proposes three chemical indices (AI, CI, and SI) and three rheological parameters (G*, f_c, and A_{MH}) as the critical evaluation indicators for rejuvenation efficiency assessment. It is interesting to explore the potential chemo-rheological connections of rejuvenated bitumen. In this section, graphs are presented to illustrate the relationships between these essential chemical indicators and rheological properties, along with their respective rejuvenation percentages, in various rejuvenator-aged bitumen blends. Figs. 21 and 22 show the correlation curves between CI (CIR) and rheological indices (corresponding rejuvenation percentages). The crucial rheological parameters exhibit a linear correlation with the CI index, but the direction of this relationship varies depending on the specific rheological parameter. As the CI parameter increases, G* , A_{MH}, and A_{MT} exhibit linear growth, whereas the f_c value displays a decreasing trend. Furthermore, the shapes of these correlation curves are affected by both the type of rejuvenator and the level of bitumen aging. With increasing bitumen aging, the G* -CI, A_{MH}-CI, and A_{MT}-CI curves shift towards the upper right, while the f_c-CI curves move towards the lower right.

Of note, the impact of bitumen aging level on the correlation curves is notably less pronounced for bio-oil rejuvenated bitumen compared to the other rejuvenators. The absolute slope values for all correlation equations involving EORB, NORB, and AORB binders first increase and then decline with the increasing bitumen aging level, whereas BORB consistently demonstrates a continuous decreasing trend. Additionally, it's worth noting that the correlation curves for EORB consistently appear to the right of those for NORB, while the relative positioning of AORB's correlation curves depends on the factors of aging level, rejuvenator dosage, and rheological parameter type.

In addition, the G*R, f_cR, A_{MH}R, and A_{MT}R tend to increase linearly as a function of CIR. The rejuvenator type greatly influences the distribution range of the correlation curves. The bio-oil rejuvenated bitumen exhibits the widest correlation curves for all rheological indices, followed by AORB, NORB, and EORB binders. The increment in aging level results in the clockwise movement of f_cR-CIR curves for all rejuvenated binders. The changing trends of G*R-CIR, A_{MH}R-CIR, and A_{MT}R-CIR curves versus the aging degree of bitumen depend on rejuvenator type. For BORB, all correlation curves show a clockwise variation as the aging level grows. Nevertheless, the other three rejuvenated binders (EORB, NORB, and AORB) have the opposite trend. In addition, the order of slope values in correlation equations for rejuvenated bitumen is EORB > NORB > AORB > BORB, regardless of the aging level and rheological parameter type. The sensitivity of rheological rejuvenation percentages to CIR follows A_{MT}R > A_{MH}R > G*R > f_cR.

The correlation curves between critical rheological indices with SI are displayed in Fig. 23. The distribution scopes of associated curves for all rejuvenated binders are similar, differing from aforementioned CI-based correlation curves. The G* , A_{MH}, and A_{MT} values of rejuvenated bitumen intensify, and the f_c parameter reduces linearly as the SI grows. The aging level influences the correlation curves of EORB and NORB more than BORB and AORB, especially to G* , A_{MH}, and A_{MT} indices. Moreover, the fc-SI curve of BORB also dramatically affects the aging degree. For AORB, the sensitivity of these rheological indices to aging degree is the lowest. For BORB, EORB, and NORB binders, the escalation of the aging degree leads to the counter-clockwise rotation of G* -SI, A_{MH}-SI, and A_{MT}-SI correlation curves while the f_c-SI curves move toward the bottom left. Further, the absolute slope values of correlation equations tend to decrease as the aging degree grows.

The correlation curves of rheological indices-based rejuvenation percentages and SIR are plotted in Fig. 24. As the SIR rises, the G*R, f_cR, A_{MH}R, and A_{MT}R magnify linearly. All SIR-based correlation curves tend to move down as the aging level of bitumen deepens, and the corresponding slope values reduce gradually. The sensitivity of f_cR values of rejuvenated bitumen to the SIR is the lowest. The rejuvenator type also affects the correlation curves between rheological index-based rejuvenation percentages and SIR. For instance, the correlation curves of AORB binders are located at the bottom left. Further, the chemo-rheological correlations of rejuvenated bitumen are strongly determined by the rejuvenator dosage and aging degree of bitumen, and the latter has a greater impact.

The connections between the rheological indices and AI index and their rejuvenation percentages are presented in Figs. 25 and 26. Similarly, the rheological indices exhibit a linear positive (G*, A_{MH}, and A_{MT}) or negative (f_c) relationship with the AI. Both aging level and rejuvenator dosage influence these chemo-rheological correlation curves of rejuvenated bitumen, which reduces as the AI increases (especially for G*, A_{MH}, and A_{MT}). High aging level results in the movement of G* -AI, A_{MH}-AI, and A_{MT} correlation curves toward up left, while the f_c-AI curves transfer to the bottom left. In addition, the aging degree shows the lowest effect on the variation in correlation curves of the AORB binder, which has a greater influence on the slope parameters in correlation equations of the other three rejuvenated bitumen (BORB, EORB, and AORB) than rejuvenator type.

All rheological index-based rejuvenation percentages (G*R, f_cR, A_{MH}R, and A_{MT}R) enlarge linearly versus AIR index. The difference in aging degree and rejuvenator type contributes to the difference in scope and direction of correlation curves. The aging degree has a smaller impact on the AIR-based correlation curves of AORB, while it shows a greater effect on G*R-AIR and f_cR-AIR curves of BORB, EORB, and NORB binders than rejuvenator type influence. As the aging degree grows, the G*R-AIR curves of all rejuvenated binders tend to rotate counter-clockwise, while the f_cR-AIR shows an opposite change. It is worth mentioning that the impacts of aging level and rejuvenator type on the A_{MH}R-AIR and A_{MT}R-AIR curves of rejuvenated bitumen are limited, not observed in CIR and SIR cases. Therefore, within the allowable range of certain errors, there is a unified linear correlation between the AIR with A_{MH}R and A_{MT}R values of all rejuvenated binders without any impact from the aging degree of bitumen and rejuvenator type.

