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**Abstract:** High content SBS polymer modified asphalt (HCPMA) can serve as pavements under heavy traffic and a wide range of temperatures. However, the applicability and validity of standard rheological evaluation methods to characterize HCPMA are still unclear. In this study, the influence of SBS content on the conventional properties and rheological behavior of HCPMA was analyzed. A higher content of SBS can improve the performance grade of the asphalt binder to PG100-34. The slope of the  $J_{nr}$ - $\sigma$  linear curves from the MSCR under various stress levels decreases when the SBS content increases. The slope of the  $J_{nr}$ - $\sigma$  linear curves can replace  $J_{nrdiff}$  as an indicator of stress sensitivity. A higher content of SBS can also decrease the flexural creep stiffness and increase the creep rate of the binders. The binder fatigue resistance parameter increases and the binder yields at higher strain, with increasing SBS content. These results show that a higher content of SBS can further improve the resistance to rutting, thermal cracking, and fatigue. Current standard rheological methods should be modified when evaluating HCPMA. The results also show that conventional tests are not valid for evaluating the performance of HCPMA.

**Keywords:** high content SBS polymer modified asphalt; multiple stress creep and recovery; performance grade; stress sensitivity; linear amplitude sweep

## 1. Introduction

Pavements for extremely heavy traffic, open-graded friction course (OGFC), and long-span steel bridge decks are subject to higher traffic loads and frequencies, larger deformations, and a wider range of service temperatures than usual roads [1]. Therefore, high-performance pavement materials are in need.

Styrene-butadiene-styrene tri-block copolymer (SBS) is one of the most efficient and widely applied asphalt modifiers. Polybutadiene (PB) blocks can increase the flexibility of asphalts at low temperatures, and polystyrene (PS) blocks can improve the elasticity of asphalts at high temperatures, which means that SBS can improve both the rutting and thermal-cracking resistance of asphalts. Due to the partial solubility and compatibility between SBS and asphalt, SBS-modified asphalts consist of two phases, i.e., polymer-rich and asphaltene-rich phases, allowing the modified asphalts to maintain the basic structure and physical characteristics of both SBS and asphalts. In conventional SBS-modified asphalt, the SBS-rich phase disperses in the continuous asphaltene-rich phase. By increasing the SBS content, phase inversion occurs and a continuous polymer-rich phase where the asphaltene-rich phase is dispersed can be obtained [2]. This is essential because the rheological properties of modified asphalt can significantly reflect those of SBS [3]. The SBS content of conventional modified asphalt is approximately 3~5% by asphalt weight, while that of high content SBS polymer modified asphalt (HCPMA), such as high-viscosity modified asphalt for OGFC, is higher than 6% [4,5]. Although the initial construction cost is slightly



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**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). higher, pavements with HCPMA can maintain an excellent service level for a longer time, reduce the frequency of maintenance and repair, and save costs throughout its life cycle.

Increasing the SBS content can bring better performance while facing some challenges, such as insufficient compatibility between SBS and asphalt and unacceptable high melt viscosity, which would result in a risk of hot-storage instability and low workability [6]. HCPMA does not exhibit the expected performance unless these problems are solved. Improving the compatibility between SBS and asphalt, increasing the mixing time and temperature to obtain a uniform dispersion, and decreasing the melt viscosity are crucial for achieving hot-storage stability and workability. Choosing SBS with a linear structure [7–10], smaller molecular weight [11], and higher diblock percentage, while adding a compatibilizer can improve the compatibility between SBS and asphalt. Masson et al. [12] found that the PB block of SBS was swollen by the alkanes of asphalts via differential scanning calorimetry. Saturate-rich or aromatic-rich oils [13] and resins [14] can act as compatibilizers. Oil can also decrease the melt viscosity of SBS-modified asphalt. Yin et al. [15–17] found that naphthenic oil assisted SBS to become swollen in asphalt and improved the low-temperature properties of the modified asphalt. Naphthenic oil contains fewer polycyclic aromatic hydrocarbons (PAHs) than aromatic oil, and thus it is more environmentally friendly [18].

