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Original Article

Nonlinear rheological behavior of asphalt based on large amplitude oscillation shear test under different loading, aging, and modification conditions

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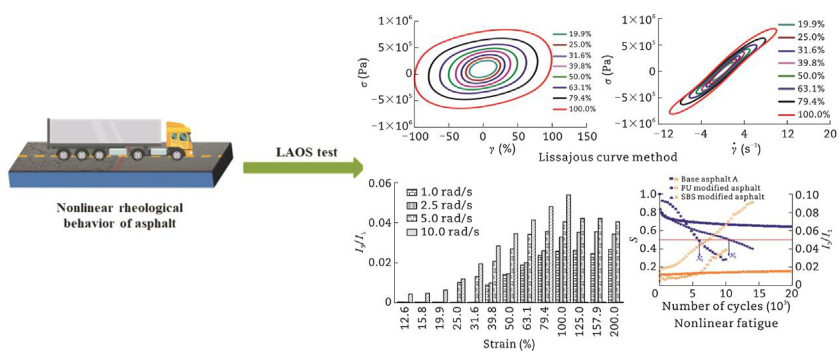
HIGHLIGHTS

- The decrease in temperature, increase in frequency and strain, and aging effect signally rise the nonlinearity of asphalt.
- The zero-strain nonlinear coefficient can accurately characterize the nonlinear viscoelasticity of asphalt.
- The fatigue failure process of asphalt is accompanied by an increasing degree of nonlinearity.
- 4% PU modifier exhibits a higher nonlinear lifting effect and a lower fatigue performance improvement effect than 4% SBS.

GRAPHICAL ABSTRACT



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ABSTRACT

To reveal the effects of environmental and loading conditions, as well as asphalt properties on the nonlinear rheological behavior of asphalt, the large amplitude oscillation shear (LAOS) test was introduced, and the Fourier transform rheology, Lissajous curve method, and the LAOS fatigue test have been applied to investigate the nonlinear rheological behavior of asphalt binders. The research results indicate that a decrease in temperature, an increase in shear frequency and strain level, the introduction of polymer modifiers, and the aging effect of asphalt can significantly increase the nonlinearity of asphalt, manifested by the higher relative magnitude of the third harmonic and zero-strain nonlinear coefficient. For the two polymer modifiers selected in this study, the 4% polyurethane modifier exhibits a higher nonlinear lifting effect than the 4% styrene-butadiene-styrene (SBS). The impact of long-term aging on nonlinear viscoelasticity is observably greater than that of short-term aging. The zero-strain nonlinear coefficient estimated based on the average value method can accurately characterize the nonlinear viscoelasticity of asphalt, which can serve as an effective supplement to the relative magnitude of the third harmonic. All asphalts exhibit shear thinning behavior under the test temperature of 24 °C, and the decrease in test temperature, the increase in shear rate and strain level, the introduction of modifiers, and the aging effect

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of asphalt all exacerbate the shear thinning behavior of asphalt. In addition, the fatigue failure process of asphalt materials is accompanied by an increasing degree of nonlinearity.

1. Introduction

Asphalt is an important constituent part of road paving materials, which plays a bonding role in asphalt mixture and is closely related to road performance such as cracking, rutting, and fatigue of asphalt pavement (Ge et al., 2023; Yi et al., 2018). As a heat-sensitive material, the mechanical properties of asphalt represent significant temperature and frequency dependence. Under different temperatures and frequency conditions, asphalt will show completely inconsistent rheological properties. Specifically, asphalt approaches fluid properties at high temperatures or low frequencies, while it exhibits solid properties at low temperatures or high frequencies. The viscous-elastic-plastic rheological properties of asphalt, coupled with the complexity of aggregate particle topology and aggregate gradation accumulation, have brought great challenges to the research on the pavement performance of asphalt mixture. To reveal the material properties and mechanical properties of the asphalt mixture, the key is to clarify the rheological properties of asphalt.

As an important and commonly used rheological test method, the dynamic shear oscillation test is widely used to investigate the rheological properties of various complex fluids and soft solids (Armstrong et al., 2016; Jiang et al., 2023; Wei et al., 2010). In the strain control mode, dynamic shear oscillation tests can be divided into small amplitude oscillation shear (SAOS) and large amplitude oscillation shear (LAOS) according to the applied strain (Hyun et al., 2011; Kamkar et al., 2022). In traditional rheological research, SAOS has become a standard method for testing the linear rheological properties of materials. Based on the linear rheology theory, this method applies the shear strain of the standard sine periodic function to the sample under a small strain, and the output stress response is also a standard sine periodic function (Läuger and Stettin, 2010). At this point, the material exhibits linear viscoelasticity, and the modulus is independent of the magnitude of strain. So far, researchers have carried out a lot of research on the linear viscoelasticity of asphalt and asphalt mixtures from the aspects of testing methods (Airey et al., 2008; Barco Carrión et al., 2017; Qi et al., 2024) and constitutive models (Liang et al., 2021; Zhao et al., 2013). However, existing studies have shown that under heavy load or high-temperature conditions, asphalt will inevitably bear the load in the nonlinear viscoelastic region (Cheng et al., 2021; Safaei and Castorena, 2017). In the actual service process, the real stress or strain of asphalt is large enough to reach the nonlinear viscoelastic region, showing more complex nonlinear viscoelasticity. Therefore, it is impossible to evaluate the mechanical properties of asphalt accurately and completely in the actual service process only through the linear viscoelastic behavior of asphalt, and it is imperative to study the nonlinear viscoelasticity of asphalt.

As the main method for nonlinear viscoelasticity testing of materials, LAOS refers to the test method of applying nonlinear viscoelastic region load to materials to obtain strain or stress response curves with time and has been successfully applied to colloids, suspensions, cement mortar, and other fields (Chan 2018; Lee et al., 2019; Macias-Rodriguez et al., 2018). In recent years, plenty of scholars have begun to investigate the nonlinear viscoelastic behavior of asphalt under the LAOS method. The study of Gulzar et al. (2023) quantified the nonlinear viscoelastic response of terminally blended crumb rubber modified asphalt under the LAOS method at different test temperatures and frequencies, finding that nonlinearity manifests greatly as strain levels and frequencies increase, and temperatures decrease. González et al. (2016) compared the nonlinear rheological behaviors of two kinds of polymer modified asphalt and two types of base asphalt at 20 °C. The research of Shan et al. (2020) investigated the nonlinear rheological behavior of SBS modified asphalt and found that SBS could increase the nonlinearity and the elastic proportion of asphalt. de Loë and Hesp (2021) established the relationship between LAOS and double-edge-notched tension (DENT) tests, which proved the potential of LAOS for high-strain failure performance characterization of asphalt. Zhu et al. (2023) studied the effects of strain amplitude, polymer content, and test frequency on the non-linear viscoelastic behaviors of asphalt. Liang et al. (2020) compared the nonlinear rheological behaviors of base asphalt, epoxy asphalt, and SBS modified asphalt. Diab and You (2017) characterized the response of polymer and crumb rubber modified asphalt binders under small and large deformations. In addition, Diab and You (2018) extended the LAOS method to asphalt mastics and found that a mastic that exhibits the best performance in the linear state is not guaranteed to perform superiorly in the nonlinear state. Most of the existing studies focus on comparing the nonlinear evaluation indexes of different kinds of asphalt under certain limited test conditions so far. However, the current research on the effect of loading conditions on the nonlinear rheological behavior of asphalt is not thorough, and the selection range of temperature, strain level, and shear frequency is not large enough and comprehensive enough. In addition, most current studies do not consider the impact of asphalt properties on its nonlinear rheology, such as the impact of aging effects on the nonlinear rheological behavior of asphalt, and the nonlinear differences between asphalts of the same penetration and from different sources. Furthermore, most current research focuses on the modified asphalt itself, while ignoring the changes in the nonlinear rheological behavior of asphalt before and after the addition of modifiers.

Given this, based on the LAOS test under strain control mode, this study expands the rheological properties of asphalt from the linear domain to the nonlinear domain, and the nonlinear rheological behavior of asphalt is analyzed comprehensively by using the Fourier rheology, Lissajous curve method, and LAOS fatigue test from the three aspects of test conditions (temperature, frequency, and strain level), asphalt properties (aging state and origins), and the use of modifiers. The research results provide a foundation for establishing an asphalt evaluation system based on nonlinear rheology.

2. Materials and methods

2.1. Materials

2.1.1. Base asphalts and modifiers

Two base asphalts (named A and B) with the performance grade (PG) of 64-22 and the penetration grade of 60/70 were selected in this study, and their physical properties are listed in Table 1.

Two commonly used polymer modifiers, styrene-butadiene-styrene (SBS) and polyurethane (PU), were adopted in this study. The SBS modifier used in this study was LG501S type produced by LG Chem Group of Korea, and its mass fraction of styrene is 31% and the density is 0.94 g/cm^3. Besides, the PU modifier used was produced by BASF Group of Germany, and its active reactive group was isocyanate with the main component of methylene diphenyl diisocyanate (MDI) (Li et al., 2022). The PU modifier was a dark brown transparent liquid, with a density of

Table 1
Basic physical properties of the base asphalts.