5. Conclusions and recommendations

This study aims to investigate the coupling effects of rejuvenator type/dosage and aging degree of bitumen on the chemical and rheological properties of rejuvenated bitumen, and propose the critical chemo-rheological indicators for rejuvenation efficiency evaluation. Moreover, the potential connections between these vital chemical and rheological indices of different rejuvenator-aged bitumen blends are explored preliminarily. The main conclusions drawn from this paper and some recommendations are listed as follows:

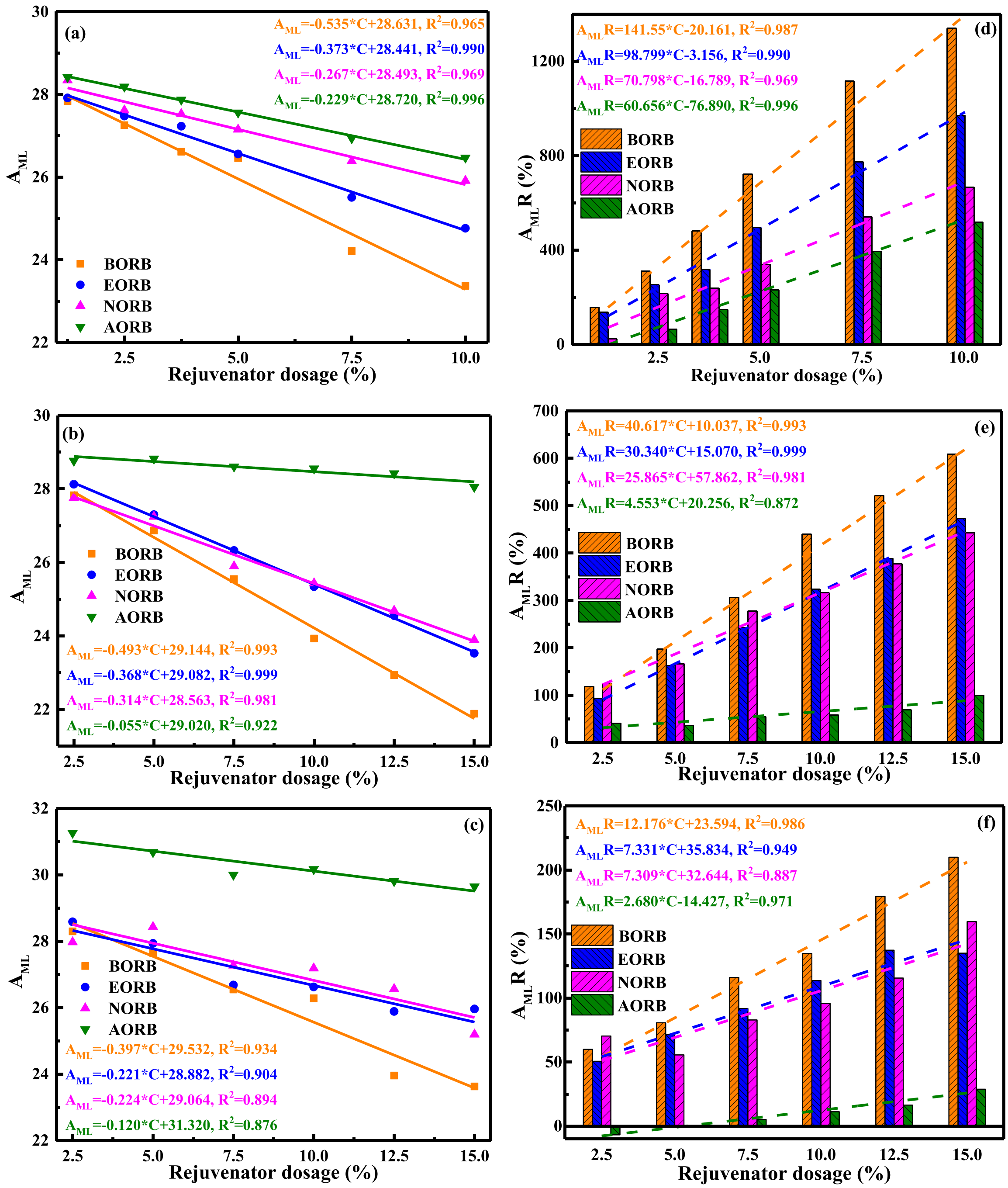

Fig. 17. The A_{ML} and $A_{ML}R$ values of various rejuvenated bitumen.

5.1. Conclusions

(1) The rejuvenation percentages of chemical indices (AI, AII, BAII, LCI, CI, and SI) show a linear relationship with rejuvenator dosage. Their variation trends strongly depend on the rejuvenator type and bitumen aging level. All rejuvenators can restore AI, CI, and SI values, but they fail to regenerate the AII parameter of aged bitumen. Moreover, the BAII value of aged bitumen can be recovered by rejuvenators except the bio-oil. Adding the aromatic-oil and naphthenic-oil is able to turn back the LCI parameter to the virgin bitumen level, which does not appear in bio-oil and engine-oil cases.

(2) All rejuvenators can regenerate the G* , δ, f_c, and A_{ML}, A_{MH}, and A_{MT} of aged bitumen in varying degrees. Nevertheless, these rejuvenators cannot restore G_c except the aromatic-oil. Furthermore, rheological indices exhibit different correlations with rejuvenator dosage and sensitivity degrees to rejuvenator type and aging degree of bitumen.