Effective and accurate evaluation methods are of vital importance for material research, development, and construction quality control, which should be validated based on the relationship between material properties and actual pavement performance. Empirical and rheological tests are widely used in research and the paving industry. Empirical tests based on experience are not directly related to performance characteristics, while rheological tests attempt to directly relate the measured results of binders to roadway performance [19,20]. Rheological tests are represented by the Superpave performance grading (PG) system developed in the Strategic Highway Research Program (SHRP). However, the PG system was initially developed based on a study of neat asphalt and then improved based on conventional polymer modified asphalts. Further research is required to characterize HCPMA. Studies on the applicability of HCPMA can further help obtain a better understanding of the mechanism and make more universal improvements.

In this paper, asphalts modified with various SBS contents were prepared using naphthenic oil as a compatibilizer. The properties of HCPMA were determined using conventional binder tests and rheological analyses. The aim of this study was to quantify the differences in the rheological behavior of HCPMA with various SBS contents and to analyze the applicability and validity of parameters in standard rheological evaluation methods to characterize HCPMA.

## 2. Materials and Methods

# 2.1. Materials

Base asphalt (Donghai 70#) was produced by Sinopec Group (Shanghai, China) and its basic properties are shown in Table 1. The fractions of base asphalt were analyzed using thin-layer chromatography with flame ionization detection (TLC-FID, Iatroscan MK-5s). The naphthenic oil (KN4006), produced by China National Petroleum Corporation (Karamay, China), was used as the compatibilizer and its basic properties are shown in Table 2. SBS (Grade 1301 L) was a customized type of asphalt modification with high content. The molecular weight was smaller and the diblock percentage was higher than other SBS commonly applied for asphalt modification, such as LG501 and Kraton D1101, because smaller molecule weight and higher diblock percentage of SBS could contribute to better compatibility between base asphalt and SBS, better storage stability, and lower melt viscosity of HCPMA [21]. Its basic properties are shown in Table 3 in comparison with the commonly used SBS for asphalt modification mentioned above. The determination of average molecular weights and the molecular weight distribution for naphthenic oil and SBS were performed using a Size Exclusion Chromatography (HLC-8320GPC, Tosoh Corporation, Tokyo, Japan) equipped with a Refraction Index (RI) detector, an Ultraviolet

(UV) detector at a flow rate of 0.5 mL/min, and a temperature of 40.0  $^{\circ}$ C, with sample concentration of about 0.5 mg/mL in tetrahydrofuran (THF). The properties of LG501 and Kraton D1101 were also listed to compare with 1301 L since molecular weight could be different when tested with different methods and equipment.

Table 1. Basic properties of base asphalt.

Penetration at 25 °C	Softening Point	Ductility at 10 °C		Fractio	on/%	
/0.1 mm	∕°Č	/cm	Saturates	Aromatics	Resins	Asphaltenes
68	48.0	21	9.9	53.7	17.4	19.0

Table 2. Basic properties of naphthenic oil.

Kinematic Viscosity/mm <sup>2</sup> ·s		Molecular Weight			Carbo	Carbon-Type Composition/%		
at 40 $^{\circ}$ C	at 100 $^\circ C$	$\stackrel{-}{M}_n/{ m g}{\cdot}{ m mol}^{-1}$	$\stackrel{-}{M}_w/{ m g}{\cdot}{ m mol}^{-1}$	$\overline{M}_w/\overline{M}_n$	C <sub>P</sub>	C <sub>N</sub>	C <sub>A</sub>	
52.6	5.92	460	502	1.09	49	50	1	

Table 3. Basic properties of SBS.

				Mo	olecular Weight	
SBS	S/B	Diblock/%	Architecture	$\stackrel{-}{M}_n/{ m g}{\cdot}{ m mol}^{-1}$	$\stackrel{-}{M}_w/\mathrm{g}\cdot\mathrm{mol}^{-1}$	$\overline{M}_w/\overline{M}_n$
1301 L	30/70	24.9	Linear	$1.34 imes 10^5$	$1.49  imes 10^5$	1.12
LG501 *	31/69	12.8	Linear	$1.65  imes 10^5$	$1.79  imes 10^5$	1.09
D1101 *	31/69	18.1	Linear	$1.51 \times 10^5$	$1.73  imes 10^5$	1.14

\* LG501 and Kraton D1101 were listed to compare with 1301 L.

