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Rheological and Aging Properties of Ultraviolet Absorber/Styrene-Butadiene-Styrene-Modified Bitumens

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ABSTRACT: The prevention of the ultraviolet (UV) aging of styrene–butadiene–styrene (SBS)-modified bitumen is of great significance because UV aging is considered to be a critical factor in the deterioration of the performance of bituminous roads. In this study, we selected two UV absorbers (octabenzone and bumetrizole) as modifiers to evaluate their effects on the rheological and UV-aging properties of different SBS-modified bitumens (linear and radial types) by means of dynamic shear rheological testing. The thermooxidative aging of the binders was also investigated through thin-film oven testing. The results show that for the linear- and radial-SBS-modified bitumens, octabenzone only enhanced their elasticity at high temperatures, but bumetrizole could increase the elastic response at intermediate temperatures on the one hand and the viscous response at high temperatures on the other hand. The UV aging of the radial-SBS-modified bitumens could effectively be alleviated by both octabenzone and bumetrizole, whereas that of the linear ones could only be improved by bumetrizole. The influence of the UV absorbers on the thermooxidative-aging properties of the SBS-modified bitumens was dependent on the types of UV absorbers and SBS polymers. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 128: 2571–2577, 2013

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INTRODUCTION

It is well known that styrene–butadiene–styrene (SBS)-modified bitumen has been a preferred choice in high-performance-grade pavements because of its excellent mechanical and rheological properties.[1] However, the performance of SBS-modified bitumen may evolve with aging as a result of the degradation of the SBS polymer and the oxidation of bitumen.[2,3] Generally, the aging of SBS-modified bitumen has two aspects: thermooxidative aging, which mainly happens in the processes of mixing and the laying of bituminous mixtures, and photooxidative aging during the service life of a pavement.[4–6] Both thermooxidative and photooxidative aging can cause the deterioration of the SBS-modified bitumen and can thereby result in destruction of the pavement.

The thermooxidative and photooxidative aging of SBS-modified bitumen has been investigated by many researchers in recent years, but fewer researchers have paid attention to the improvement of aging resistance in SBS-modified bitumen, especially the improvement of photooxidative-aging resistance.[2–8] It is believed that modification with additives is an effective way to protect a binder from aging.[9–11] For example, antioxidants such as zinc dialkyl dithiophosphate and zinc dibutyl dithiocarbamate have been confirmed to be able to retard the thermal oxidation of SBS-modified asphalt through the inhibition of peroxides and the scavenging of radicals.[9] Layered silicates, including sodium montmorillonite and organophilic montmorillonite, can also efficiently improve the thermooxidative-aging resistance of SBS-modified asphalt.[10] Also, Zhang et al.[11] found that the ultraviolet (UV) aging of SBS-modified bitumen could be prevented to some extent by the existence of organophilic montmorillonite.

For years, UV absorbers have been selected as anti-aging additives to protect most polymers from photooxidative degradation; their use has led to excellent improvements in the aging resistance of polymeric materials.[12–14] In our previous research, several types of UV absorbers were used to modify base bitumens. The results revealed that the addition of UV absorbers not only significantly affected the aging properties of the bitumens but also had obvious effects on their mechanical and rheological properties.[15,16] However, it is worth mentioning that the method related to the modification of the photooxidative-aging resistance of SBS-modified bitumen with UV absorbers has rarely been reported thus far.

In this study, two UV absorbers that are commonly used in the prevention of the photooxidative aging of other polymers (e.g., plastic, rubber, coatings) were selected as modifiers to evaluate their effects on the rheological and UV-aging properties of different SBS-modified bitumens by means of dynamic shear rheometry (DSC) tests. The thermooxidative aging of the UV absorber/SBS-modified bitumens was also investigated because thermooxidative aging is inescapable for all binders during their application. Also, the influence of the UV absorbers on the physical properties and storage stability of the SBS-modified bitumens was studied to evaluate the use of these modified materials on roads.

EXPERIMENTAL

Materials

An 80/100 penetration-grade base bitumen was used in this study. The physical properties of the base bitumen were as follows: penetration = 89 dmm (25°C), ductility > 150 cm (15°C), softening point = 46.1°C, and viscosity = 0.29 Pa s (135°C).