(3) The results reveal that the AI, CI, and SI act as effective chemical indices to assess the rejuvenation efficiency of BO, EO, NO, and AO rejuvenated bitumen. Meanwhile, the CI parameter is more appropriate to be a critical chemical index than the AI and SI. In addition, the G* , f_c, and A_{MH} are recommended as essential rheological indices for rejuvenation efficiency evaluation. Importantly, for these proposed critical chemical and rheological indicators, the magnitude of rejuvenation efficiency for four rejuvenators is BO > EO > NO > AO.

(4) The linear correlations between the critical chemical and rheological indices are observed, significantly affected by rejuvenator type and bitumen aging level. Within the allowable range of certain errors,

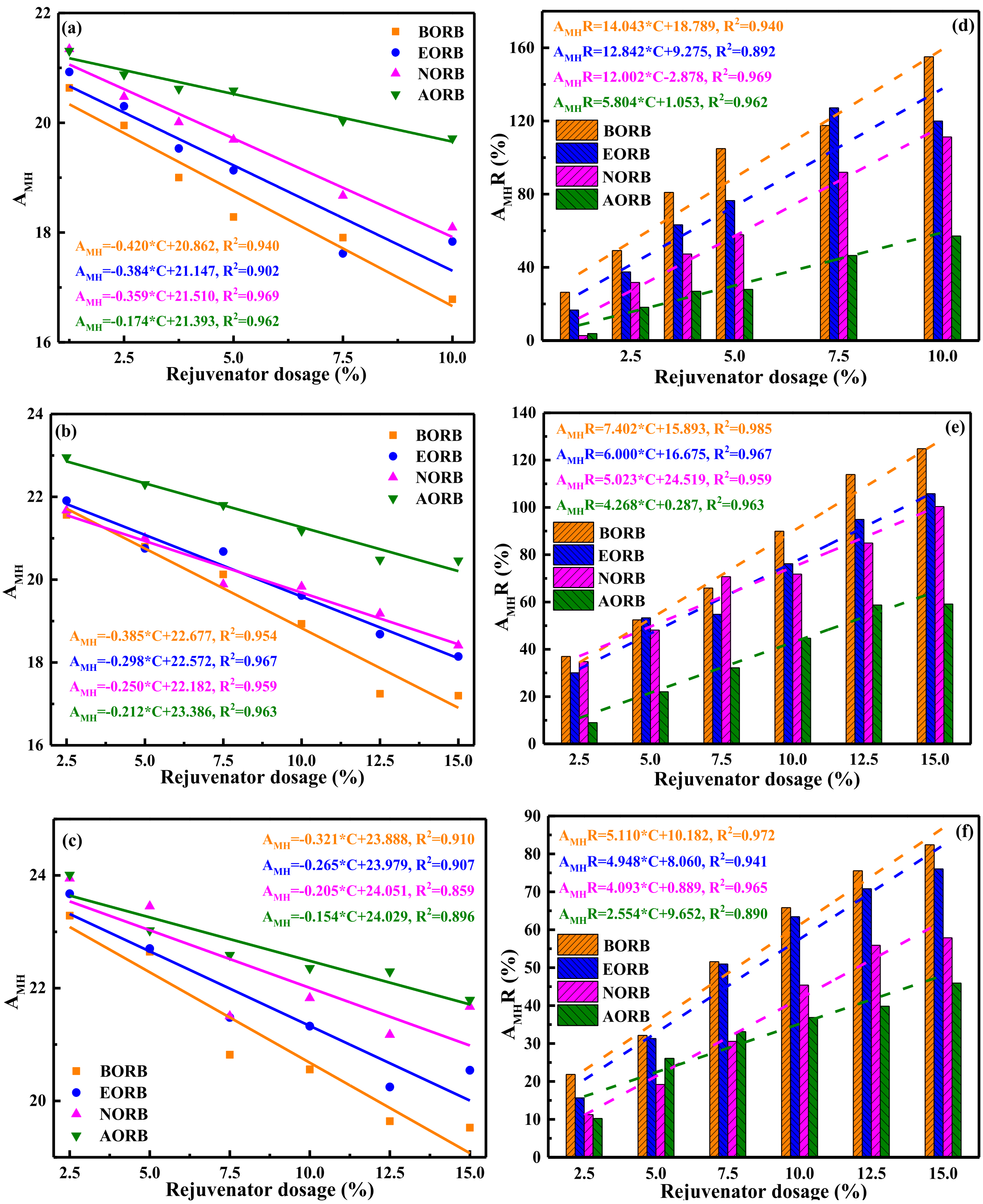

Fig. 18. The A_{MH} and $A_{MH}R$ values of various rejuvenated bitumen.