Item	Penetration at 25 °C (0.1 mm)	Ductility at 15 °C (cm)	Softening point (°C)	Brookfield viscosity at 135 °C (Pa·s)
Test result A	60.8	>100	49.5	0.493
Test result B	66.9	>100	48.9	0.383
Specification	AASHTO T49	AASHTO T51	AASHTO T53	AASHTO T316

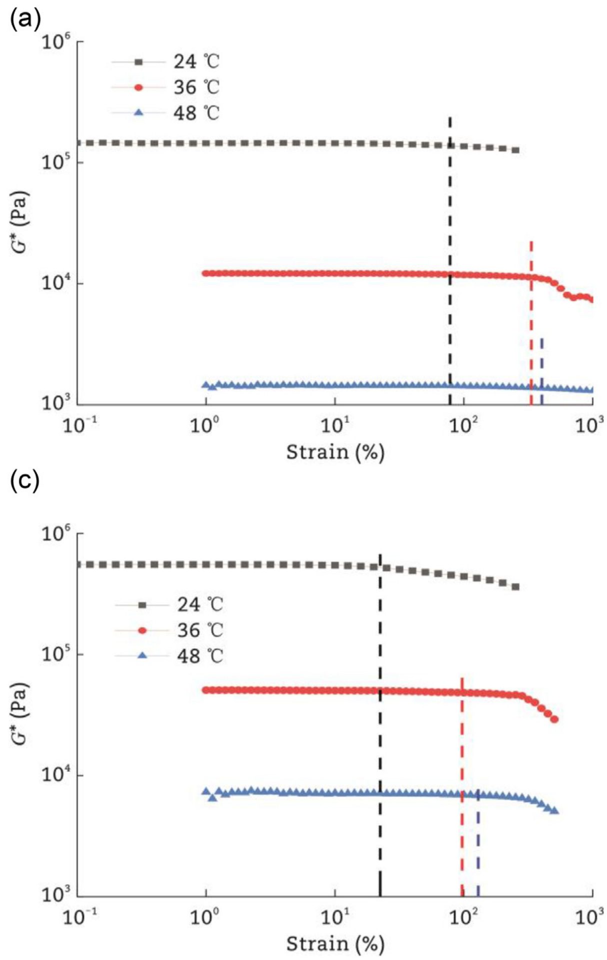

Fig. 1. Strain sweep test results of base asphalt at different frequencies. (a) 1.0 rad/s (b) 2.5 rad/s (c) 5.0 rad/s (d) 10.0 rad/s.

1.2 g/cm^3 and a viscosity of 0.21 Pa·s at room temperature (25 °C). For SBS modifier, this study selected the recommended dosage from the manufacturer, which is 4% of the asphalt mass and also a commonly used dosage (Chen et al., 2021). Meanwhile, as a better comparison between the two modifiers, the PU modifier was also selected with a dosage of 4%. In addition, to eliminate the effect of additives like stabilizing agent on the performance of asphalt, the introduction of stabilizing agent in SBS is not considered in current research.

2.1.2. Preparation of aging asphalt, SBS modified asphalt, and PU modified asphalt

The ROSS HSM 100L high shear mix was used for the preparation of modified asphalt in this study. The preparation procedure of SBS modified asphalt can be divided into three steps. At first, the base asphalt was heated at 170 °C to achieve a full flow state. Second, the SBS was added into the base asphalt stirring at 170 °C for 15 min with a stirring rate of 1500 revolutions per minute (RPM). Third, the stirring rate was increased to 4000 RPM at 170 °C for 1 h. In addition, the preparation procedure of PU modified asphalt can also be divided into three steps. In the beginning, the base asphalt was heated at 145 °C. Second, the PU modifier was added to the base asphalt. In this step, the stirring rate was kept at 300 RPM. Finally, the stirring rate was increased to 4000 RPM and stirred for 2 h. In the whole procedure of PU modified asphalt, the stirring temperature was kept at 145 °C.

According to the AASHTO T 240, the rotating thin film oven (RTFO) test was conducted on the base asphalt to simulate the short-term aging state of asphalt, and the pressurized aging vessel (PAV) test was carried out to simulate the long-term aging state of asphalt referring to AASHTO R28. In this study, base asphalt A was selected to prepare the modified asphalt to investigate the effect of using modifiers on the nonlinear rheological behavior of asphalt. Base asphalt B was used to prepare the aged asphalt to investigate the aging effect on the nonlinear rheological behavior of asphalt.

2.2. LAOS test

In this study, the ARES-G2 manufactured by TA Instruments of the United States was used to conduct the LAOS test. The conical plate with a uniform flow field was selected, the time sweep mode was adopted, and the original input strain and output stress were captured using the transient mode of the data acquisition mode. To reduce the interference of the equipment's inertia to the nonlinear test, the LAOS test was carried out in the low-frequency region, and four shear frequencies (1.0, 2.5, 5.0, and 10.0 rad/s) and three test temperatures (24 °C, 36 °C, and 48 °C) were selected respectively. 8 mm diameter conical plates were used at 24 °C and 36 °C, and 25 mm diameter conical plates were adopted at 48 °C.

To preliminarily determine the linear viscoelasticity range of the selected base asphalt under different test temperatures and frequency conditions, the base asphalt was subjected to the strain sweep tests at 24 °C, 36 °C, and 48 °C, respectively. Besides, the strain ranges at 24 °C, 36 °C and 48 °C were 0.1%–250%, 1%–500% and 1%–1000%, respectively (Yang et al., 2023). The results of the strain sweep test are presented in Fig. 1. It can be found that under any given temperature and frequency, the complex modulus of asphalt first remains unchanged and then decreases rapidly with the increase of strain. The strain level corresponding to a certain degree of decrease in complex modulus is generally referred to

Table 2
Strain selection of LAOS test at different loading conditions and test temperatures.

Frequency (rad/s)	Temperature (°C)	Strain (%)									
1.0	24	39.8	50.0	63.1	79.4	100.0	125.0	157.9	200.0	–	–
	36	140.7	157.9	177.2	200.0	223.1	250.0	280.8	315.1	–	–
	48	250.0	280.8	315.1	353.5	396.6	445.1	500.0	560.3	–	–
2.5	24	39.8	50.0	63.1	79.4	100.0	125.0	157.9	200.0	–	–
	36	140.7	157.9	177.2	200.0	223.1	250.0	280.8	315.1	–	–
	48	223.1	250.0	280.8	315.1	353.5	396.6	445.1	500.0	–	–
5.0	24	25.0	31.6	39.8	50.0	63.1	79.4	100.0	125.0	157.9	200.0
	36	100.0	111.8	125.0	140.7	157.9	177.2	200.0	223.1	–	–
	48	177.2	200.0	223.1	250.0	280.8	315.1	353.5	396.6	–	–
10.0	24	12.6	15.8	19.9	25.0	31.6	39.8	50.0	63.1	79.4	100.0
	36	79.4	88.8	100.0	111.8	125.0	140.7	157.9	177.2	–	–
	48	140.7	157.9	177.2	200.0	223.1	250.0	280.8	315.1	353.5	396.6

as the critical strain point between the linear viscoelastic region and the nonlinear viscoelastic region of asphalt materials (Wu et al., 2023). Before the critical strain point, the asphalt material can be within the linear viscoelastic range, and beyond this value, the material enters the nonlinear viscoelastic range. In this study, the strain point corresponding to the reduction of the complex modulus to 95% of the initial modulus is selected as the critical strain (Liang et al., 2020; Wu et al., 2023). From the figure, the increase in testing temperature and the decrease in shear frequency both improve the critical strain level of asphalt. For example, at the test temperature of 24 °C, the critical strains of asphalt at four shear frequencies of 10.0, 5.0, 2.5, and 1.0 rad/s are 12.6%, 25%, 39.8%, and 79.1%, respectively. Under the shear frequency of 5.0 rad/s, the critical strains of asphalt at three test temperatures of 24 °C, 36 °C, and 48 °C are 25.0%, 99.6%, and 125.4%. To observe the transition of asphalt materials from linear to nonlinear regions, the strain range selected in this study includes a portion of the strain values in the linear rheological region. In this study, the loading strain selection of the LAOS test at different temperatures and frequencies are summarized in Table 2.

2.3. Fourier transform rheology method

2.3.1. Relative magnitude of the third harmonic I_3/I_1

Due to the nonlinear rheological characteristics of materials, in the LAOS test, when the input strain is a standard sinusoidal periodic function, the output strain is no longer a standard sinusoidal periodic function but contains the contribution of higher harmonics. The Fourier rheology method aims to quantify this non-standard sinusoidal periodic output signal, which converts the time-domain response of stress into frequency-domain response through the Fourier transform, including the weight of amplitude and phase angle of different orders and their corresponding frequency in the total response (González et al., 2016). The Fourier expansion of the response stress $\sigma(t)$ of the non-sinusoidal periodic function can be expressed as follows.

$$\sigma(t) = \sum_{n=1}^{\text{odd}} \sigma_n \sin(n\omega t + \delta_n) \tag{1}$$

where ω is the shear frequency, t is the time, n is the order of the high-order harmonic, σ_n is the amplitude of the n-order output stress harmonics, δ_n is the phase angle of the n-order output stress harmonics.

If Eq. (1) is Fourier transformed, it will show obvious signals at the positions of ωt, $3\omega t$, $5\omega t$ and $7\omega t$ on the Fourier rheological map. Most materials behave as isotropic fluid in the LAOS test, and their mechanical response should only have odd non-zero harmonic components (Shan et al., 2018). In the Fourier rheology method, the relative magnitude of the third harmonic I_3/I_1 is often used as the main index for evaluating the degree of nonlinear rheology of asphalt. The larger the I_3/I_1, the higher the degree of nonlinearity of the material.

2.3.2. Zero-strain nonlinear coefficient Q_0

Hyun and Wilhelm (2009) found that I_3/I_1 is proportional to the quadratic of the strain amplitude (γ^2) regardless of the shear frequency and test temperature, and proposed the nonlinear coefficient $Q = \frac{I_3/I_1}{\gamma^2}$ for the first time to characterize the nonlinear viscoelasticity of the material. In addition, referring to the definition of zero-shear viscosity (the viscosity value when the shear rate is 0 or infinitely close to 0), Hyun and Wilhelm (2009) defined the plateau value of the Q-γ curve in log-logarithmic coordinates as the zero-strain nonlinear coefficient Q_0. This parameter does not include the load parameter, which better reflects the nonlinear viscoelasticity of the material. The definitions of Q_0 are as follows.

$$Q_0 = \lim_{\gamma \to 0} Q = \lim_{\gamma \to 0} \frac{I_3/I_1(\gamma)}{\gamma^2} \tag{2}$$

Hyun and Wilhelm (2009) also proposed an analytical solution for Q_0, however, this analytical solution is too complex and not suitable for this study. On this basis, Abbasi et al. (2013) proposed two approximate solutions to Q_0. Wilhelm (2002) observed that $I_3/I_1(\gamma)$ generally exhibits a sigmoid dependence on the strain amplitude (γ), which can be described empirically.

$$I_3 \Big/ I_1(\gamma) = A\left(1 - \frac{1}{1 + B\gamma^C}\right) \tag{3}$$

where A is the maximum value of the I_3/I_1, B is the inflection point of the curve, C is the scaling exponent.