Two types of SBS were used, those with linear and radial structures, respectively. The average molecular weight of the linear SBS was 110,000 g/mol. The average molecular weight of the radial SBS was 350,000 g/mol. The block ratio of both the linear and radial SBS polymers was 30 : 70. The sulfur, a bright yellow crystalline solid, was a commercial product.

The two UV absorbers used were octabenzone and bumetrizole. The physical properties of the octabenzone were as follows: melting point = 48°C and light transmittance = 82% (450 nm) and 91% (500 nm). The physical properties of the bumetrizole were as follows: melting point = 140°C and light transmittance = 85% (450 nm) and 92% (500 nm).

Preparation of UV Absorber/SBS-Modified Bitumen

The UV absorber/SBS-modified bitumen was prepared by melt blending. First, the base bitumen was heated to 180°C until it became fluid. Second, a suitable content of SBS (3.5 wt %) was slowly added to the base bitumen and sheared for 1 h with a high-speed shearing mixer. The shearing temperature was 180°C, and the shearing rate was 4000 rpm. Finally, the UV absorber (0.6 wt %) and sulfur (0.1 wt %) were mixed with the blend at 180°C with a mechanical stirrer. After the mixtures were stirred for 2 h, the UV absorber/SBS-modified bitumens were obtained. To compare with the UV absorber/SBS-modified bitumens preferably, the SBS-modified bitumens without UV absorbers were also treated by the same method.

The samples were denoted as follows: linear styrene–butadiene–styrene modified bitumen (LSMB), radial styrene–butadiene–styrene modified bitumen (RSMB), octabenzone/linear styrene–butadiene–styrene modified bitumen (OLSMB), bumetrizole/linear styrene–butadiene–styrene modified bitumen (BLSMB), octabenzone/radial styrene–butadiene–styrene modified bitumen (ORSMB), and bumetrizole/radial styrene–butadiene–styrene modified bitumen (BRSMB).

Aging Procedures

The thin-film oven test (TFOT) was performed according to the standard ASTM D 1754/D1754M-09 before UV aging. The residue from the TFOT was placed into a UV irradiation oven to undergo the UV aging. The UV lamp was 500 W with a main wavelength of 365 nm, and the average intensity of UV radiation on the sample surface was about 1400 $\mu W/cm^2$. The thickness of the bitumen film was about 2 mm. The temperature of the UV aging was 60°C, and the UV aging cycle was 6 days.

DSR Testing

The dynamic rheological properties of the binders before and after aging were measured by a dynamic shear rheometer (model MCR101, Anton Paar Co., Graz, Austria). The binders were first heated until they were sufficiently fluid. Then, the hot binders were poured into a silicone rubber mold and cooled to room temperature. We removed the bitumen disk from the silicone mold and placed it on the lower plate (25 mm in diameter) of the DSR device. After the plate was warmed so that a good adhesion was achieved between the plate and the binder, the gap between the top and bottom plates was gradually reduced to the target value plus 50 μm. The specimen squeezed between the plates was then trimmed flush with the edge of the plate. After trimming, the final adjustment was made to the designated gap (1 mm). The DSR test was performed under the strain-controlled mode. The angular frequency was set at 10 rad/s. The range of sweeping temperature was 30–90°C, and the temperature increment rate was 2°C/min.

Physical Properties Testing

The penetration (at 25°C), ductility (at 5°C), softening point, and viscosity (at 135°C) were measured in accordance with the standards ASTM D 5-06e1, ASTM D 113-07, ASTM D 36/D36M-09, and ASTM D 4402-06, respectively.

Ultraviolet–Visible (UV–Vis) Spectroscopy

The absorbance and reflectance of the UV absorbers were measured with a UV–vis spectrophotometer (UV2550, Shimadzu Corp., Shimane, Japan). $BaSO_4$ was used as a standard in the UV–vis experiment. The wavelength range was selected within the UV region of 220–400 nm.

Storage Stability Testing

The samples were poured into an aluminum foil tube with a diameter of 32 mm and a height of 160 mm. We sealed the tube and put it into the oven vertically. After it was stored in the oven at 163°C for 48 h, the tube was taken out and cooled to ambient temperature. We cut the tube into three equal sections and determined the softening point of the top and bottom sections. The difference of the softening point between the top and bottom sections represented the storage stability of the binders.