# 2.2. Preparation of HCPMA

A content of at least 7% SBS in modified asphalt is needed to achieve a continuous polymer-rich phase [22]. The amounts of SBS added to the base asphalt were 7.5%, 9.0%, 10.5%, and 12.0% by weight, respectively (coded as HC7.5, HC9.0, HC10.5, and HC12.0).

Naphthenic oil was added to decrease the melt viscosity, thus guaranteeing the workability of asphalt-aggregate mixtures and improving the compatibility between SBS and asphalt. The different contents of naphthenic oil corresponding to the SBS content were added to control different HCPMA with similar penetration, as shown in Table 4.

Table 4. Conventional properties of HCPMA.

Item		HC7.5	HC9.0	HC10.5	HC12.0
Constitution	base asphalt (Donghai 70#)	85.0	82.0	79.0	76.0
Composition	Naphthenic oil (KN4006)	7.5	9.0	10.5	12.0
/ %	SBŜ (1301 L)	7.5	9.0	10.5	12.0
Penetration at 2	5 °C/0.1 mm	73	75	78	78
Softening point	∕°C	87.0	89.0	91.5	93.5
Ductility at 5 °C	C/cm	82	79	75	75
Elastic recovery	at 25 °C/%	99.5	100	100	100
Elastic recovery at $5 ^{\circ}C/\%$		97.5	97.5	99.0	99.5
Toughness at $25 ^{\circ}\text{C/N} \cdot \text{m}$		18.1	17.5	16.7	15.2
Tenacity at 25 °C/N⋅m		15.8	15.4	14.3	13.2
Brookfield viscosity at 135 °C/Pa·s		1.34	1.56	2.34	2.96
$\Delta T_{R\&B}/^{\circ}C$	$\Delta T_{R\&B}/^{\circ}C$		1.5	1.5	2.0
RTFOT Residue					
Mass change/%		-0.180	-0.331	-0.302	-0.591
Retained penetration at 25 °C/%		91	90	87	94
Ductility at 5 °C/cm		46	65	76	72

The base asphalt was heated to  $180 \degree C$ , and then naphthenic oil and SBS were added to the asphalt. Subsequently, the blend was heated to  $190 \degree C$  and sheared at 5000 rpm for two hours. After that, the samples were stirred at  $180 \degree C$  for another two hours.

## 2.3. Conventional Test of HCPMA

Conventional tests were performed according to ASTM D5, ASTM D36, ASTM D113-17, ASTM D6084, ASTM D5801, AASHTO T316, ASTM D7173, AASHTO T240, and ASTM D6521, respectively for penetration, softening point, ductility, elastic recovery, toughness/tenacity, Brookfield viscosity, separation tendency, rolling thin-film oven test (RTFOT), and pressurized aging vessel (PAV). As the elastic recovery at 25 °C of HCPMA with different contents of SBS showed little difference, elastic recovery at 5 °C was also tested.

An optical microscope (Leica, Wetzlar, Germany) was utilized to observe the morphology of HCPMA under a magnification of 500 times. A small droplet of the sample was placed on a glass slide and covered with a cover slip to make a thin film. The morphology images of all samples could be correctly captured by the computer.

## 2.4. High-Temperature Properties of HCPMA

The oscillation temperature sweep was performed on a dynamic shear rheometer (DSR, Discovery HR-1, TA Instruments) to obtain complex modulus (*G*\*), phase angle ( $\delta$ ), and rutting parameter ( $|G^*|/\sin \delta$ ) of asphalt binders from 52 °C to 106 °C with a step of 6 °C according to ASTM D7175. The tests were carried out with a frequency of 10.0 rad/s using 25 mm parallel plate geometry with a gap of 1000 µm at a strain in the linear viscoelastic region. Three replicates were tested for each binder and the average value was reported.