It is not difficult to find that, based on Eq. (3), when $C = 2$, $Q_0 = AB$. However, this solution (special value method) is only suitable for asphalt materials with C approximately equal to 2 at low load levels (Abbasi et al., 2013). The second solution is the average value method, which takes the average value of multiple Q values within the platform range of the Q-γ curve as the Q_0 (Abbasi et al., 2013). Based on the above two approximate solutions, this study calculated the Q_0 of asphalt, which are recorded as Q_{01} and Q_{02}, respectively.

2.4. Lissajous curve method

2.4.1. Lissajous curve geometry method

By plotting the input strain and output stress, a closed stress-strain curve can be obtained, which is the Lissajous curve. In the field of linear rheology, the output stress is a standard sinusoidal periodic function, so the Lissajous curve is a standard ellipse. As a contrast, in the field of nonlinear rheology, because the output stress will deviate from the standard sinusoidal function, the Lissajous curve will also deviate from the standard ellipse. Similarly, a closed curve can be obtained by mapping the strain rate and stress. Generally, the stress-

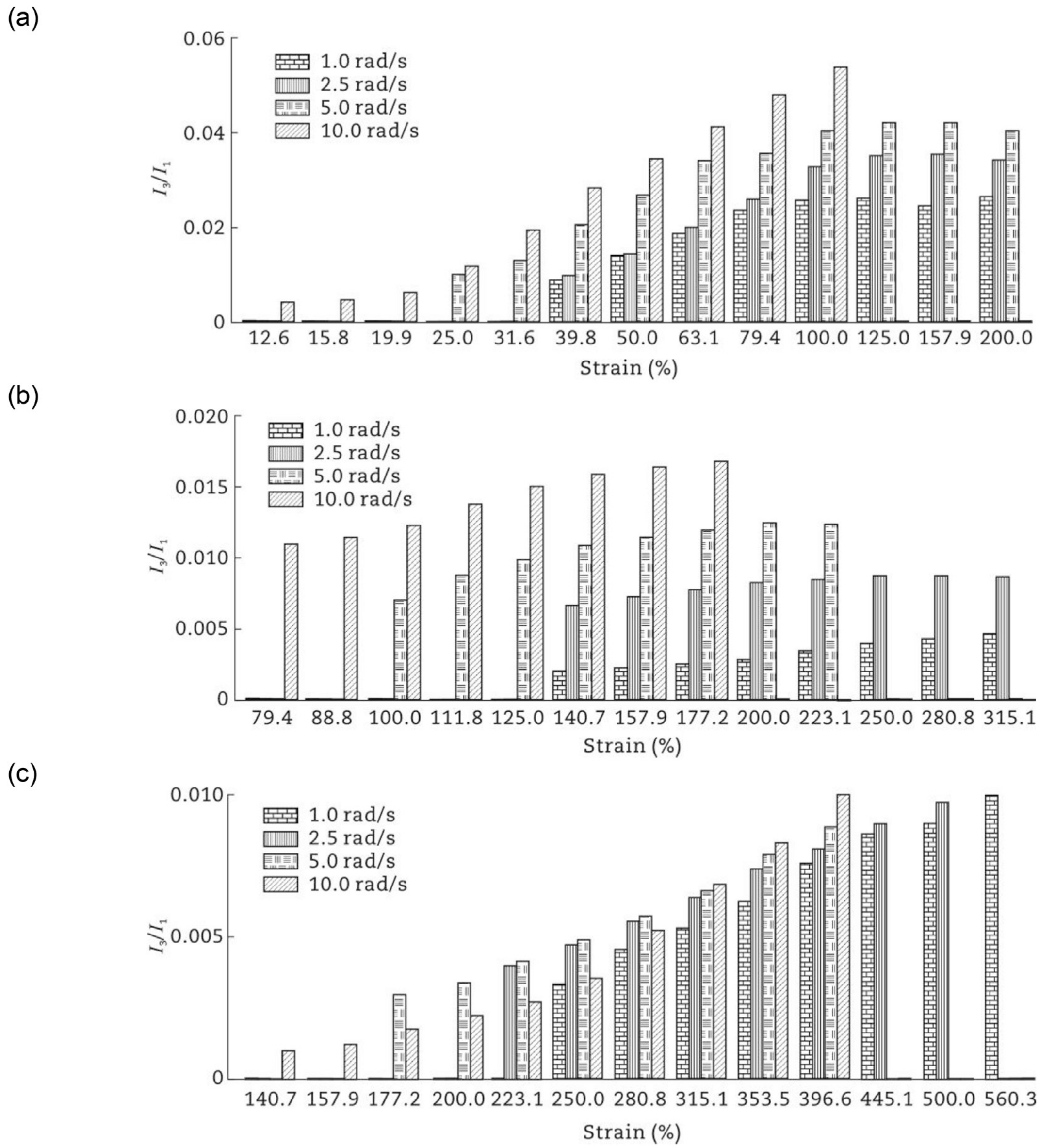

Fig. 2. I_3/I_1 of base asphalt A at different temperatures. (a) 24 °C. (b) 36 °C. (c) 48 °C.

strain curve is defined as the elastic Lissajous curve, and the stress-strain rate curve is defined as the viscous Lissajous curve. The linear and nonlinear rheological responses of materials can be more intuitively represented by Lissajous curves, so it is still a convenient method to identify the nonlinear rheological behavior of materials. In addition, by observing the shape change of the Lissajous curve, special nonlinear rheological properties of materials can be obtained, such as in-loop strain stiffening, in-loop strain softening, and yield (González et al., 2016).

2.4.2. Lissajous curve parameters method

It is worth noting that the Lissajous curve geometry method can only qualitatively characterize the nonlinear rheological characteristics of asphalt. Given this, researchers proposed certain nonlinear viscoelastic parameters based on the Lissajous curve geometry to quantitatively characterize the nonlinear viscoelastic behavior of asphalt materials. According to Ewoldt et al. (2008), the minimum-strain modulus G'_{M}, the large-strain modulus G'_{L}, the minimum-rate dynamic viscosity η'_{M}, and the large-rate dynamic viscosity η'_{L}, were defined. G'_{M} is the tangent slope when the strain $\gamma = 0$ (minimum strain) in the elastic Lissajous curve; G'_{L} is the slope of the straight line from $\gamma = \gamma_0$ (maximum strain) to the origin in the elastic Lissajous curve; η'_{M} is the tangent slope when the strain rate $\dot{\gamma} = 0$ (minimum strain rate) in the viscous Lissajous curve; η'_{L} is the slope of the straight line from $\dot{\gamma} = \dot{\gamma}_0$ (maximum strain rate) to the origin in the viscous Lissajous curve. The calculation formula for the above four parameters is as follows.

$$G'_{\mathrm{M}} = \left.\frac{\mathrm{d}\sigma}{\mathrm{d}\gamma}\right|_{\gamma=0} \tag{4}$$

$$G'_{\mathrm{L}} = \left.\frac{\sigma}{\gamma}\right|_{\gamma=\gamma_0} \tag{5}$$

$$\eta'_{\mathrm{M}} = \left.\frac{\mathrm{d}\sigma}{\mathrm{d}\dot{\gamma}}\right|_{\dot{\gamma}=0} \tag{6}$$

$$\eta'_{\mathrm{L}} = \left.\frac{\sigma}{\dot{\gamma}}\right|_{\dot{\gamma}=\dot{\gamma}_0} \tag{7}$$

Furthermore, the strain-stiffening ratio S and the shear-thickening ratio T were established based on the four parameters above as follows (Ewoldt et al., 2008).

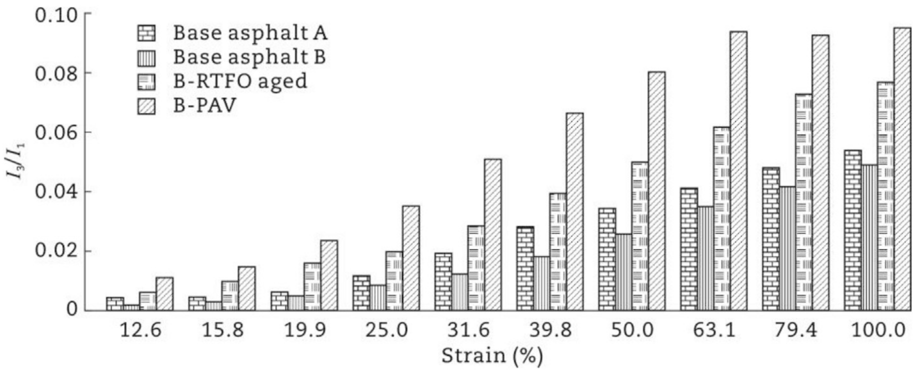

Fig. 3. I_3/I_1 of base asphalt A and base asphalt B under different aging states.