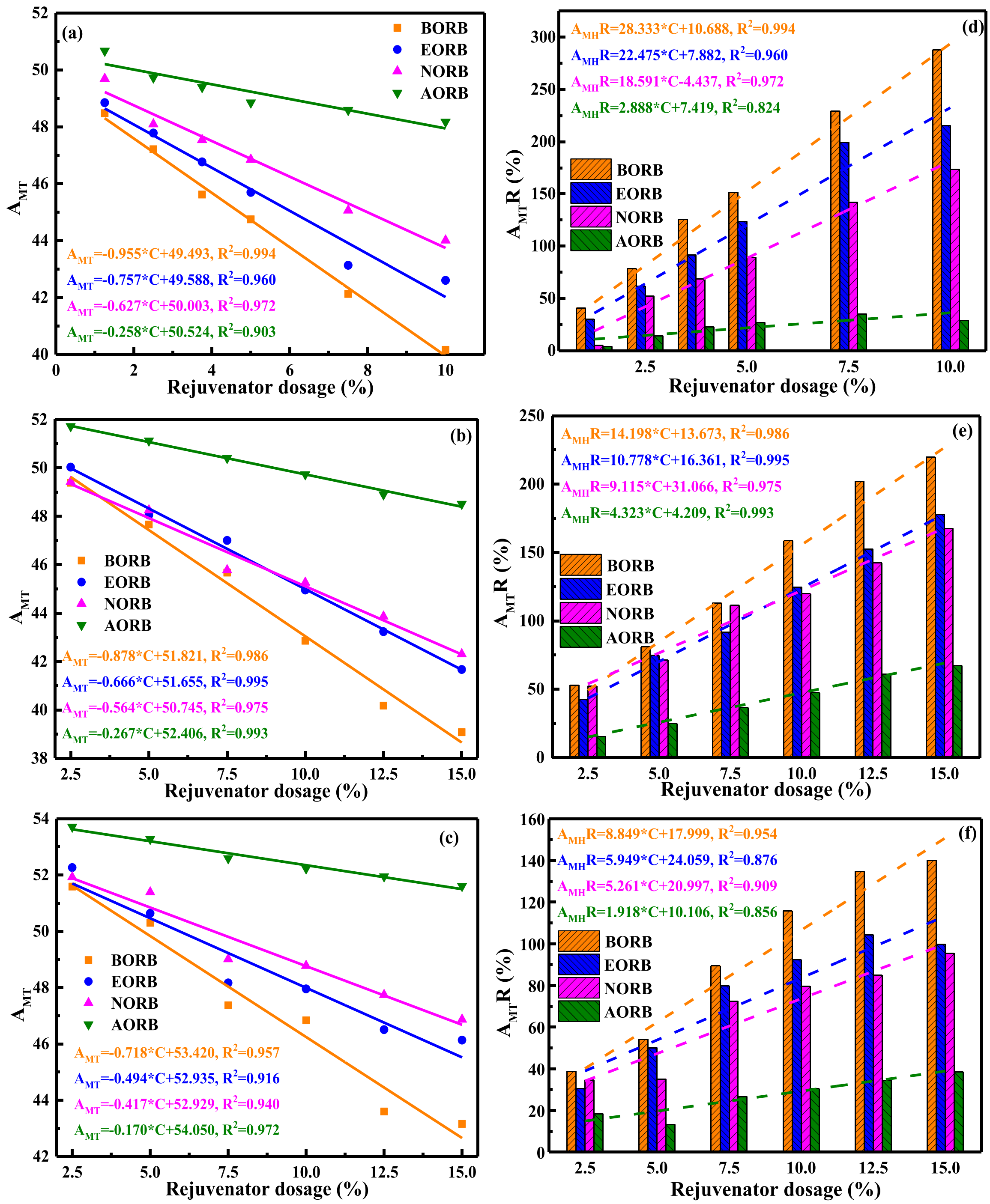

Fig. 19. The A_{MT} and $A_{MT}R$ values of various rejuvenated bitumen.

Table 6
Analysis of critical rheological parameters for rejuvenation efficiency evaluation.

Potential evaluation indicators	G^*	δ	G_c	f_c	A_{ML}	A_{MH}	A_{MT}
Recovery possibility	O	O	XXO[3]	O	O	O	O
Sensitivity to rejuvenator type	O	OXX[2]	O	O	O	O	O
Sensitivity to rejuvenator dosage	O[1]	O	O	O	O	O	O
Sensitivity to aging level	O	X	O	XO[4]	O	O	O
Rejuvenation percentage scope (%)	0-135	0-65	-40-30	0-60, 0-18, 0-1.1	0-1200	0-160	0-300