The standard multiple stress creep recovery test (MSCR) should be performed with a creep of 1.0 s and recovery of 9.0 s, with 20 cycles at 0.1 kPa stress and 10 cycles of 3.2 kPa at the high-temperature grade determined by  $|G^*|/\sin \delta$  of the original binder. The average percent recovery at 0.1 kPa  $R_{0.1}$  and the average nonrecoverable creep compliance  $J_{nr0.1}$  were calculated with the second 10 cycles. Recovery at 3.2 kPa  $R_{3.2}$ , nonrecoverable creep compliance  $J_{nr3.2}$  and the percent difference in nonrecoverable creep compliance  $J_{nrdiff}$  were also calculated. In this study, the percent difference in recovery  $R_{diff}$  was calculated to evaluate the stress sensitivity along with  $J_{nrdiff}$ . The MSCR was tested with RTFOT residue at different temperatures and stress. Three replicates were tested for each binder and the average value was reported.

### 2.5. Low-Temperature Properties of HCPMA

The flexural creep stiffness test was performed by the bending beam rheometer (BBR, Cannon, State College, PA, USA). Specimen after RTFOT and PAV was prepared by molds with the size of 127 mm  $\times$  12.7 mm  $\times$  6.35 mm (length  $\times$  width  $\times$  height). A load of 980  $\pm$  50 mN was applied to the middle of the specimen for 240 s at the required temperature with a span of 102  $\pm$  1.0 mm. The deflection was measured. The flexural creep stiffness *S* and the creep rate *m*-value of HCPMA were calculated according to ASTM D6648. Three replicates were tested for each binder and the average value was reported.

#### 2.6. Fatigue Resistance of HCPMA

The fatigue resistance of HCPMA was evaluated by the linear amplitude sweep (LAS) test. PAV residues were tested by DSR using 8 mm parallel plate geometry with a gap of 2000  $\mu$ m at 25 °C. The test was carried out by oscillation shear using a frequency sweep to determine the undamaged properties and then tested using a strain sweep by linearly increasing amplitude from 0 to 30% at 10 Hz to cause accelerated fatigue damage. The binder fatigue performance model was calculated as Equation (1), where parameters A and B were calculated according to AASHTO TP101-14. The binder fatigue performance parameter  $N_f$  could be predicted at the given maximum expected binder strain. It should be

noted that the damage at failure was defined as the damage accumulation that corresponds to the peak stress.

$$N_f = A(\gamma_{\max})^{-b} \tag{1}$$

# 3. Results and Discussion

#### 3.1. Conventional Properties of Different HCPMA

The composition and binder properties of HCPMA are shown in Table 4. The different contents of naphthenic oil were added to control the penetration of HCPMA with various SBS contents between 60 and 80 dmm. The results showed that softening point increased and ductility decreased slightly when the content of SBS and naphthenic oil increased. Elastic recovery at both 25 °C and 5 °C showed little difference and were all nearly 100%. These results illustrated their excellent elasticity with high content SBS in the modified asphalt.

The force-elongation curves of the toughness test are shown in Figure 1. For HCPMA, when the force returned to zero, the asphalt column did not break, but the tension head was pulled out from the column. This was because the cohesion in the HCPMA was stronger than the adhesion between the tension head and HCPMA. Consequently, the toughness and tenacity became smaller with the increase in SBS content.



Figure 1. Force-elongation curves of the toughness test of HCPMA.

With naphthenic oil as a compatibilizer and processing aid agent, the Brookfield viscosity at 135 °C was lower than 3.0 Pa·s and the difference in  $T_{R\&B}$  from top to bottom was less than 2 °C, which suggested that naphthenic oil could bring good workability and storage stability even when the SBS content was high.

Mass change and retained penetration after RTFOT also showed little difference. Ductility after RTFOT increased with the increase in SBS content, which indicated that the aging resistance had improved.

The optical microscope images of the HCPMA were observed, as shown in Figure 2. The HCPMA exhibited biphasic morphology. The asphaltene-rich phase was dark while the SBS-rich phase, which was swelled by the maltene of asphalt, was yellow-colored. The asphaltene-rich phase was dispersed while the SBS-rich phase was continuous, which indicated that the properties of HCPMA could markedly reflect those of SBS polymer.