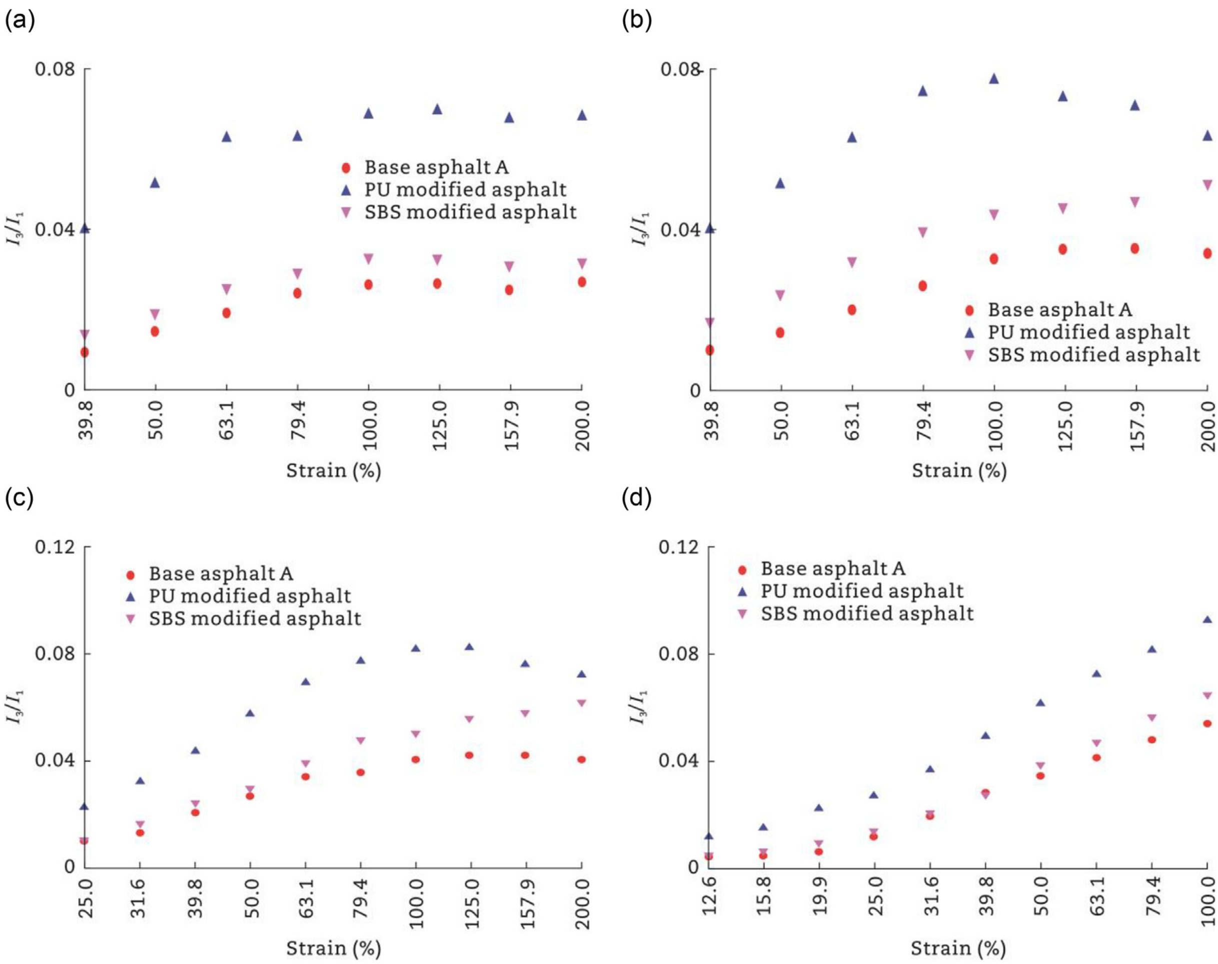

Fig. 4. I_3/I_1 of asphalt binders at different frequencies. (a) 1.0 rad/s (b) 2.5 rad/s (c) 5.0 rad/s (d) 10.0 rad/s.

$$S = \frac{G'_L - G'_M}{G'_L} \tag{8}$$

$$T = \frac{\eta'_L - \eta'_M}{\eta'_L} \tag{9}$$

When $S > 0$, it means that when the strain increases, the slope of the stress-strain curve increases continuously, showing the behavior of strain stiffening, otherwise it is strain softening. Similarly, when $T > 0$, the asphalt material is shear thickening, otherwise it is shear thinning. In addition, when the material behaves as linear viscoelasticity, $G'_M = G'_L$, $\eta'_M = \eta'_L$. When the material enters the nonlinear viscoelastic stage, $G'_M \neq G'_L$ and $\eta'_M \neq \eta'_L$. In this study, the nonlinear rheological characteristics of asphalt were further quantitatively characterized using the above Lissajous curve parameters according to the input strain and output stress responses obtained from the experiments.

3. Results and discussions

3.1. Analysis results of I_3/I_1

3.1.1. Effect of test conditions on I_3/I_1

The I_3/I_1 of base asphalt A at four shear frequencies (1.0, 2.5, 5.0, and 10.0 rad/s), three test temperatures (24 °C, 36 °C, and 48 °C), and large

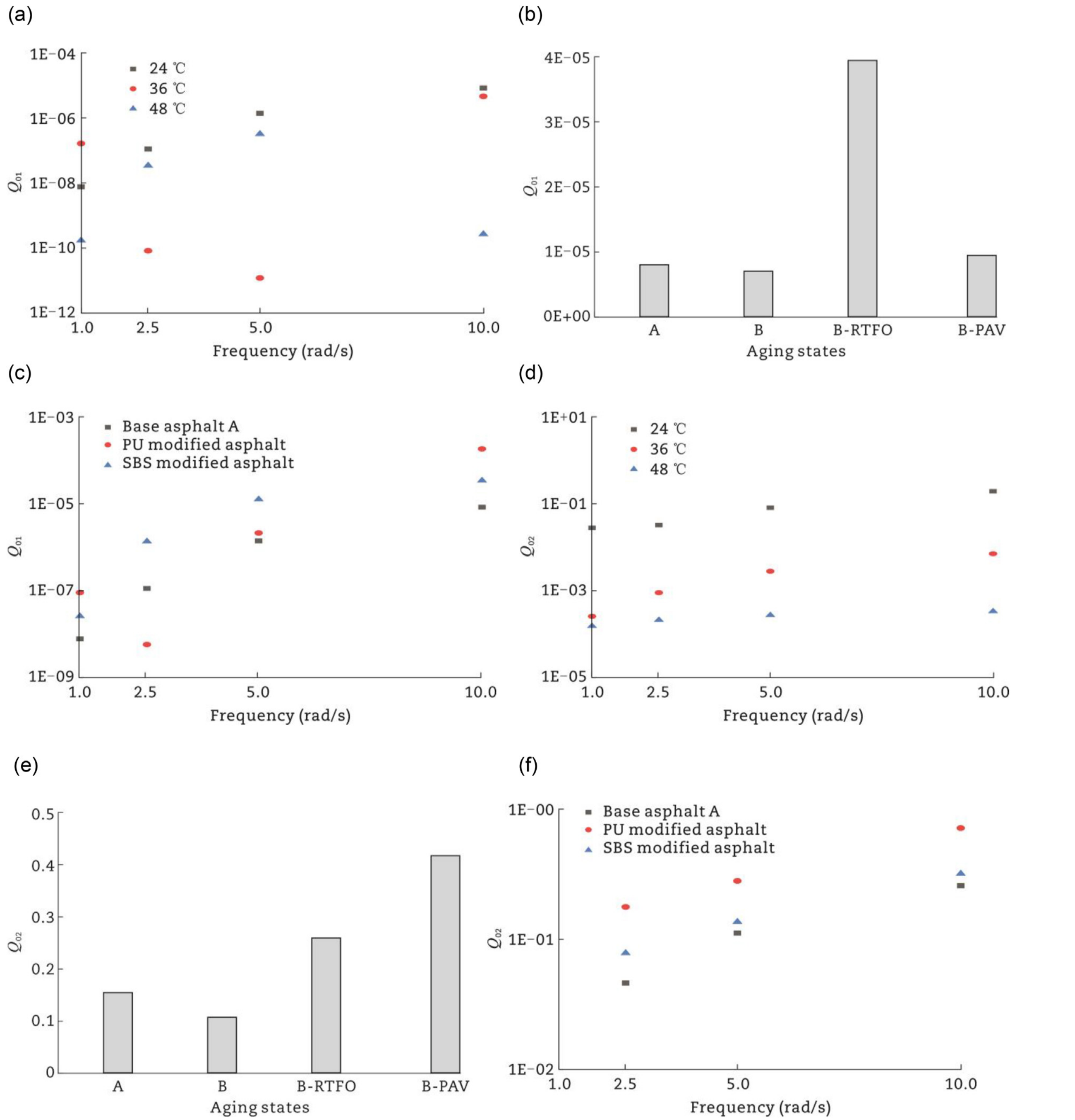

Fig. 5. Q of asphalt binders. (a) Q_{01} at different temperatures. (b) Q_{01} at different aging states. (c) Q_{01} at different modifications. (d) Q_{02} at different temperatures. (e) Q_{02} at different aging states. (f) Q_{02} at different modifications.

strain level ranges are shown in Fig. 2. From Fig. 2, the increase in strain levels, the improvement of shear frequencies, and the promotion of test temperatures all increase the nonlinear rheological degree of asphalt binders, which manifests as the higher I_3/I_1. In addition, it is worth noting that the correlation between I_3/I_1 and the shear frequency is not obvious at 48 °C, and the values of I_3/I_1 between different frequencies are extremely low and very close. Even at some strain levels, there are cases where the I_3/I_1 corresponding to a shear rate of 5.0 rad/s is higher than that of 10.0 rad/s. This is because asphalt has a very low degree of nonlinear rheology at relatively high temperatures, and the test results at this point are easily affected by the signal noise of the instrument itself, thereby affecting the accuracy. Therefore, 48 °C was the highest temperature adopted in this study.