RESULTS AND DISCUSSION

Dynamic Rheological Properties

Temperature Dependence of the Phase Angle (δ). δ reflects the viscoelastic behavior of the binders.[17] The δ versus temperature graphs of the different UV absorber/SBS-modified bitumens before aging are displayed in Figure 1. Octabenzone showed little influence on the viscoelastic properties of LSMB before aging, as indicated by the similar values of δ in the temperature range from 30 to 65°C. At temperatures higher than 65°C, the δ values of OLSMB were lower than those of LSMB; this

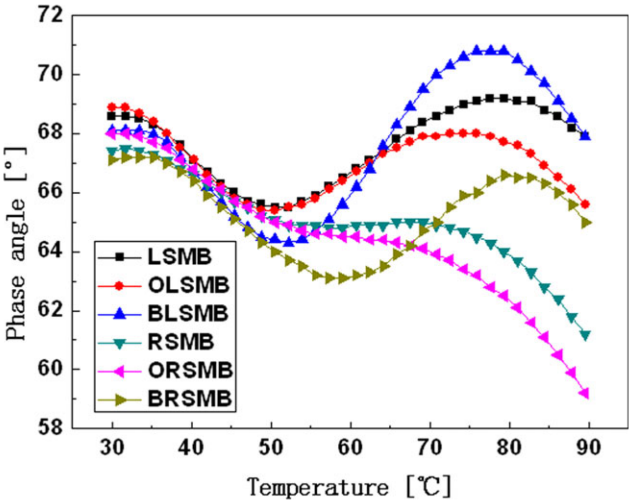

Figure 1. δ versus temperature of different UV absorber/SBS-modified bitumens before aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

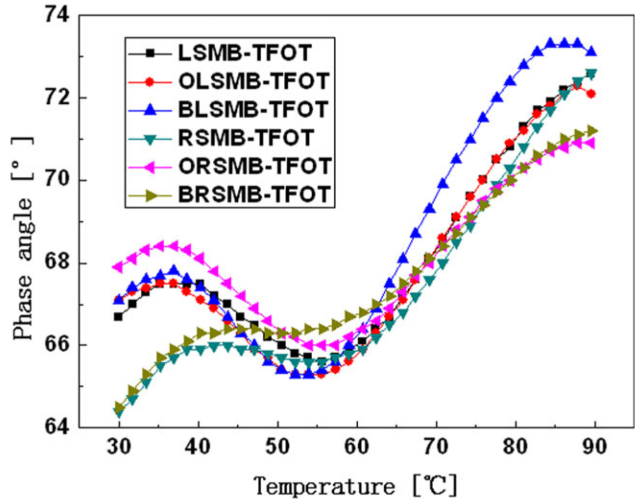

Figure 2. δ versus temperature of different UV absorber/SBS-modified bitumens after TFOT aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

indicated an increased elasticity of LSMB at higher temperatures with the introduction of octabenzone. For BLSMB, the δ curve went across LSMB curve at about 65°C. Compared with LSMB, a decrease in δ with an obviously reduced depth of the δ minimum was observed below the crossover point (65°C), but an increase in δ was seen when the temperature exceeded 65°C. Consequently, the elastic behavior of LSMB was enhanced by the bumetrizole at intermediate temperatures, but at high temperatures, the viscous behavior was increased.

As shown in Figure 1, a smooth decrease in δ appeared in the curves of RSMB and ORSMB; this indicated a more elastic behavior with increasing temperature. Similar to LSMB, the elasticity of RSMB was also enhanced by the octabenzone at high temperatures, as shown by the smaller δ values. The δ curve of BRSMB was much similar than that of BLSMB; this showed that the addition of bumetrizole increased the elastic response at intermediate temperatures on the one hand and the viscous response at high temperatures on the other hand for both LSMB and RSMB.