Figure 2. Optical microscope images of HCPMA.

## 3.2. Oscillation Temperature Sweep

The complex modulus  $G^*$  is the ratio of the peak stress to the peak strain in oscillatory shear. Storage modulus G' describes the amount of energy stored and released elastically in each oscillation, while loss modulus G'' describes the amount of energy dissipated in each oscillation. Phase angle  $\delta$  is the phase lag between the stress and strain in oscillation and is a measure of the viscoelastic character of a material. If  $\delta$  is near 90°, the asphalt is more viscous, while if  $\delta$  is near 0°, the asphalt is more elastic. The complex modulus  $G^*$ , phase angle  $\delta$ , and rutting parameter  $|G^*|/\sin \delta$  of the original HCPMA and RTFOT residues from 52 °C to 106 °C obtained from the oscillation temperature sweep are shown in Figure 3. The complex modulus decreased while the phase angle increased with the increase in temperature. The complex modulus increased while the phase angle decreased with the increase in SBS content. The rutting parameter proved to correlate with rutting resistance. Increasing SBS content led to improving the rutting parameter, which indicated that better rutting resistance of asphalt mixtures could be expected. The increase in the rutting parameter by increasing SBS content was caused by both the decrease in the phase angle and the increase in the complex modulus. This suggested that SBS increased both elasticity and modulus.



**Figure 3.** Oscillation temperature sweep of the HCPMA. (**a**) Complex modulus; (**b**) phase angle; (**c**) rutting parameter with solid lines for original samples, and dash lines for RTFOT residues.

The complex moduli of the RTFOT residues were not strictly higher than those of the original samples. This might attribute to better aging resistance of HCPMAs, which needs further research.

The Superpave PG system limits the value of the rutting parameter to a minimum of 1.00 kPa for the original binder and to a minimum of 2.20 kPa for RTFOT residue. The upper service temperature grades of HC7.5, HC9.0, HC10.5, and HC12.0 were PG76-, PG88-, PG94-, and PG100-, respectively. However, the highest upper service temperature is 82 °C in the PG system. The upper service temperature grade was designed to be related to the average seven-day maximum pavement design temperature, which could not reach a temperature as high as 88 °C. Cheng et al. [23] and Lin et al. [24] also obtained higher upper service temperature than 82 °C by HCPMA. How to grade the HCPMAs that are out of the PG range needs further discussion. Another problem was that the interval of 6 °C was slightly coarse to describe the properties of asphalts. The critical temperature that could meet the requirement of the PG system was calculated as the true grade. The upper service temperature grade and true grade are shown in Table 5.

Sample	PG Grade	True Grade	MSCR-PG
HC7.5	PG76-22	PG79.8-26.9	PG82S-22
HC9.0	PG88-28	PG92.3-29.6	PG82S-28
HC10.5	PG94-28	PG98.7-32.4	PG82H-28
HC12.0	PG100-34	PG101.5-39.0	PG82H-34

Table 5. PG grades, true grades, and MSCR PG.

### 3.3. Multiple Stress Creep Recovery Test

MSCR has been proven to be a better method to evaluate the high-temperature performance of modified asphalt than dynamic shear as the nonlinear viscoelasticity of modified asphalt is considered [20]. According to AASHTO T350, the test should be performed at 3.2 kPa because the stress level of 3.2 kPa was considered to be close to most real traffic load conditions and showed a good correlation with rutting from the field study [25]. The specification AASHTO MP19-10 covers asphalt binders graded by performance using MSCR, which could be called MSCR-PG. The MSCR test should be performed at the upper service temperature determined by the rutting parameter of original samples, as  $J_{ur3,2}$  in MSCR-PG replaces  $|G^*|/\sin \delta$  of the RTFOT residues in former PG and  $J_{nr_{3,2}}$  of 4.0 kPa<sup>-1</sup> is considered to provide equivalent stiffness to  $|G^*| / \sin \delta$  of 2.2 kPa. The upper service temperature grade is specified as four traffic grades according to  $J_{nr3.2}$ .  $J_{nr3.2}$  is an indicator of rutting resistance. A lower  $J_{nr}$  indicates better rutting resistance and vice versa. Meanwhile,  $J_{nrdiff}$  is limited to a maximum of 75% for each traffic grade. As the upper service temperature grade is only determined by  $|G^*|/\sin \delta$  of original binders in the MSCR-PG system instead of  $|G^*| / \sin \delta$  of both the original binders and RTFOT residues, the upper service temperature grades of HC7.5, HC9.0, HC10.5, and HC12.0 changed from PG76-, PG88-, PG94-, and PG100- to PG94-, PG100-, PG106-, and PG106-, respectively. Further discussion is needed regarding the level of temperature the MSCR should be tested at when the upper service temperature is higher than 82  $^{\circ}$ C, as the HCPMAs were not taken into consideration when MSCR was developed. In this study, MSCR was tested at different temperatures for comparison. The results are shown in Figure 4.