3.1.2. Effect of asphalt properties on I_3/I_1

Fig. 3 presents the I_3/I_1 comparison of base asphalt A and base asphalt B under different aging states (unaged, RTFO aged, and PAV aged) at the test temperature of 24 °C and shear frequency of 10 rad/s. It can be found from Fig. 3 that the difference in I_3/I_1 values between base asphalt A and B is relatively small. This indicates that for the two materials with the same penetration grade selected in this study, the source of asphalt will not significantly change the nonlinear rheological characteristics of asphalt. In addition, the I_3/I_1 value of base asphalt B after RTFO and PAV aged exhibited a significant increase, and the increase was more pronounced when the strain level was higher. Moreover, the I_3/I_1 value of the B-PAV aged sample was much higher than that of the B-RTFO aged sample. This indicates that aging has a significant impact on the

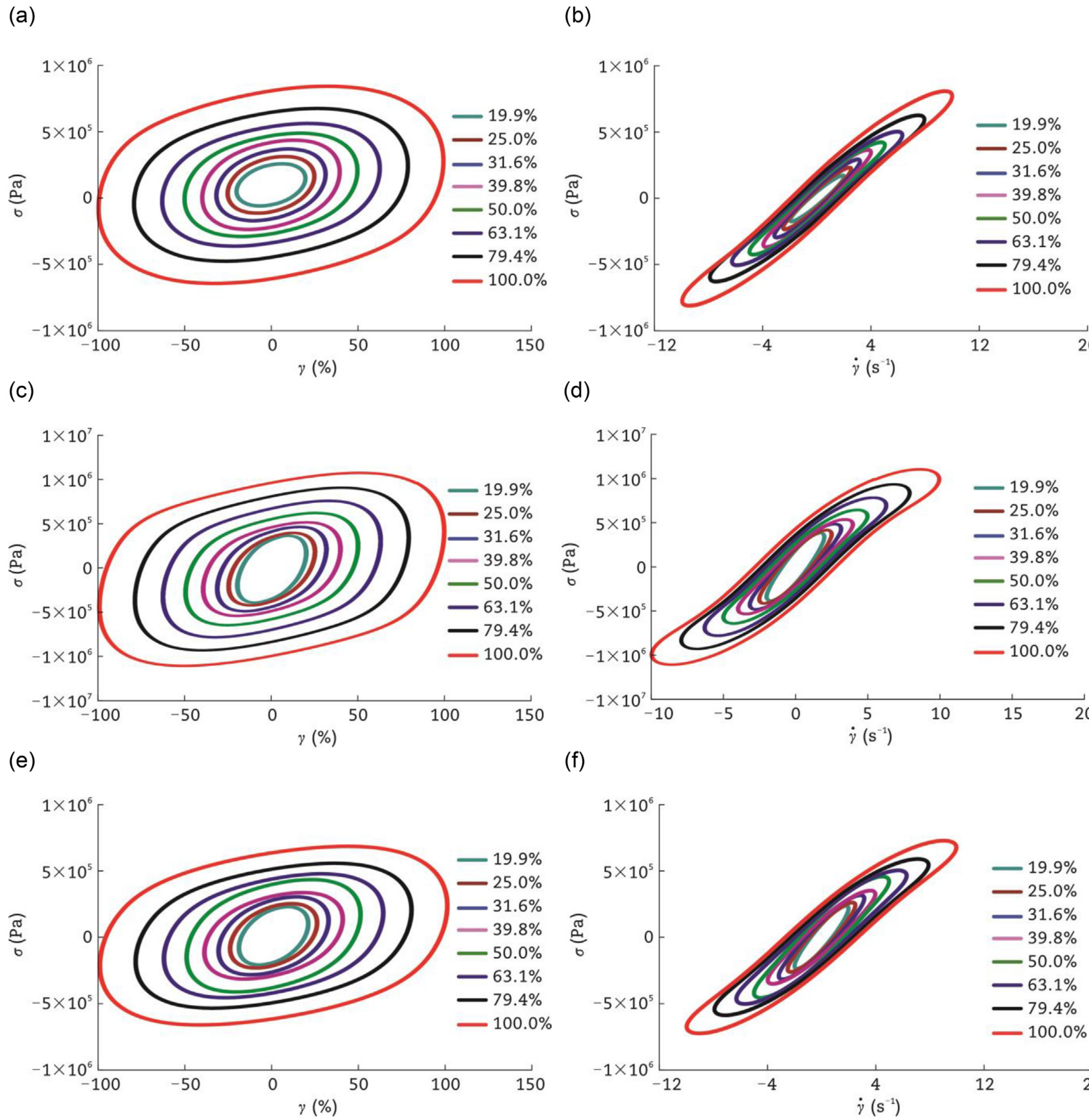

Fig. 6. Lissajous curves at different strain levels. (a) Elastic Lissajous curve of base asphalt A. (b) Viscous Lissajous curve of base asphalt A. (c) Elastic Lissajous curve of PU modified asphalt. (d) Viscous Lissajous curve of PU modified asphalt. (e) Elastic Lissajous curve of SBS modified asphalt. (f) Viscous Lissajous curve of SBS modified asphalt.

nonlinear characteristics of asphalt, and as the degree of aging increases, asphalt exhibits more remarkable nonlinear rheological characteristics (Fan et al., 2021).

3.1.3. Effect of modifiers on I_3/I_1

Fig. 4 presents the variation of the I_3/I_1 with the strain level of base asphalt A, PU modified asphalt, and SBS modified asphalt at the test temperature of 24 °C and four shear frequencies. As we can see in Fig. 4, the incorporation of modifiers can have a significant effect on the level of nonlinear rheology of asphalt. Among the three asphalt binders, the nonlinear rheological degree of PU modified asphalt is the strongest, followed by SBS modified asphalt, and the nonlinear rheological degree of base asphalt is the weakest, which is consistent with the conclusion of the Lissajous curve geometry above. The I_3/I_1 of PU modified asphalt is significantly higher than that of base asphalt and SBS modified asphalt, indicating that the addition of polyurethane has greatly changed the internal structure of asphalt so that PU modified asphalt shows a strong nonlinear rheological level. Therefore, for modified asphalt, it is easier to exhibit significant nonlinear rheological characteristics than base asphalt, which illustrates the necessity to investigate the performance of modified asphalt by using nonlinear rheological theory.

3.2. Analysis results of Q_0

Fig. 5 shows the nonlinear coefficients Q_{01} and Q_{02} of asphalt binders using two approximate solutions. From Fig. 5, under different loading, aging and modification conditions, the Q_{01} of asphalt does not present a uniform change law. In contrast, Q_{02} exhibits the same rule as I_3/I_1, that is, the decrease of test temperature, the increase of shear frequency, the introduction of modifiers and the aging effect of asphalt can significantly improve the nonlinear viscoelasticity of asphalt, which manifests as an increase in Q_{02}. In addition, the nonlinear degree of PU modified asphalt is stronger than that of SBS modified asphalt, which also verifies the previous conclusion. To conclude, as far as the experimental data

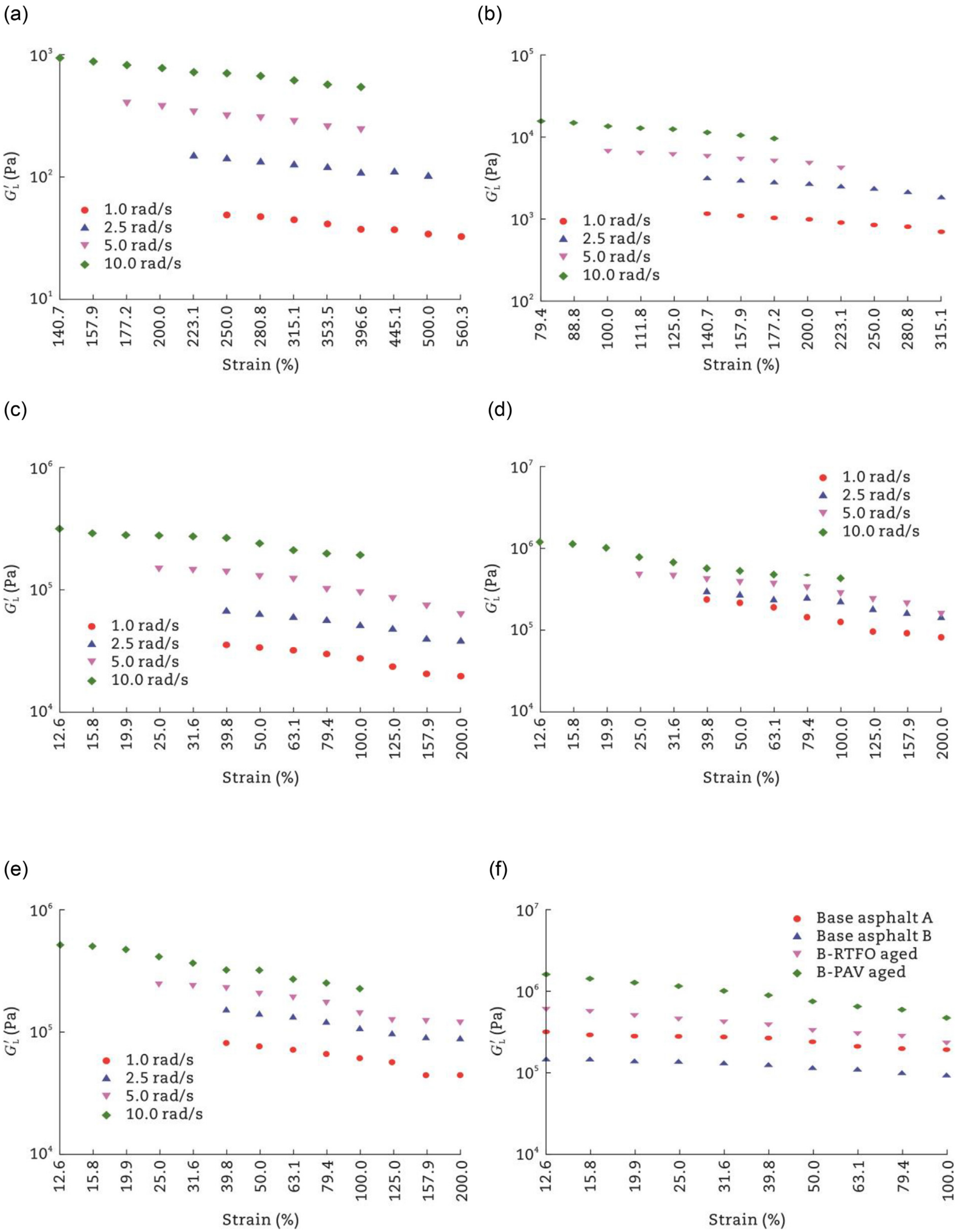

Fig. 7. G'_L results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

collected based on the samples in this study is concerned, the zero-strain nonlinear coefficient Q_0 obtained based on the average value method can accurately characterize the nonlinear viscoelasticity of asphalt, while the Q_0 obtained based on the special value method is not ideal.

