



## Weather aging resistance of different rubber modified asphalts



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### HIGHLIGHTS

- Weather aging of rubber modified asphalts was carried out under natural environment.
- Rheological properties of modified asphalts changed with the structural evolution.
- For the given base asphalt, SBSMA showed the worst weather resistance, while TB showed the best.

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### ABSTRACT

The service life of polymer modified asphalt strongly depends on its weather resistance because of its exposure to the natural environment. Three most widely used rubber modified asphalts, namely styrene–butadiene–styrene (SBS) modified asphalt (SBSMA), crumb rubber modified asphalt (CRMA) and terminal blend rubber modified asphalt (TB) are exposed outdoor for weather aging. Effect of aging on the chemical composition of different rubber modified asphalts is analyzed by using infrared spectroscopy. The rheological properties of base asphalt and modified asphalts before and after aging are characterized by using the dynamic shear rheometer (DSR) and dynamic mechanical analysis (DMA). Results show that the chemical structure and rheological properties of modified asphalts change significantly as a result of the weather aging. Different rubber modified asphalts show the different aging behavior. For the given base asphalt, SBSMA is the most susceptible to weather aging, followed by CRMA and TB, respectively.

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### 1. Introduction

Asphalt materials are exposed to the environmental factors such as sunlight, moisture, oxygen and heat that affect the properties of asphalt has been widely investigated [1–3]. The combined interaction of these factors causes asphalt being susceptible to the change of physical, chemical and rheological property, which reduces the durability of asphalt pavements [4,5].

Polymers have been considered as the most cost effective additives to improve the durability of asphalt pavements [6–9]. However, just like base asphalt (BA), polymer modified asphalts (PMAs) also suffer from aging under the action of environmental factors [10–12]. The aging of PMAs not only includes the aging of asphalt and modifiers, but also their interactions in aging process, which contributes to the deterioration of asphalt pavement, and the service life is shorten [12–14]. In terms of the aging of polymers,

the irreversible changes in polymer structure and properties caused by aging have been widely investigated [15–17].

The aging of PMAs, such as styrene–butadiene–styrene modified asphalt (SBSMA) has been taken a great deal of investigation from aspects of chemical structure and physical rheology. Lu and Issacson [18] reports that the polymer couldn't resist the formation of carbonyl group in aging process of SBSMA. The rheological properties evolution of aged modified asphalt depends on a combined effect of asphalt oxidation and polymer degradation. Ruan et al. [19] investigate the dynamic shear properties and extensional properties of PMAs, which shows that oxidative aging reduces the temperature susceptibility of asphalt, damages the polymer network in modified asphalt. Yut and Zofka [10] study the correlation between chemistry and rheology of aged PMAs through DSR and FTIR. They find that the severity of aging procedure affects the viscosity of PMA more than polymer composition and concentration. Complex modulus can be fairly estimated from chemical composition elucidated by ATR-FTIR. Wang et al. [20] evaluate the aging mechanism of SBSMA based on chemical reaction kinetics, which indicates that the activation energy of asphalt will change with the degradation of SBS modifier, and the temperature sensitivity

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of asphalt is improved. Geng et al. [21] report that the aging under thermal moisture conditions changes the properties of asphalt and the color of SBS due to the increasing carbonyl content of asphalt and SBS. The presence of water is shown to accelerate the aging of SBS and its modified asphalt under the action of heat and oxygen.

Generally speaking, SBSMA shows better aging resistance than base asphalt. However, the performance of modified asphalt decreases as the degradation of SBS in aging process. Crumb rubber (CR) has been selected as modifier for the purpose of the improved aging resistance performance and tire recycling [22,23]. Crumb rubber modified asphalts are mainly divided into two types: the traditional crumb rubber modified asphalt (CRMA), and the terminal blend rubber modified asphalt (TB). TB consists of asphalt with CR modifier, which is digested at the refinery or at asphalt terminal and then delivered to the plant [23–25].

Aging of PMAs has been investigated by using the rolling thin film oven test (RTFOT), pressurized aging vessel (PAV), and ultraviolet aging (UV). It is found that two parallel reactions occur during the course of oxidation: oxidation of base asphalts and degradation of polymers, which results in changing the compatibility between asphalt and polymer [26,27]. Due to the complexity of the CRMA, the aging mechanism of CRMA is still unclear. Ghavibazoo and Abdelrahman [28] find that the traditional crumb rubber modifier particles don't prevent base asphalt from thermal aging. Otherwise, the pavement longevity is extended largely by means of the high asphalt aggregate ratio and crack resistance of rubber. Chipps [29] and Ruan [30] analyze the effect of the long-term aging on properties of TB. They find that the aging reduces the hardening and oxidative ration of TB which has better aging resistance than BA.

Currently, the field aging behaviors of PMAs are still mainly studied by simulating the influence of environmental factors in indoor conditions [2,3,26,31,32], such as RTFOT, UV with PAV and so on. However, the aging of PMAs mainly occur outdoor. The thin layer of modified asphalts exposing to the weather conditions need to be investigated.

In this study, SBS, CR and degraded rubber (the crumb rubber digested in the asphalt under high temperature and high shear) were used to prepare three different rubber modified asphalts. Their weather aging properties of different modified asphalts were investigated in terms of the rheological properties and chemical composition. The research results will gain more insight into the aging behaviors of different rubber modified asphalts under real using conditions.

## 2. Experimental

### 2.1. Materials

Base asphalt GS 90# (Saturate, 18.3%; Aromatics, 50.0%; Resin, 32.5%; Asphaltene, 9.2%) was selected as the base asphalt. CR (40 mesh) was produced by the mechanical shredding at ambient temperature, supplied by the Jiangsu Branch Yin Gert Asphalt Co. Ltd. Linear styrene-butadiene-styrene (SBS) with molecular weight is  $1.2 \times 10^5$  g/mol with S/B (30/70), produced by Petrochemical Co., Ltd, China.