With the increase in temperature, the recovery decreased and the nonrecoverable creep compliance increased. With the increase in SBS content, the recovery increased and the nonrecoverable creep compliance decreased. The rutting resistance increased with the decreasing temperature and increasing SBS content.

According to AASHTO MP 19-10, the appropriate traffic loading of asphalt binders can be graded as four levels, i.e., standard (S), heavy (H), very heavy (V), and extremely heavy (E), according to  $J_{nr3.2}$  in ranges of 2.0~4.0 kPa<sup>-1</sup>, 1.0~2.0 kPa<sup>-1</sup>, 0.5~1.0 kPa<sup>-1</sup>, and no higher than 0.5 kPa<sup>-1</sup>, respectively. The traffic grades of various HCPMA at different temperatures are shown in Figure 5. The temperature had a significant influence on the

traffic grade. Considering that 82 °C is the top upper service temperature grade of the MSCR-PG system, the traffic grades of HC7.5, HC9.0, HC10.5, and HC12.0 were PG82S-, PG82S-, PG82H-, and PG82H-, respectively, as shown in Table 5, regardless of the limit of  $J_{nrdiff}$ .



**Figure 4.** MSCR test of the HCPMA at different temperatures. (a)  $R_{0.1}$ ; (b)  $R_{3.2}$ ; (c)  $R_{\text{diff}}$ ; (d)  $J_{nr0.1}$ ; (e)  $J_{nr3.2}$ ; (f)  $J_{nrdiff}$ .



Figure 5. The traffic grade of various HCPMA at different temperatures.

There was no clear relationship between SBS content or temperature and  $J_{nrdiff}$ .  $J_{nrdiff}$  is requested to be no higher than 75% according to AASHTO MP19-10. However, the  $J_{nrdiff}$  of all HCPMA at every temperature was much higher than 75%. Huang et al. [26] and White [27] also found that  $J_{nrdiff}$  was much higher than 75% when the SBS content was high. It seemed that the HCPMA was very sensitive to stress, which was contrary to common sense. The stress of 0.1 kPa was not intended to represent real traffic load but to help the samples reach a steady-state condition.  $J_{nr0.1}$  of HCPMA was too low to achieve meaningful and reliable  $J_{nrdiff}$ . The high  $J_{nrdiff}$  could be explained by low  $J_{nr0.1}$  rather than high-stress sensitivity.

Though the difference in recovery seemed more orderly than that in nonrecoverable creep compliance, it was strain and recovery that determined the nonrecoverable strain simultaneously. Therefore, the difference in recovery should not be applied as the indicator of stress sensitivity. A new indicator and corresponding criterion should be developed to characterize the stress sensitivity of HCPMA.

The two stress levels applied in the standard MSCR test were arbitrarily selected to ensure that it could be completed in a short time as a specification test. Higher levels of stress were applied to research the relationship between the  $J_{nr}$  of the binders and the rutting resistance of mixtures [28,29]. Singh et al. [30] used the slope of the nonrecoverable creep compliance-stress linear curve to evaluate the stress sensitivity of binders. In this study, MSCR was performed with a creep of 1.0 s and a recovery of 9.0 s at 82 °C, with 10 cycles at each stress level of 3.2 kPa, 6.2 kPa, and 12.8 kPa. The average percent recovery R and the average nonrecoverable creep compliance  $J_{nr}$  were calculated. The linear curves of  $J_{nr}$ - $\sigma$  were fitted and the slope of the curves was applied to replace  $J_{nrdiff}$  as the indicator of stress sensitivity. The results are shown in Figure 6.