3.3. Analysis results of Lissajous curve geometry

Fig. 6 presents the Lissajous curves of base asphalt A, PU and SBS modified asphalts at the test temperature of 24 °C, the shear frequency

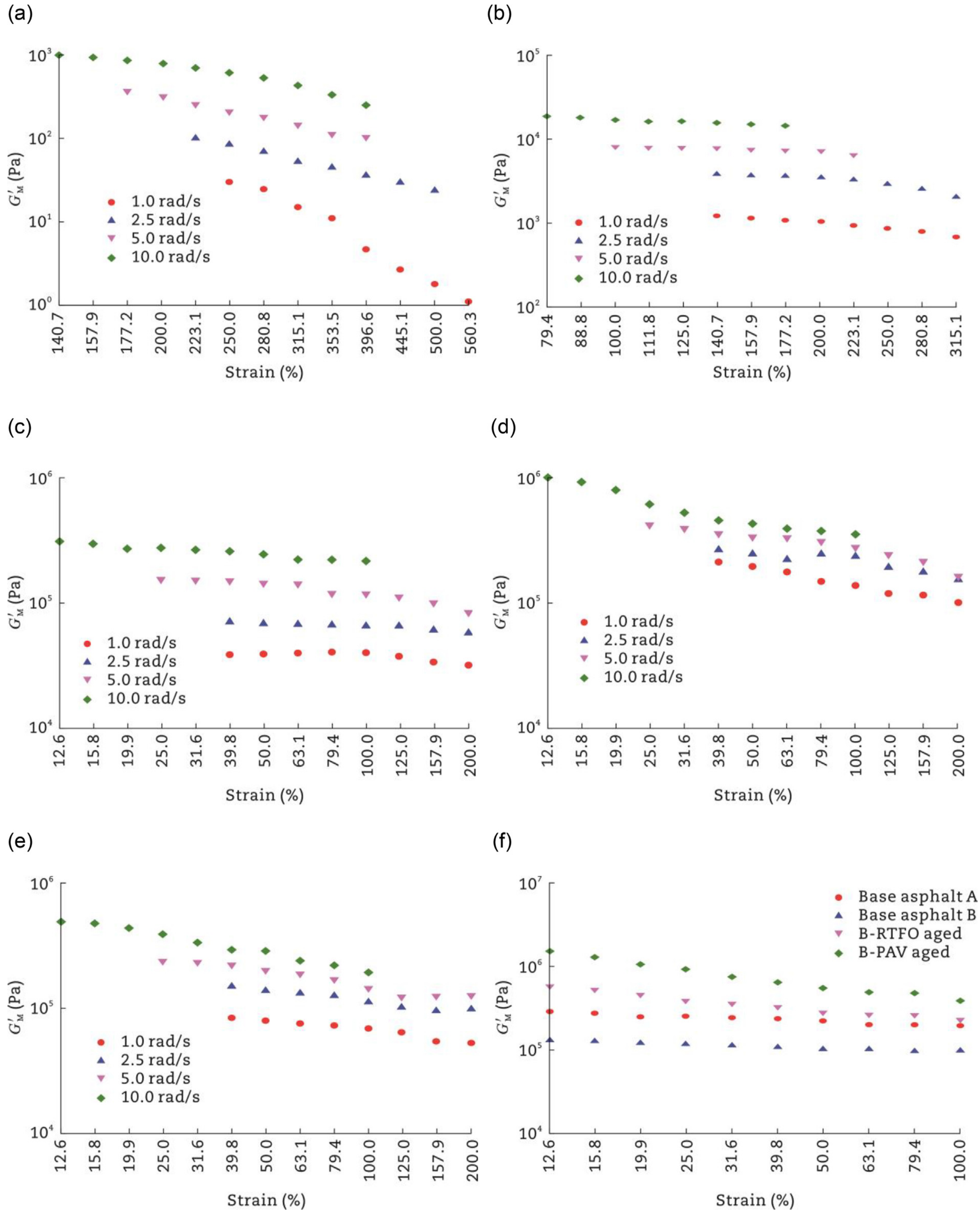

Fig. 8. G'_M results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

of 10 rad/s, and different strain levels. From Fig. 6, the Lissajous curve of asphalt exhibits significant distortion with increasing strain. The elastic Lissajous curve gradually transforms from an ellipse to an ellipse close to a parallelogram, and the viscous Lissajous curve gradually transforms from a flat ellipse to an S-shaped twisted ellipse, indicating that asphalt has significant nonlinear rheological characteristics at this point. Compared with the Lissajous curves of base asphalt A, it is found that the addition of modifiers aggravates the

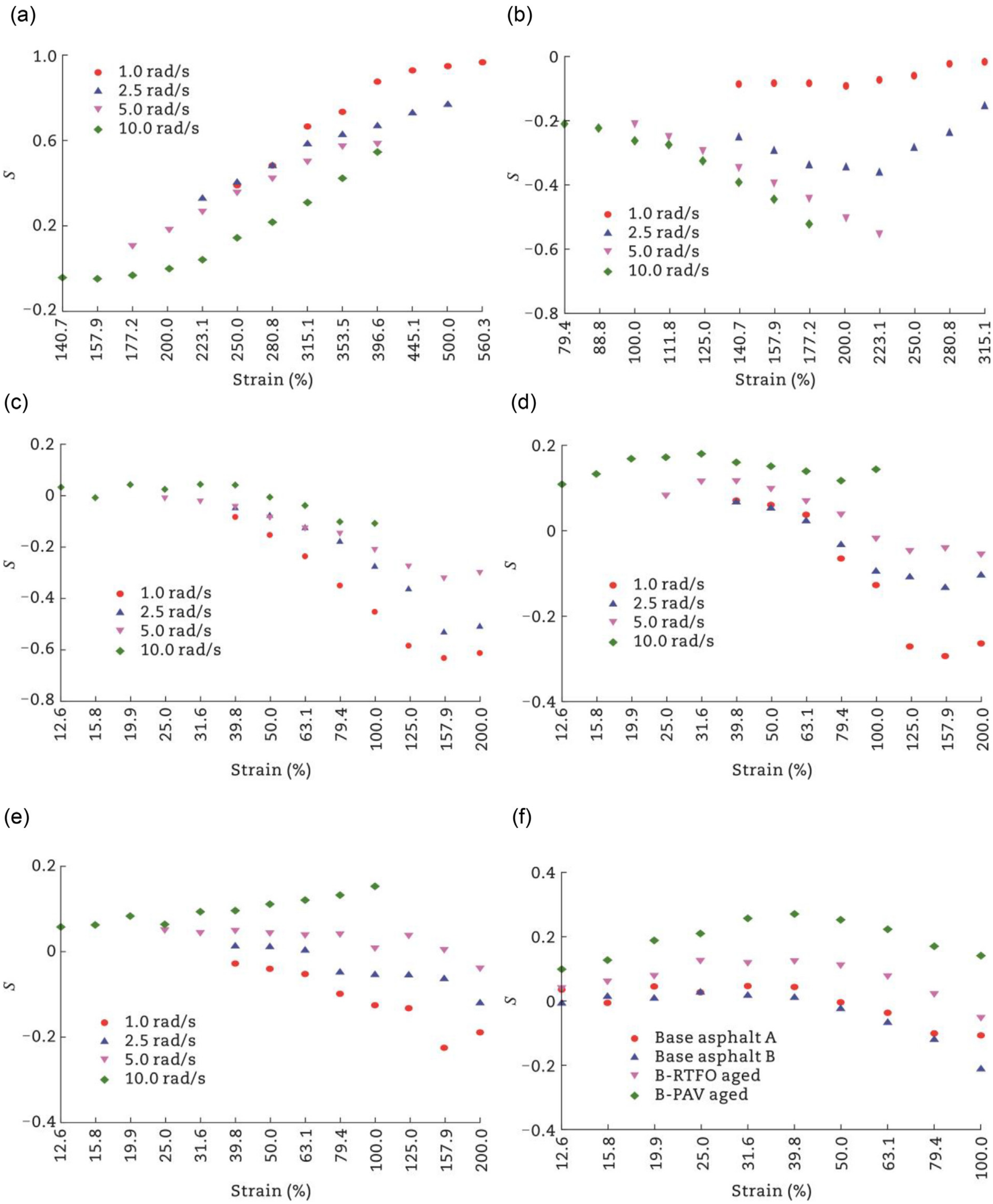

Fig. 9. *S* results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

distortion of the Lissajous curve of asphalt, in which the elastic Lissajous curve is closer to a rounded parallelogram, and the viscous Lissajous curve is more S-shaped distorted ellipse. Among the two modifiers, the effect of SBS is much more pronounced. However, as explained above, Lissajous curve geometry method can only qualitatively and cannot quantitatively characterize the nonlinear rheological properties of asphalt. Therefore, the following will discuss in detail the nonlinear rheological behavior of asphalt through Lissajous curve parameters method.

3.4. Analysis results of Lissajous curve parameters

3.4.1. Results of G'_L

Fig. 7 presents the large-strain modulus G'_L results of asphalt binders at different test conditions. For the SBS and PU modified asphalts in Fig. 7(d) and (e), the test temperature is 24 °C. For base asphalt A and base asphalt B under different aging states in Fig. 7(f), the test temperature is 24 °C, and the shear frequency is 10.0 rad/s. The remaining Lissajous curve parameters are also following this test condition.