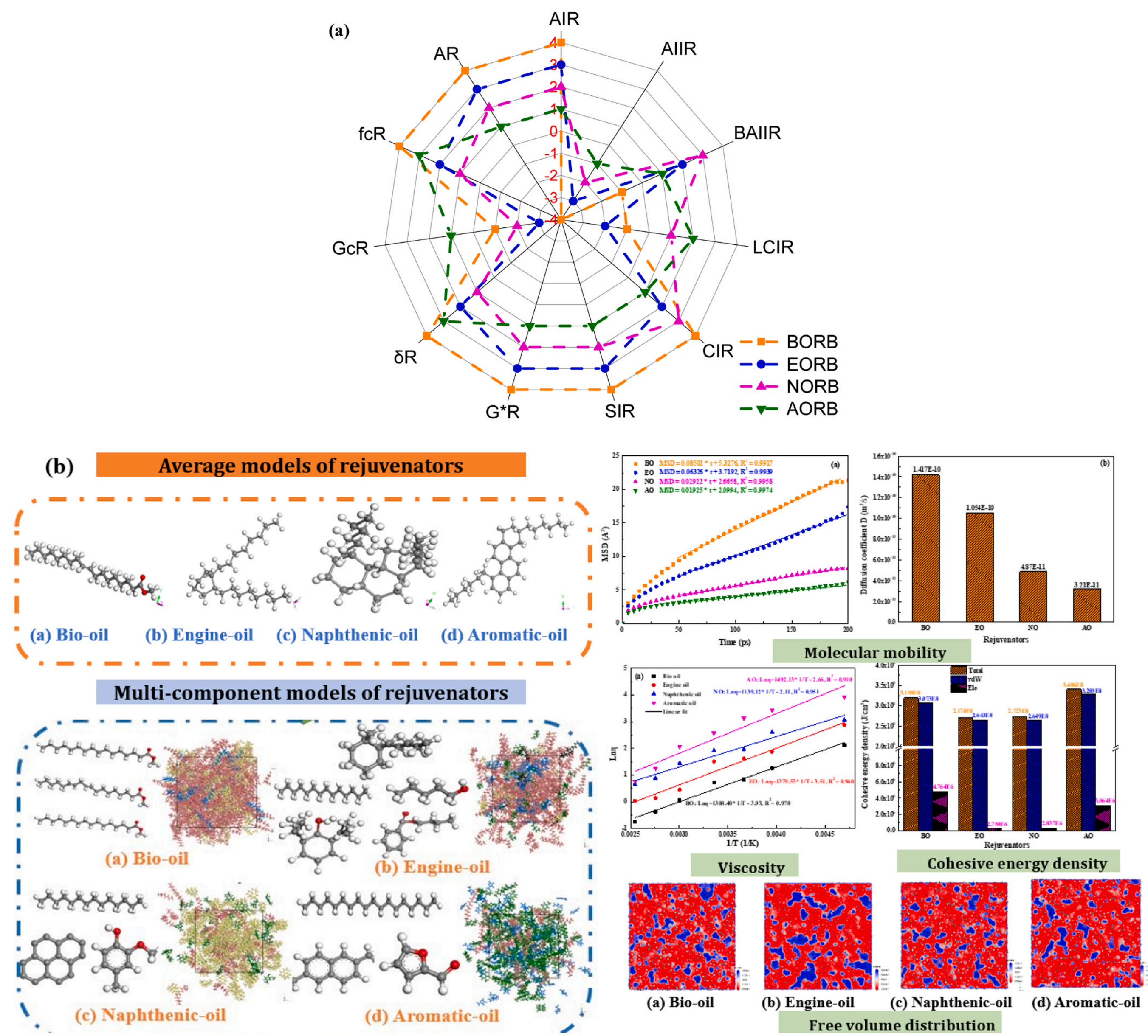

Fig. 20. Rejuvenation efficiency ranking (a) and difference in molecular structures and thermodynamic properties of rejuvenators (b).

there is a unified linear correlation between the AIR with A_{MH}R and A_{MT}R values of all rejuvenated binders without any impact from the aging degree of bitumen and rejuvenator type.

5.2. Recommendations for future work

(1) More rejuvenated binders (e.g. tall-oils, compound rejuvenator) will be characterized to validate the feasibility of proposed critical chemical and rheological indicators as well as their connections for rejuvenation efficiency evaluation.

(2) Different varieties of bitumen may demonstrate distinct responses to the effects of rejuvenators. Therefore, it is essential to conduct further research to assess the adaptability of the key chemical and rheological indicators proposed in this study to rejuvenation scenarios involving various bitumen sources.

(3) The underlying mechanism of the difference in chemical and rheological properties of various rejuvenated binders can be explored using multi-scale evaluation methods, such as molecular dynamics simulations and microstructural characterizations.

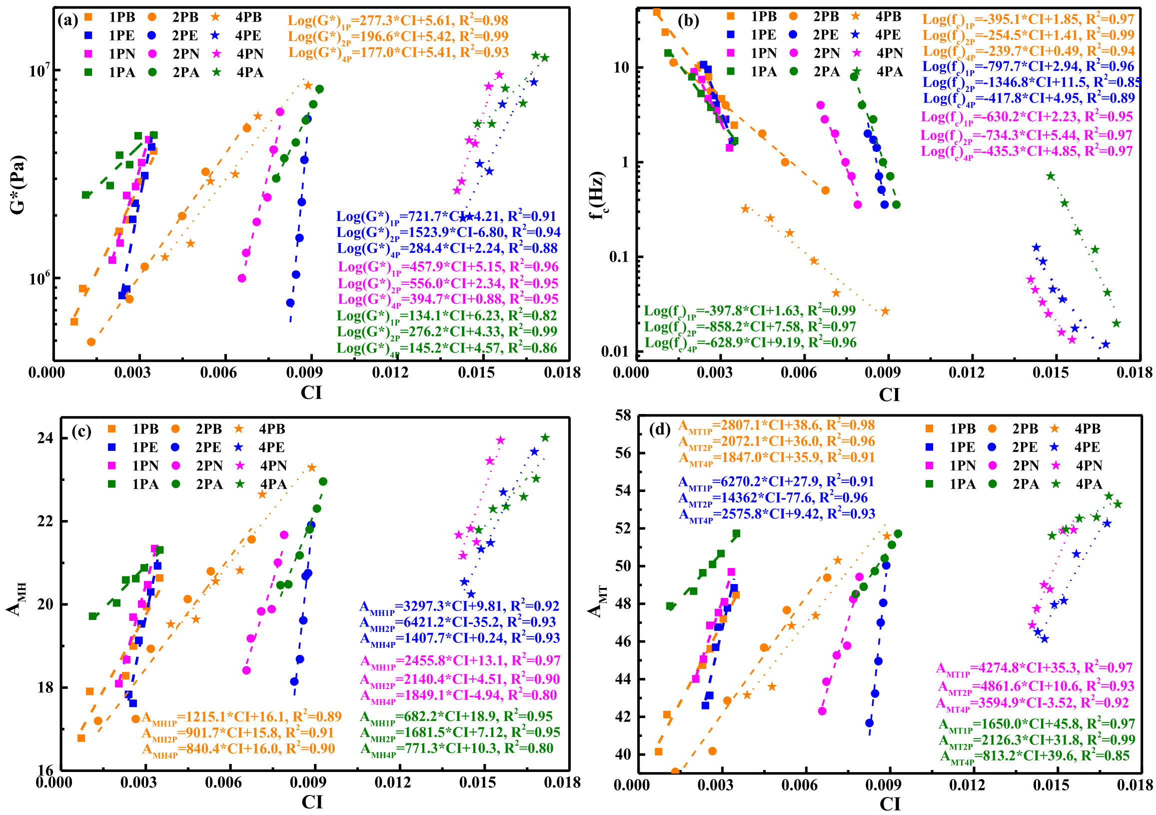

Fig. 21. Correlation curves between rheological indices with CI.