**Figure 6.** MSCR test of the HCPMA at different stress. (**a**) R; (**b**)  $J_{nr}$ - $\sigma$  curves and linear fitting with correlation coefficient.

With the increase in stress, the recovery decreased and the nonrecoverable creep compliance increased. With the increase in SBS content, the recovery increased and the nonrecoverable creep compliance decreased. The rutting resistance decreased under a higher stress level. The slope of  $J_{nr}$ - $\sigma$  linear curves decreased with increasing SBS content, which suggested that SBS contributed to less stress sensitivity of binders.

### 3.4. Flexural Creep Test

The flexural creep stiffness *S* and the creep rate *m*-value of HCPMA at different temperatures measured by BBR are shown in Figure 7. With decreasing temperature or SBS content, the flexural creep stiffness increased and the creep rate decreased. The risk of thermal cracking decreased with the increase in temperature and SBS content.



Figure 7. BBR results of various HCPMA at different temperatures. (a) Flexural creep stiffness; (b) creep rate.

*S* is requested no higher than 300 MPa and *m*-value is requested no lower than 0.300 according to AASHTO M320. When the *m*-value of HCPMA failed at a certain temperature, *S* was much lower than 300 MPa. Therefore, the *m*-value determined the lower service temperature grade of HCPMA. The *m*-value represented the relaxation capacity under the applied stress. A higher *m*-value means that the asphalt could shed the stress more quickly. However, the elasticity of SBS inhibited viscous flow at low temperatures [31], which resulted in less relaxation, thus exhibiting a relatively low *m*-value. The limit of the *m*-value perhaps underestimated the low-temperature performance of SBS-modified asphalt. More research about the correlation between the low-temperature performance of asphalt mixtures and the BBR parameters of asphalts is needed.

The critical temperature exactly at the limiting creep rate value of 0.300 was calculated by interpolating between passing and failing temperatures as the true grade. The lower service temperature grade and the true grade are shown in Table 5. With the increase in SBS content, both high- and low- temperature performance of HCPMA became better, and thus the range of performance grades became wider.

### 3.5. Linear Amplitude Sweep

The LAS test is considered an effective method for evaluating the fatigue resistance of asphalts [32–34]. According to AASHTO TP101-14, strain should be increased linearly from 0 to 30% in the amplitude test and the fatigue failure was defined as corresponding to the peak stress. In the proposed procedure, the peak stress of HC7.5, HC9.0, HC10.5, and HC12.0 was at the strain of 21.4%, 26.7%, 30.0%, and 30.0%, respectively, suggesting that HC10.5 and HC12.0 did not yield at the maximum strain of 30%. Increasing the maximum amplitude, the peak stress of HC10.5 and HC12.0 was found at the strain of 48.4% and 68.0%, as shown in Figure 8. With the increase in SBS content, HCPMA yielded a higher strain. If the maximum strain still ranged from 0 to 30% and the value of D(t) at 30% strain was determined as  $D_f$ , the value of  $D_f$  would be lower than the actual one, which would result in a lower parameter A. The  $N_f$ - $\gamma_{max}$  curve would shift downward as a whole, which means that the binder fatigue performance parameter at a given strain amplitude would be lower and the fatigue resistance of HCPMA would be underestimated. For HCPMA, a higher maximum strain should be applied to make sure that the binder yields and the peak stress is obtained.



Figure 8. Stress-strain curves of HCPMA by amplitude sweep.