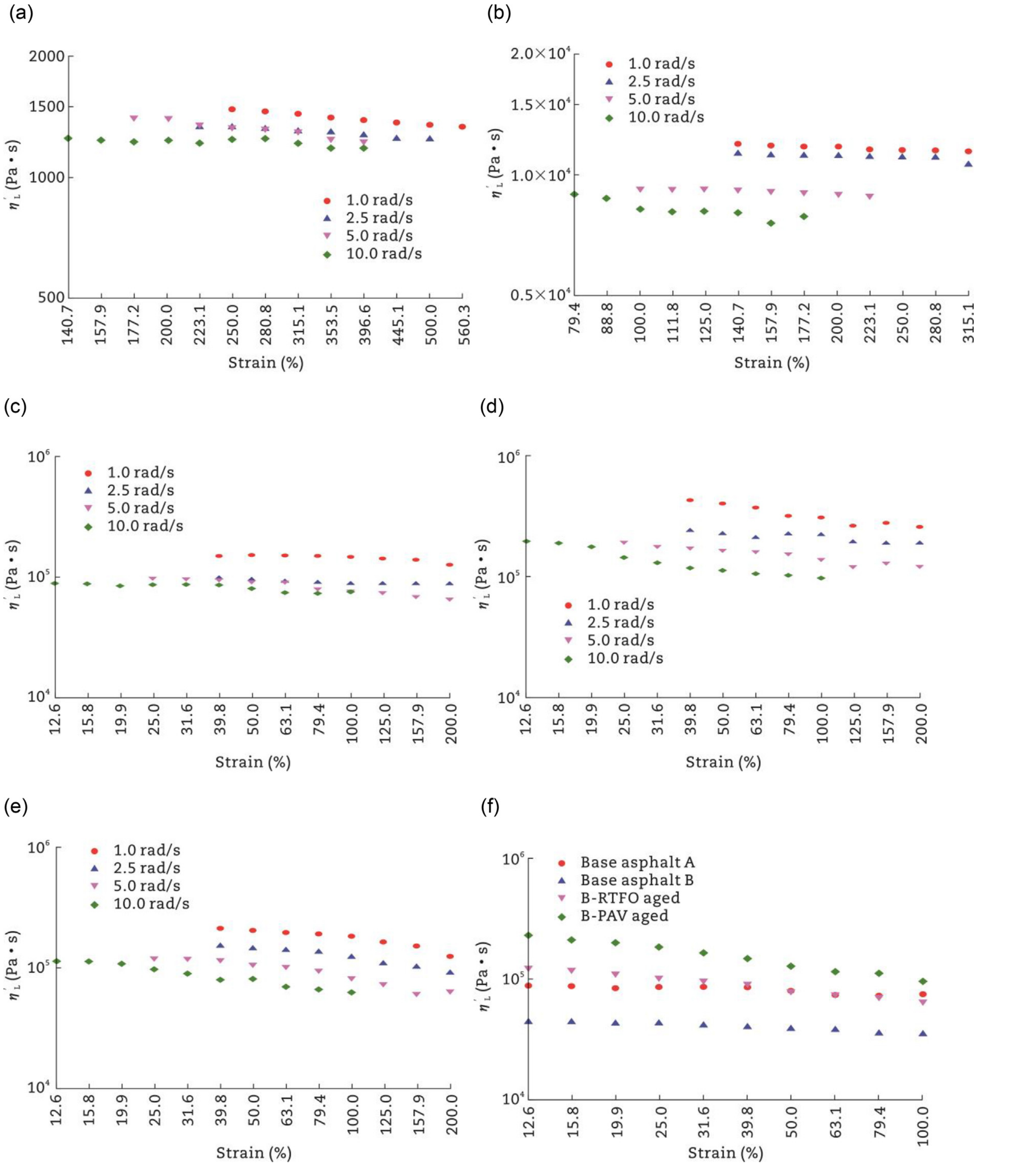

Fig. 10. η'_L results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

From Fig. 7, the rise in temperature and strain level leads to a decrease in G'_L, while the increase in shear frequency, the introduction of modifiers, and the aging effect result in an increase in G'_L. Furthermore, the improvement effect of the PU modifier on G'_L is notably higher than that of SBS, and the improvement effect of PAV aging is significantly greater than that of RTFO aging. In addition, the G'_L of base asphalt A is higher than that of base asphalt B.

3.4.2. Results of G'_M

Fig. 8 presents the minimum-strain modulus G'_M results of asphalt binders at different test conditions. It can be found that the change law of G'_M is completely consistent with G'_L, that is, the increase of temperature and strain level has a prominent negative impact on G'_M, while the increase of shear frequency, the introduction of modifiers, and the aging effect have a dramatic improvement influence. Similarly, the G'_M of base asphalt A is higher than that of base asphalt B.

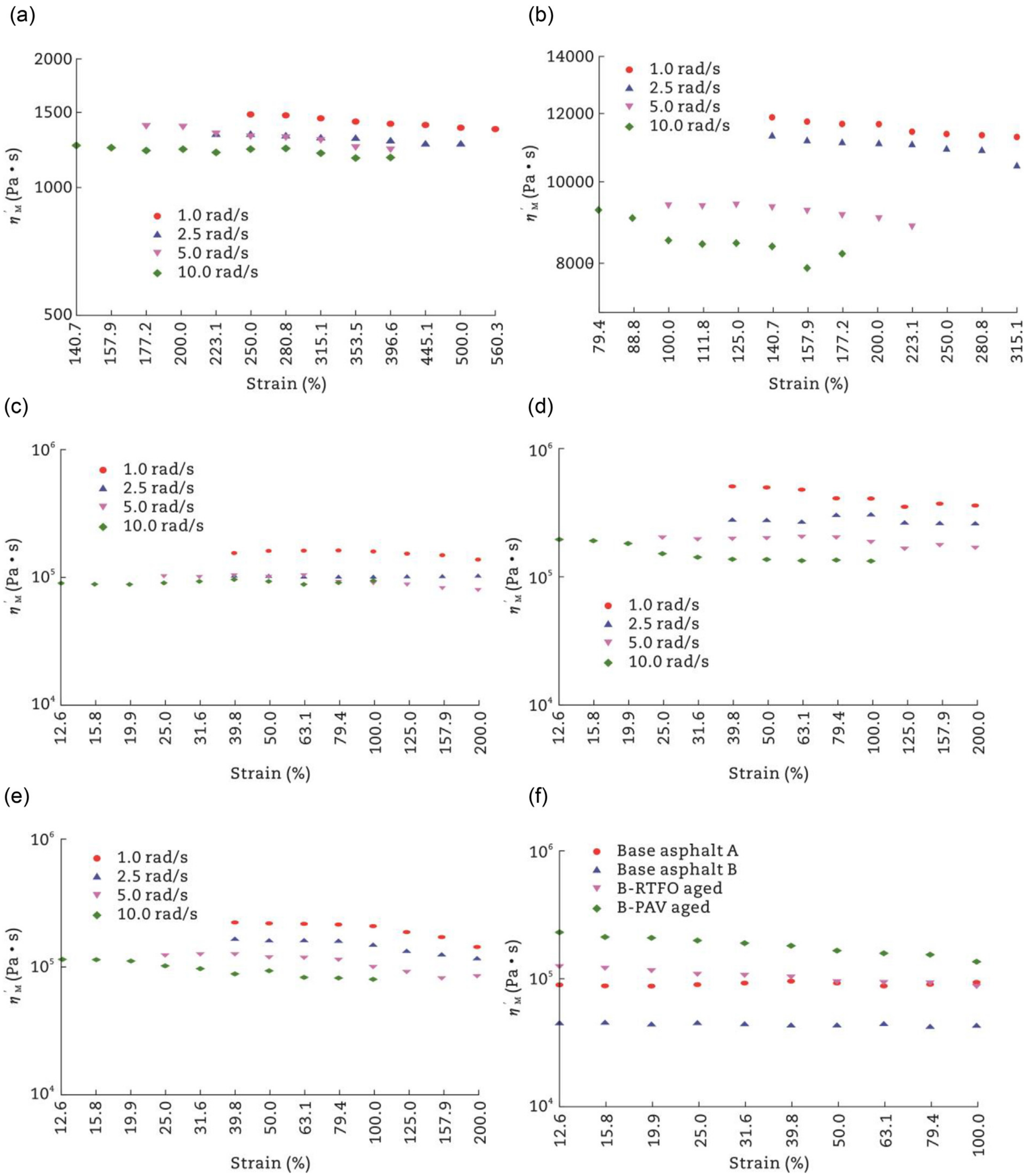

Fig. 11. η'_M results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

3.4.3. Results of S

Fig. 9 presents the strain-stiffening ratio S results of asphalt binders at different test conditions. As mentioned above, asphalt materials exhibit strain-stiffening behavior when $S > 0$ and show strain-softening behavior when $S < 0$. The S results can be divided into two categories: S results at 24 °C and S results at other test temperatures. At the test temperature of 24 °C, the increase in shear frequency, the introduction of modifiers, and the aging effect all promote the strain-stiffening behavior of asphalt, which is manifested as the higher S. On the other hand, the rise in strain level will lead to the tendency of asphalt to exhibit strain softening behavior, which is manifested as the lower S. At 48 °C, asphalt exhibits strain stiffening at all shear frequencies, and the S at this point increases with the decrease in shear frequency and the improvement in strain level. At 36 °C, asphalt exhibits strain softening at all shear frequencies, and the S at this point also increases with a decrease in shear frequency. Overall, an increase in testing temperature will promote the transition of asphalt from strain-softening behavior to strain-stiffening behavior. In addition, the S results of the two kinds of base asphalt are very close, which once again confirms that the source of the asphalt does not significantly change the nonlinear rheological characteristics under the same penetration grade.

3.4.4. Results of η'_L

Fig. 10 presents the large-rate dynamic viscosity η'_L results of asphalt binders at different test conditions. From Fig. 10, the rise in test