### 2.2. Preparation of the modified asphalts

CRMA was prepared in an open iron container by using a high shear mixer (made by Aidong Machine Co. Ltd., China). Base asphalt was first heated to fluid state, the stirring equipment was started with 500 rpm to heat evenly to 175 °C, and 22 wt% CR was added into asphalt matrix with a shearing speed of 5000 rpm for 40 min.

SBSMA was prepared by adding 4 wt% SBS to asphalt with 4000 rpm for 30 min at 175 °C.

TB was prepared with high shearing (3000 rpm) and high temperature (220 °C) for 2 h, subsequently, the blend was stored at 200 °C for 32 h.

The conventional properties of BA, SBSMA, CRMA and TB are given in Table 1.

### 2.3. Aging procedure

The weathering aging procedure of different rubber modified asphalts was adopted according to BS EN ISO 2810-2004 (Paints and varnishes-natural weathering of coatings exposure and assessment). Different modified asphalts were coated on Aluminum plate (200 mm × 200 mm) in thickness of 0.5 mm. Subsequently, the coated samples were placed into an oven (80 °C) to make sure the evenness of PMAs. After that, all samples exposed outdoor (the weather in Shanghai city with humid meso-thermal climates). The weather aging was carried out for 9 months, from August, 2014 to April, 2015. The aging environment conditions were shown in Fig. 1.

### 2.4. Characterization methods

#### 2.4.1. Fourier transform infrared (FTIR)

Changes in functional groups of asphalt and the modified asphalts before and after aging were conducted with Attenuated Total Reflectance Infrared Spectrometer (ATR-FTIR) (iz10, Nicolet, USA). The scan ranges from 600  $\text{cm}^{-1}$  to 4000  $\text{cm}^{-1}$  at a resolution of 4  $\text{cm}^{-1}$ .

Carbonyl group C=O (centered around 1700  $\text{cm}^{-1}$ ) and sulfoxide group S=O (centered around 1030  $\text{cm}^{-1}$ ), as well as the butadiene double bonds C=C (centered around 966  $\text{cm}^{-1}$ ) were monitored to characterize the aging degree of modified asphalts in spectra. Among them, the decrease of C=C content in SBSMA means the degradation of SBS in asphalt during weather aging (especially thermal, oxidation, UV radiation). The carbonyl index (CI), sulfoxide index (SI) and butadiene double bonds index ( $I_{\text{SBS}}$ ) were calculated by integral to evaluate the aging degree by following formulas [33]:

$$\text{CI} = A_{\text{C=O}}/A, \text{SI} = A_{\text{S=O}}/A, I_{\text{SBS}} = A_{\text{SBS}}/A$$

where the C=O ranges from (1690  $\text{cm}^{-1}$  to 1710  $\text{cm}^{-1}$ ); S=O (1020  $\text{cm}^{-1}$  to 1034  $\text{cm}^{-1}$ ); A (700  $\text{cm}^{-1}$  to 2000  $\text{cm}^{-1}$ );  $I_{\text{SBS}}$  (940  $\text{cm}^{-1}$  to 990  $\text{cm}^{-1}$ ).

This carbonyl indices for BA and modified asphalts in production (carbonyl content of modified asphalt was prepared later subtracting the carbonyl content of base asphalt) and aging process was analyzed.

#### 2.4.2. Dynamic shear rheometer (DSR)

High temperature rheological characterization of BA and modified asphalts was carried out by using a dynamic shear rheometer (rotary rheometer, Gemini 200HR, Bohlin Instruments, UK) configured in parallel plate geometry. Temperature sweep tests were conducted in constant-strain oscillation mode at a fixed frequency of 10 rad/s. The temperature ranged from 46 °C to 86 °C with an increment of 3 °C/min. The parallel plate of 25 mm diameter with a gap width of 1 mm between the two plates was used. The constant strain was 1%. Rheological parameters such as complex modulus ( $G^*$ ) and phase angle ( $\delta$ ) as a function of temperature for all samples were applied to evaluate the rheological properties of asphalts in aging process.

#### 2.4.3. Dynamic mechanical analysis (DMA)

Dynamic mechanical analysis (DMA) was implemented with a dynamic mechanical analyzer (Q800, TA instruments, USA). Samples were cut into size of  $22 \times 5 \times 2$  mm<sup>3</sup>. The storage modulus  $G'$  and  $\tan \delta$  was measured at a frequency of 10 Hz with a strain of 0.01% in tensile mode. The temperature ranged from –50 °C to 5 °C at a heating rate of 3 °C/min.

## 3. Results and discussion

### 3.1. Rheological properties of different PMAs at high temperature

Different asphalts show different aging behavior because of the different aging sensitivity of molecular structures. The weather aging will cause the composition evolution of different asphalts, which changes the viscoelastic properties of asphalts. The moduli of asphalts are sensitive to temperature. Therefore, complex modulus ( $G^*$ ) and phase angle ( $\delta$ ) of BA and different PMAs before and after aging are applied to evaluate the rheological properties.

**Table 1**

Properties of base asphalt and different modified asphalts.

Properties	BA	SBS MA	CRMA	TB	Standard
Penetration (0.1 mm)	94.0	67.4	54.9	69.4	ASTM D5
Softening point (°C)	49.2	63.4	58.9	53.6	ASTM D36
Ductility (5 °C)/cm	0	21.3	8.5	11.7	ASTM D113

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