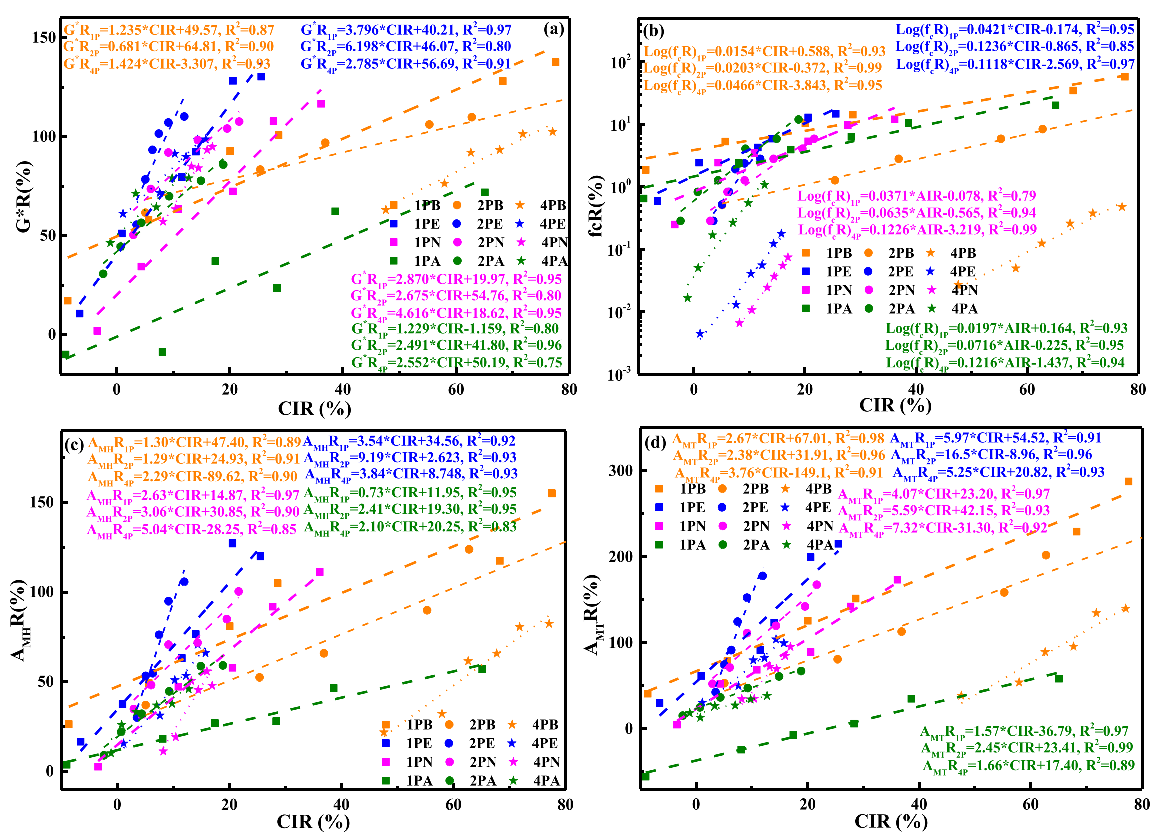

Fig. 22. Correlation curves between rheological rejuvenation percentages and CIR.