The temperature dependence of δ of different UV absorber/SBS-modified bitumens after TFOT aging is shown in Figure 2. The viscoelastic properties of the binders were obviously changed after TFOT aging, as indicated by the distinct shape of the δ curves. The TFOT aging made OLSMB and BLSMB exhibit a similar trend to that of LSMB. However, the depths of the δ minimum of OLSMB and BLSMB at about 54°C were larger than that of LSMB. Therefore, the elastic behaviors of the UV absorber/LSMBs were more pronounced than those of the control samples after TFOT aging because the δ minimum was often attributed to the polymer elastic network or entanglement in the binder.[2,3,18]

As shown in Figure 2, a smooth increase in δ without the δ minimum took place in the curves of RSMB and BRSMB after TFOT aging. This was interpreted to mean that the polymer elastic network of both RSMB and BRSMB disappeared during TFOT aging, and thus, the binders showed a viscoelastic behavior similar to the base bitumen. With respect to the TFOT-aged ORSMB, it could be considered that the degradation of the polymer elastic network was inhibited by octabenzone because the δ minimum still existed in the δ curve.

The influence of UV aging on the temperature dependence of δ of different UV absorber/SBS-modified bitumens is illustrated in Figure 3. Although the δ curves of the UV-aged LSMB with or without UV absorbers were very like those of the TFOT-aged ones, the depth of the δ minimum decreased for all binders during UV aging; this showed that the UV aging degraded the SBS polymers further. The δ minimum of BLSMB was nearly the same as that of LSMB after UV aging, whereas that of OLSMB remained smaller than that of LSMB. The effect of UV aging on δ of RSMB with or without UV absorbers was almost

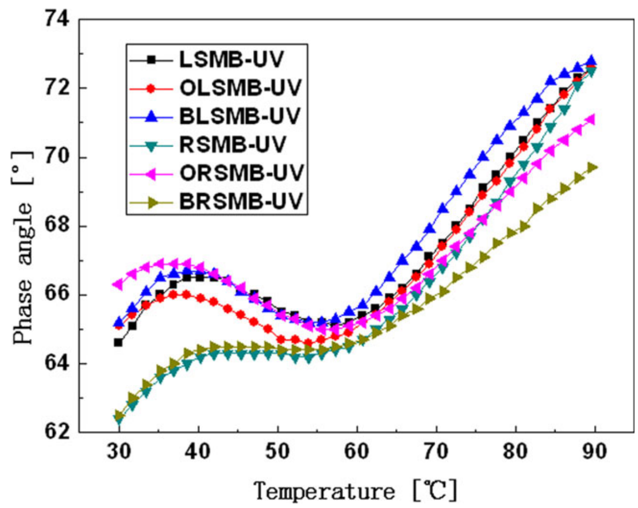

Figure 3. δ versus temperature of different UV absorber/SBS-modified bitumens after UV aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

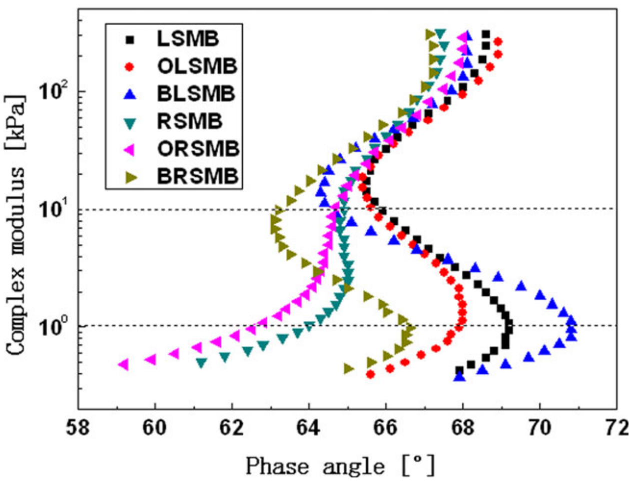

Figure 4. Black diagrams of different UV absorber/SBS-modified bitumens before aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

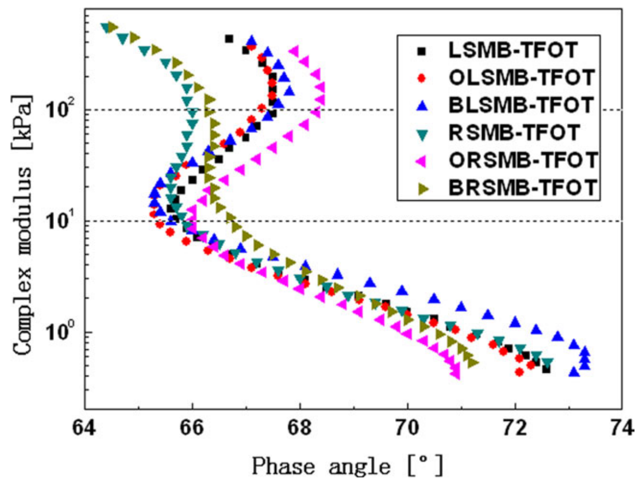

Figure 5. Black diagrams of different UV absorber/SBS-modified bitumens after TFOT aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the same as that of TFOT aging. The different rheological and aging effects of octabenzone and bumetrizole on the studied SBS-modified bitumens were likely related to the chemical structures of the UV absorbers (seen in Ref. 16).