Another concern is about the range of test data used to determine the fitting parameters  $C_1$  and  $C_2$ . In the specification AASHTO TP101-14, data points at up to 30% strain, excluding those corresponding to damage accumulation less than 10, should be applied to calculate  $C_1$  and  $C_2$  by fitting the relationship between C(t) and D(t). As the strain of HCPMA at peak stress may be higher than 30%, a higher maximum strain should be applied to achieve the peak stress, and test data with a strain of higher than 30% are obtained. The range of data used for calculation needs more discussion. When the LAS test was originally developed by Johnson and Bahia, the amplitude sweep was conducted with a strain of

0.1~20%. Hintz et al. [32] found that the damage accumulation was insufficient for polymer modified asphalts and using additional higher strains did not substantially affect the results. They recommended increasing the maximum strain to 30% to achieve more significant material decaying, which was adopted by AASHTO TP101-14. The HCPMA decays more slowly than the polymer modified asphalts that Hintz et al. [32] investigated. It seems that a higher maximum strain (e.g., 70%) should be adopted to evaluate HCPMA. However, the complex modulus fluctuates at a relatively high shear strain after the materials yield, as shown in Figure 9. It is hard to calculate parameters using the same high maximum strain. A more reasonable proposal is to select the range of data according to the strain at which each binder yields.



Figure 9. Complex modulus-strain curves of HCPMA by amplitude sweep.

The relative parameters were calculated in Table 6. With increasing SBS content, the parameter  $\alpha$  increased, indicating that the sensitivity of the storage modulus to the frequency of HCPMA decreased in the linear viscoelastic region. The damage intensity at failure  $D_f$  increased, suggesting that the binder could accumulate more damage before fatigue failure. The binder fatigue performance parameters  $N_f$  with various applied shear strains are shown in Figure 10. The fatigue resistance of HCPMA was improved by increasing the SBS content.

Table 6. The fffect of SBS content on the parameters of HCPMA from LAS.



Figure 10. N<sub>f</sub>-shear strain curves of HCPMA with different SBS content.

# 4. Conclusions

The rheological characterization of high content SBS polymer modified asphalt has been seldom researched. In this study, HCPMA was prepared with high content SBS using balanced naphthenic oil. The influence of the SBS content on the conventional properties and rheological behavior of binders was comprehensively investigated. The applicability and validity of current standard rheological evaluation methods on HCPMA were discussed. The conclusions were drawn as follows:

- (1) Higher content of SBS in HCPMA can improve the rutting resistance and decrease the sensitivity to temperature and stress. With the increase in SBS content, the rutting parameter increases, caused by both a decrease in the phase angle and an increase in the complex modulus. The upper service temperature grade of HCPMA can far exceed 82 °C, which is the highest temperature grade in the PG system. The creep recovery increases, while there is a decrease in the nonrecoverable creep compliance and the slope of  $J_{nr}$ - $\sigma$  linear curves. The slope of  $J_{nr}$ - $\sigma$  linear curves from various stress levels can replace  $J_{nrdiff}$  as an indicator of stress sensitivity.
- (2) Higher content of SBS can improve the resistance of thermal cracking. With increasing SBS content, the flexural creep stiffness decreases, and the creep rate increases. The flexural creep stiffness is much lower than the limit value at the temperature where the creep rate fails.
- (3) Higher content of SBS can improve fatigue resistance. With increasing SBS content, the binder yields at a higher strain, and the binder fatigue performance parameters  $N_f$  increases. When the LAS test is used to evaluate HCPMA, a maximum strain higher than 30% should be applied to obtain the peak stress.
- (4) With naphthenic oil as a compatibilizer and processing aid agent, HCPMAs exhibit excellent resistance to rutting, thermal cracking, and fatigue, as well as good workability and hot-storage stability.
- (5) Conventional tests are not valid for characterizing HCPMA. The ductility, elastic recovery, mass change, and retained penetration after RTFOT are not sensitive to SBS content when it is high. The toughness and tenacity even decrease with an increase in SBS content.

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## Abbreviations

SBS	styrene-butadiene-styrene tri-block copolymer
РВ	polybutadiene
PS	polystyrene
HCPMA	high content SBS polymer modified asphalt
PG	performance grading
RTFOT	rolling thin-film oven test
PAV	pressurized aging vessel
DSR	dynamic shear rheometer
MSCR	multiple stress creep recovery test
BBR	bending beam rheometer
LAS	linear amplitude sweep test

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