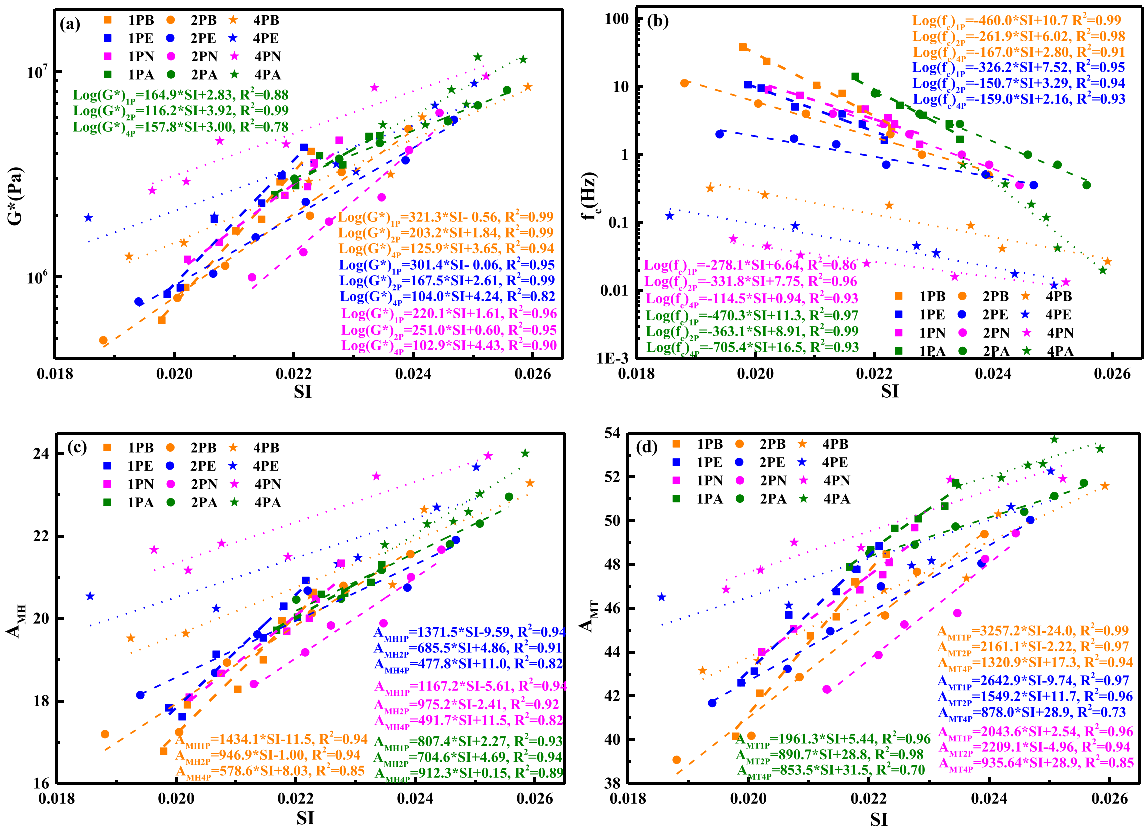

Fig. 23. Correlation curves between rheological indices with SI.

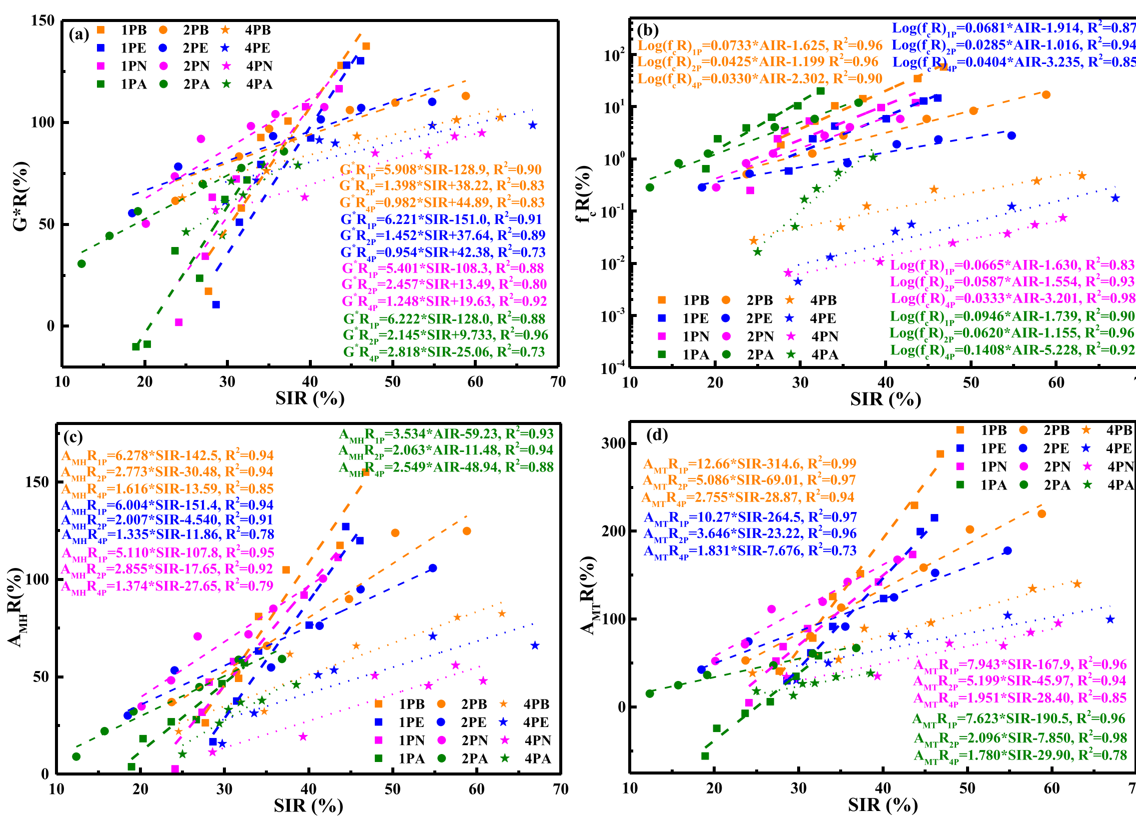

Fig. 24. Correlation curves between rheological rejuvenation percentages and SIR.