Black Diagrams. The effects of the UV absorbers on the rheological parameters (complex modulus and δ) of the SBS-modified bitumens were combined in the form of black diagrams (Figure 4), which are considered to be rheological fingerprints.[19,20] As shown in Figure 4, the black diagrams could be divided into three parts by complex moduli of about 10 and 1 kPa. δ decreased with the reduction of the complex modulus above a complex modulus value of 10 kPa (part I), but the δ went toward a higher value with the reduction of the complex modulus between 10 and 1 kPa (part II). The same phenomenon as in part I was observed when the complex modulus was lower than 1 kPa (part III). However, RSMB and ORSMB showed a continuous shift toward elastic response with the reduction of the complex modulus, as indicated by the decreasing δ values within the whole range of the complex modulus in the curves of RSMB and ORSMB.

Moreover, the addition of octabenzone led to a smaller δ for both LSMB and RSMB only when the complex modulus was lower than 10 kPa; this indicated an enhanced elastic response of the control binders with the introduction of octabenzone. However, the bumetrizole affected the black diagrams of LSMB and RSMB in a little more complex manner: on the one hand, the elastic response of the binders increased at intermediate complex modulus values, and on the other hand, the viscous response was increased at lower complex modulus values by bumetrizole.

The black diagrams of different UV absorber/SBS-modified bitumens after aging are displayed in Figures 5 and 6. Clearly, the black diagrams were obviously changed after the TFOT aging (Figure 5) and UV aging (Figure 6) for all of the binders. There were also three parts in the black diagrams, but the dividing points of the complex modulus changed at 100 and 10 kPa. At complex modulus values lower than 10 kPa, δ continuously increased with decreasing complex modulus; this indicated a transformation from more elastic behavior to more viscous behavior after aging. These changes could be attributed to the breakdown of the SBS polymer network during the aging processes, and consequently, the effectiveness of the SBS polymer in modifying the bitumen rheology was reduced, and a higher viscous behavior was shown.[2,3,21]

In Figures 5 and 6, the black diagram patterns of LSMB were not obviously changed by the octabenzone and bumetrizole after TFOT and UV aging; this evidenced a lesser influence of the UV absorbers on the viscoelastic properties of the aged binders. However, a significant difference was observed when the octabenzone was added to RSMB; the black diagram patterns of RSMB were nearly unchanged with the addition of bumetrizole during the TFOT and UV aging.

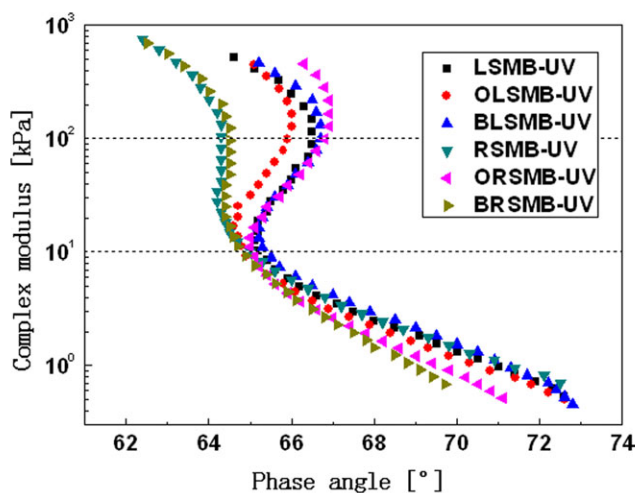

Figure 6. Black diagrams of different UV absorber/SBS-modified bitumens after UV aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