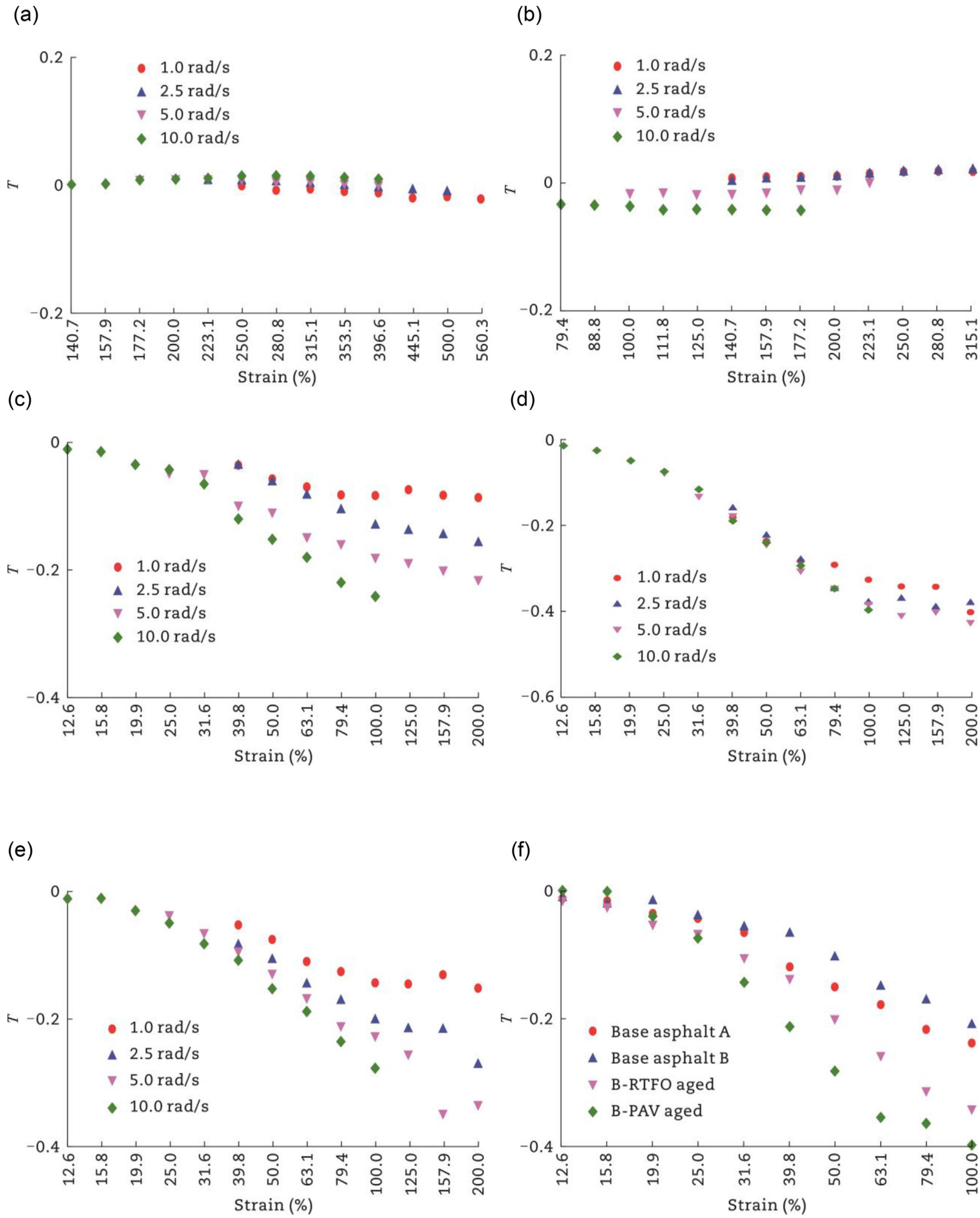

Fig. 12. *T* results of asphalt binders. (a) Base asphalt A at 48 °C. (b) Base asphalt A at 36 °C. (c) Base asphalt A at 24 °C. (d) PU modified asphalt. (e) SBS modified asphalt. (f) Base asphalt B under different aging states.

temperature, shear frequency, and strain level leads to a decrease in η'_L. The introduction of modifiers and the aging effect contribute to an increase in η'_L. Besides, the improvement effect of the PU modifier on η'_L is notably higher than that of SBS, and the lifting effect of PAV aging is observably greater than that of RTFO aging, which is consistent with the results above. In addition, the η'_L of base asphalt A is higher than that of base asphalt B.

3.4.5. Results of η'_M

Fig. 11 presents the minimum-rate dynamic viscosity η'_M results of asphalt binders at different test conditions. According to Fig. 11, the variation pattern of η'_M and η'_L is completely consistent, that is, the increase in testing temperature, strain level, and shear frequency has a distinctly negative impact on η'_M, while the introduction of modifiers and aging effect have a remarkable improvement effect on η'_M. In addition,

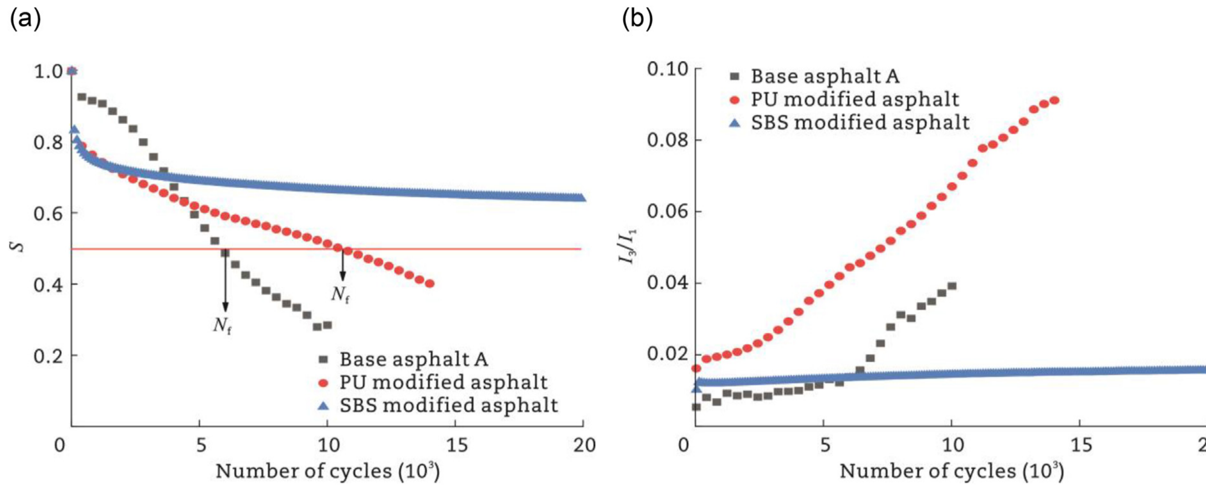

Fig. 13. Fatigue test results of asphalt based on LAOS test. (a) Fatigue results. (b) Nonlinearity results.

the influence of the PU modifier is observably greater than that of SBS, and the effect of PAV aging is more significant than that of RTFO aging. Similarly, the η'_M of base asphalt A is higher than that of base asphalt B.

3.4.6. Results of shear-thickening ratio T

Fig. 12 presents the shear-thickening ratio T results of asphalt binders at different test conditions. As mentioned above, $T > 0$ represents the shear-thickening behavior of asphalt, and $T < 0$ represents the shear-thinning behavior. From Fig. 12, all asphalts under the test temperature of 24 °C exhibit the shear thinning behavior, i.e., $T < 0$. Furthermore, the decrease in test temperature, the increase in shear frequency and strain level, the introduction of modifiers, and the aging effect all intensify the shear thinning behavior of asphalt, which is manifested as the lower T. While at the test temperatures of 36 °C and 48 °C, the T value of asphalt fluctuates around 0 with changes in the shear frequency and strain level. The effect of the PU modifier on T is also much greater than that of SBS. In addition, the T results of two kinds of base asphalt are adjacent, which is consistent with the S results above.

3.5. Relationship between the nonlinearity and fatigue performance of asphalt

To establish the relationship between the nonlinearity and fatigue performance of asphalt preliminarily, this study developed a fatigue test of asphalt based on the LAOS test. According to the results of the strain sweep test above, the critical strain of the linear region and the nonlinear region of base asphalt A is 12.6% when the temperature is 24 °C and the shear rate is 10.0 rad/s. Therefore, the strain level of 20%, the test temperature of 24 °C, and the shear rate of 10.0 rad/s were selected as the representative test condition to conduct the LAOS test. Besides, it is worth noting that this study attempts to establish the relationship between the nonlinearity and fatigue performance of asphalt based on this testing condition as an example. More investigation into the impact of different testing conditions will be conducted in future studies. The fatigue performances of base asphalt A, PU modified asphalt, and SBS modified asphalt were compared.

Fig. 13 presents the fatigue test results of three asphalts based on LAOS test, and the fatigue life of asphalt is indicated as N_f. It is found that the complex shear modulus of asphalt decreases with the application of cyclic load. Generally, the number of loads corresponding to the initial complex modulus decrease by 50% is called the fatigue life of asphalt (Wang et al., 2016). From the figure, the modulus of SBS modified asphalt still did not drop to 50% after 20,000 cyclic loads. SBS modified asphalt obviously exhibits the best fatigue performance, followed by PU modified asphalt, and the worst is base asphalt A. In addition, as the number of cyclic loading increases, S decreases gradually, and I_3/I_1 increases gradually, which indicates that the nonlinearity of asphalt will increase during the fatigue failure process.

4. Conclusions

In this paper, the LAOS test has been introduced, and the Fourier transform rheology, the Lissajous curve method, and the LAOS fatigue test have been applied to investigate the nonlinear rheological behavior of asphalt materials under different loading, aging, and modification conditions. The following conclusions can be drawn from this study.

(1) The decrease in test temperature, increase in shear frequency and strain level, introduction of modifiers, and aging effect of asphalt all signally rise the nonlinearity of asphalt. The effect of long-term aging is observably greater than that of short-term aging. Under the same penetration grade, the origin of the asphalt will not markedly change the nonlinear rheological characteristics.
(2) The zero-strain nonlinear coefficient Q_0 estimated based on the average value method can accurately characterize the nonlinear viscoelasticity of asphalt, which exhibits a completely consistent pattern with I_3/I_1 and can serve as an effective supplement to I_3/I_1. As far as the asphalt materials selected in this study is concerned, the Q_0 obtained based on the special value method is not ideal.
(3) All asphalt under the test temperature of 24 °C exhibits shear thinning behavior. The decrease in test temperature, the increase in shear rate and strain level, the introduction of modifiers, and the aging effect of asphalt all exacerbate the shear thinning behavior of asphalt.
(4) The fatigue failure process of asphalt is accompanied by an increasing degree of nonlinearity. 4% PU modifier exhibits a higher nonlinear lifting effect and a lower fatigue performance improvement effect than 4% SBS after adding to asphalt.

More studies such as the correlation between the nonlinear rheological properties and performance indicators of asphalt and the field performance of asphalt mixtures, will be further addressed in subsequent research.

CRediT authorship contribution statement

Jiaqiu Xu: writing–original draft, methodology, data curation, conceptualization. **Zijia Wang:** writing–original draft, investigation. **Zepeng Fan:** investigation, formal analysis, data curation. **Junfu Liu:** methodology, conceptualization. **Guoyang Lu:** writing–original draft, methodology, conceptualization. **Dawei Wang:** writing–review & editing, supervision, methodology.

Declaration of competing interest

Dawei Wang is a contributing editor and Guoyang Lu is a young academic editor of Journal of Road Engineering, and they were not involved in the editorial review or the decision to publish this article. The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Zijia Wang is currently employed by Civil Aviation Research Base (Beijing) Co., Ltd.

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