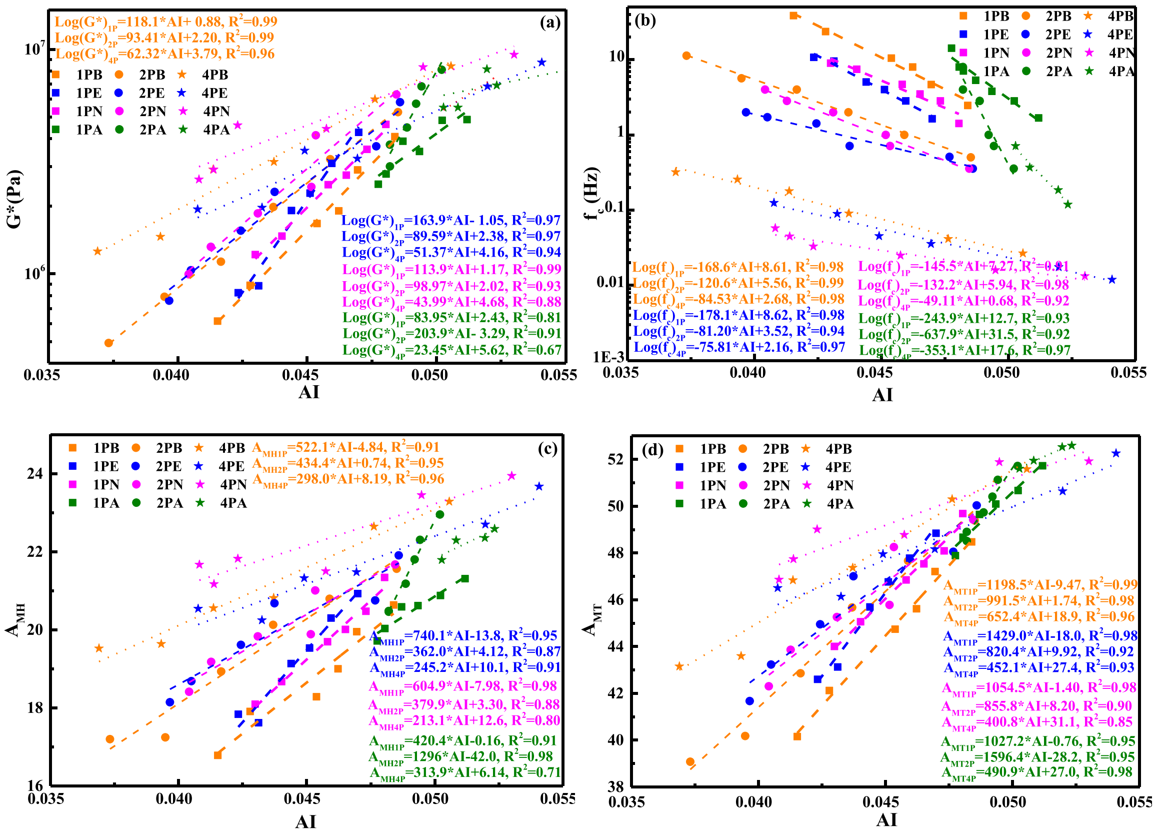

Fig. 25. Correlation curves between rheological indices and AI.

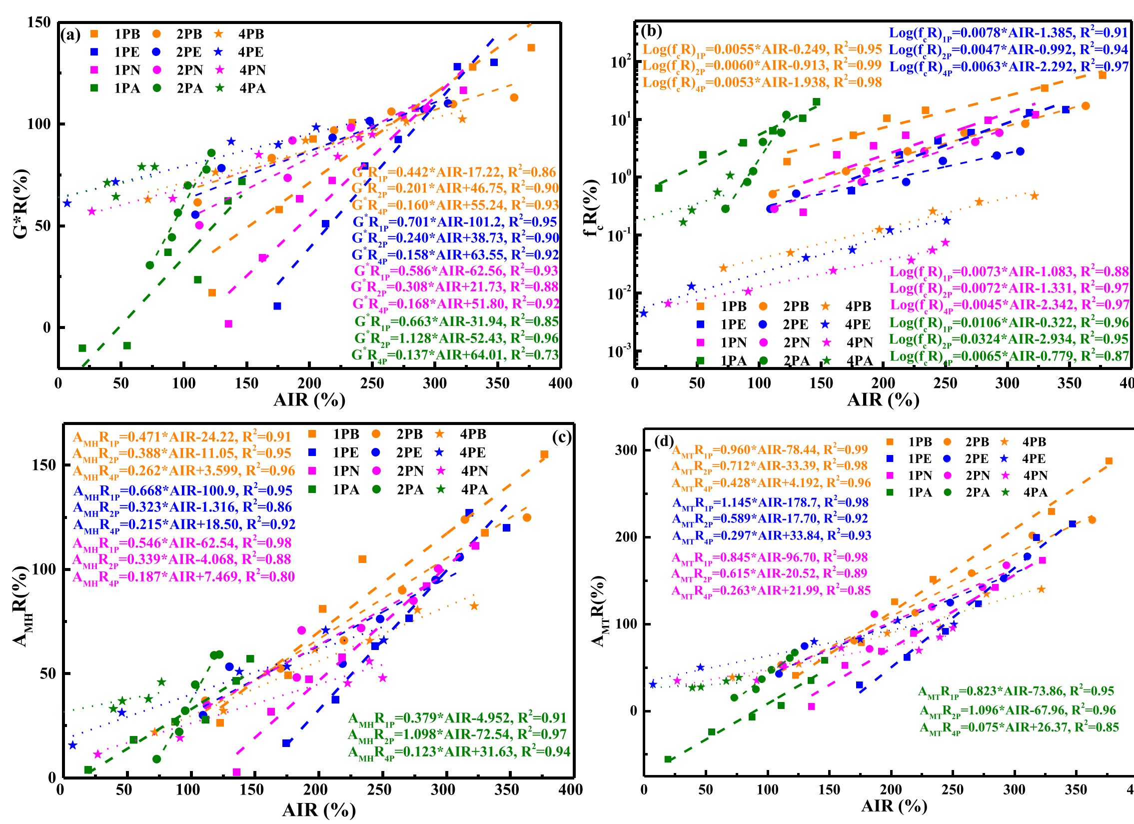

Fig. 26. Correlation curves between rheological rejuvenation percentages and AIR.

(4) The critical evaluation indices regarding the rejuvenation efficiency of various rejuvenator-aged bitumen blends on high-temperatures, low-temperatures, and fatigue performance will be recommended.

CRediT authorship contribution statement

Erkens Sandra: Methodology, Project administration, Resources, Supervision. **van Aggelen Michèle:** Methodology, Resources, Writing – review & editing. **Lin Peng:** Methodology, Writing – review & editing. **Ren Shisong:** Data curation, Formal analysis, Investigation, Methodology, Validation, Writing – original draft. **Liu Xueyan:** Project administration, Resources, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.conbuildmat.2023.134774.

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