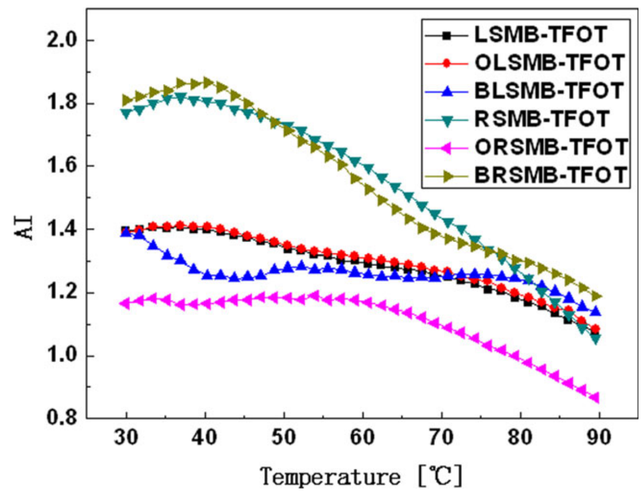

Figure 7. AIs of different UV absorber/SBS-modified bitumens after TFOT aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

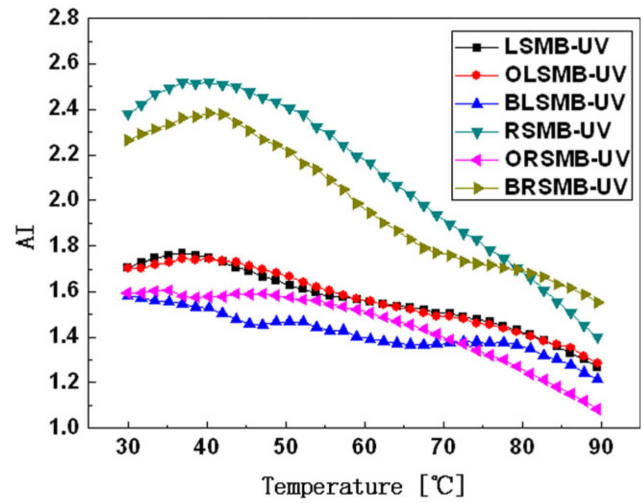

Figure 8. AIs of different UV absorber/SBS-modified bitumens after UV aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Aging Properties

The aging degrees of the binders could be judged on the basis of changes in the rheological properties before and after aging. In this study, the aging index (AI), which was calculated according to eq. (1), was used to evaluate the aging properties of the different UV absorber/SBS-modified bitumens:

$$AI = G^*_{\text{Aged}}/G^*_{\text{Unaged}} \quad (1)$$

where G^*_{Unaged} is the complex modulus before aging and G^*_{Aged} is the complex modulus after aging (kPa). Although it is believed that AI is not an accurate parameter for characterizing the aging properties of polymer-modified bitumens,[2] AI based on the complex modulus (G^*) ratio is still applicable in the absence of anything better because the complex modulus can reflect a combined effect of polymer degradation and bitumen oxidation. Generally, high values of AI indicate a high degree of bitumen aging.

TFOT Aging. The AI values of different UV absorber/SBS-modified bitumens after TFOT aging are displayed in Figure 7. It was clear that the thermooxidative-aging properties of LSMB were influenced little by octabenzone and bumetrizole, as revealed by the similar AI values of the three binders after TFOT aging. As to RSMB, octabenzone improved its resistance to the thermooxidative aging remarkably because the AI values of ORSMB were much smaller than those of RSMB. However, BRSMB showed similar AI values to RSMB; this indicated that the bumetrizole slightly affected the thermooxidative aging of RSMB. Also, LSMB exhibited better thermooxidative-aging resistance than RSMB, as indicated by the smaller AI values of LSMB than RSMB.

UV Aging. Figure 8 displays the AI values of different UV absorber/SBS-modified bitumens after UV aging. The AI curve of OLSMB overlapped that of LSMB after the UV aging, but that of BLSMB was lower than that of LSMB. This implied that the UV-aging resistance of LSMB was influenced little by octabenzone but was much improved by bumetrizole. As shown in Figure 8, the ability of RSMB to resist UV aging could be improved by both octabenzone and bumetrizole in that the AI values of RSMB were reduced by the octabenzone and bumetrizole after UV aging within a temperature domain of 30–80°C. Moreover, LSMB also had better UV-aging resistance than RSMB.

To evaluate the effects of the pure UV irradiation on the aging extent of different UV absorber/SBS-modified bitumens, the AI values between TFOT and UV aging were also calculated on basis of eq. (1). The results are shown in Figure 9. Evidently, the pure UV aging of LSMB and RSMB could be prevented by the UV absorbers (except for OLSMB). Although the AI curve of OLSMB was crossed with that of LSMB, the AI values of OLSMB were lower than that of LSMB at most temperatures from 30 to 90°C.

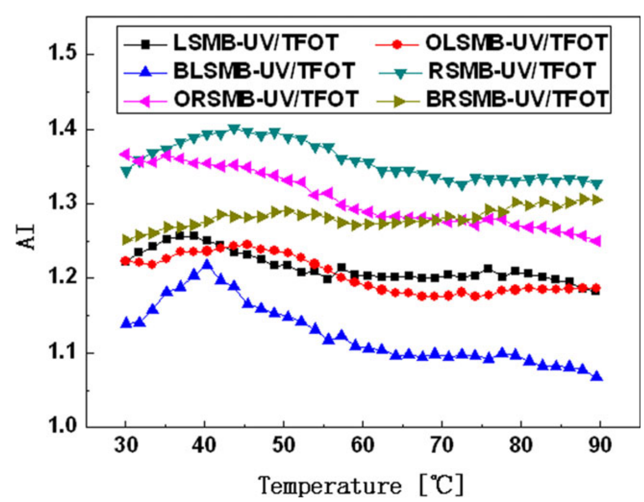

Figure 9. AIs of different UV absorber/SBS-modified bitumens between TFOT and UV aging. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

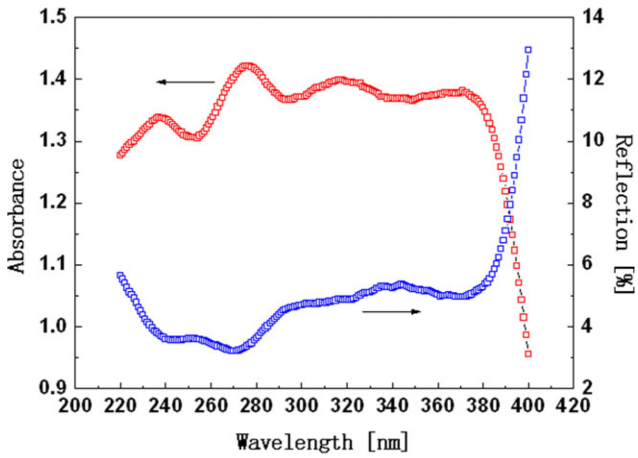

Figure 10. UV–vis spectra of the octabenzone. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The reason the UV absorbers could improve the UV-aging resistance of the SBS-modified bitumens may have been related to the mechanisms of the UV absorber itself. As displayed in Figures 10 and 11, the UV–vis spectra of octabenzone and bumetrizole manifested in that both octabenzone and bumetrizole had high absorbances to UV rays. With respect to the UV irradiation at 365 nm at which the UV aging was conducted in this study, the absorbances of octabenzone and bumetrizole were 1.379 and 0.995, respectively. When the binders were exposed to the UV irradiation, the UV absorbers could absorb the photon energy from the UV rays and then convert them into heat energy. The heat energy generated could dissipate into the surrounding environment naturally and harmlessly.[22] As a result, the binders were protected to some extent with the introduction of the UV absorbers. Octabenzone and bumetrizole had different anti-aging effects on the binders because the UV absorbers may have shown a specific choice of the SBS-modified binders; this was also revealed for the base bitumen in our previous research.

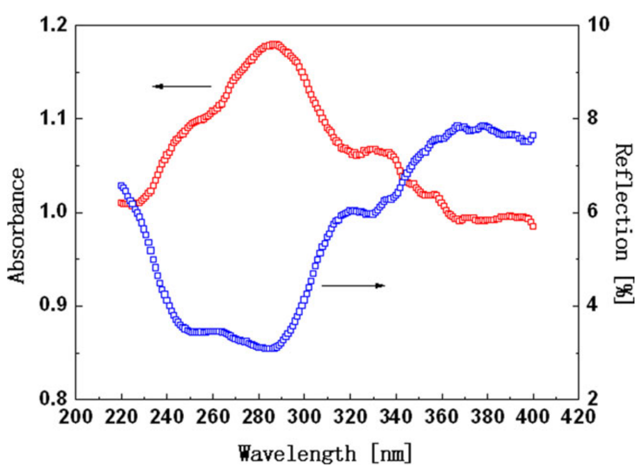

Figure 11. UV–vis spectra of the bumetrizole. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Physical Properties of the UV Absorber/SBS-Modified Bitumens before Aging

Binder	Penetration at 25°C (0.1 mm)	Ductility at 5°C (cm)	Softening point (°C)	Viscosity at 135°C (Pa s)
LSMB	62	32.5	67.8	1.52
OLSMB	69	34.1	79.0	1.38
BLSMB	66	39.8	75.5	1.41
RSMB	64	24.0	86.7	1.55
ORSMB	71	32.1	89.0	1.54
BRSMB	64	28.3	91.4	1.53

Physical Properties

The physical properties of the UV absorber/SBS-modified bitumens before aging were measured to investigate the property modification of the SBS-modified bitumens by the UV absorbers. The results of the physical properties are shown in Table I. Octabenzone and bumetrizole showed similar modifications of the physical properties for LSMB and RSMB. The penetration, ductility, and softening point of the two types of SBS-modified bitumen increased, whereas the viscosity decreased with the introduction of the two UV absorbers. As a result, octabenzone and bumetrizole could bring beneficial effects to the physical properties of the SBS-modified bitumens. It is known that these physical parameters are the basic indices for evaluating whether a binder is qualified for application on a specific road. From Table I, it could be concluded that the studied UV absorber/SBS-modified bitumens still met the requirements for road construction and even surpassed the control samples at some of these properties.

Storage Stability

The storage stability is one of the most critical parameters for SBS-modified bitumens because segregation may occur when polymer-modified bitumens are blended and stored for weeks, and this determines the normal application of a binder in road construction and usage.[23,24] The results of the storage stability test are listed in Table II. The softening point difference of all the binders was less than 2.5°C. After modification with the two UV absorbers (octabenzone and bumetrizole), the storage stability of the SBS-modified bitumens was improved, especially that

Table II. Differences in the Softening Points of the UV Absorber–SBS-Modified Bitumens

Binder	Softening point (°C)		
	Top	Bottom	Difference
LSMB	77.8	76.8	1.0
OLSMB	74.7	74.2	0.5
BLSMB	72.3	72.2	0.1
RSMB	88.6	87.3	1.3
ORSMB	89.3	88.2	1.1
BRSMB	90.2	89.8	0.4

of the binders mixed with bumetrizole. Thereby, the UV absorber/SBS-modified bitumens could be regarded as having good storage stability and were suitable use in bituminous pavement.

CONCLUSIONS

The rheological properties of the SBS-modified bitumens were modified by the introduction of UV absorbers to different extents. For LSMB and RSMB, the octabenzone enhanced their elasticity only at higher temperatures (or lower complex moduli), whereas the addition of bumetrizole increased the elastic response at intermediate temperatures (or moderate complex modulus values) on the one hand and the viscous response at high temperatures (or low complex modulus values) on the other hand.

After aging by TFOT and UV, a transformation from a more elastic response to a more viscous response took place at high temperatures (or low complex modulus values) for all of the binders; this was attributed to the breakdown of the SBS polymer network during the aging processes. Regardless of the aging procedures, LSMB exhibited a better ability to resist aging than RSMB.

The UV-aging properties of RSMB could be effectively alleviated by both octabenzone and bumetrizole because of their high absorbance and protective mechanisms to UV rays, whereas that of the linear ones could only be improved by bumetrizole. The influence of the UV absorbers on the thermooxidative-aging properties of the SBS-modified bitumens was dependent on the types of UV absorbers and SBS polymers; octabenzone improved the thermooxidative-aging resistance of RSMB remarkably but had little influence on LSMB. Bumetrizole showed no improvement for the thermooxidative-aging resistance for either LSMB or RSMB.

The UV absorber/SBS-modified bitumens showed enhanced physical properties and storage stability and well satisfied the requirements of road construction. Consequently, the binders possess great potential for providing bituminous pavements with excellent mechanical properties and aging resistance.

ACKNOWLEDGMENTS

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