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Chemo-Rheological Characterization of Aging Behaviors of Warm-Mix High-Viscosity Modified Asphalt

Bei Chen[1]; Fuqiang Dong[2]; Xin Yu[3]; Shisong Ren[4]; and Changjiang Zheng[5]

Abstract: Today, high-viscosity modified asphalt (HVMA) is widely used in drainage asphalt pavements. However, due to its high construction temperature, HVMA is prone to thermal oxidative aging, which reduces its life cycle and increases the maintenance costs of asphalt pavements. In this study, the effects of warm-mix additives on the physical, rheological and chemical properties of HVMA were studied to determine optimum warm-mix conditions. First, the effects of warm-mix technologies (foam warm mix, Sasobit, Evotherm, and GLWBR at 3%, 3%, 0.8%, and 0.8%, respectively) on the physical and rheological properties of HVMA were studied. Then, two aging methods [thin film oven test (TFOT) and pressure aging vessel (PAV)] were applied to simulate the short-term and long-term thermal oxidation processes of HVMA and evaluate the influences of different warm-mix technologies on HVMA aging. The results showed that the four warm-mix technologies, especially foam warm mix and Sasobit, reduced the construction temperature of HVMA. In addition, warm-mix technologies also improved the high-temperature rheological properties of HVMA; however, they had adverse effects on the low-temperature cracking resistance of asphalt. Based on asphalt aging index fluctuations, it was evident that warm-mix technology was not conducive to the antiaging performance of HVMA, and foam warm mix had the weakest influence on the antiaging performance of high-viscosity asphalts. Furthermore, according to the results of an analysis of the carbonyl changes in aging asphalts, asphalt aging index can be applied to predicting the degree of aging of warm-mix HVMA. **DOI: 10.1061/(ASCE)MT.1943-5533.0004501.** *© 2022 American Society of Civil Engineers.*

Author keywords: Warm-mix additive; High-viscosity modified asphalt (HVMA); Rheological characteristics; Aging index; Aging degree.

Introduction

Porous asphalt pavements are widely used in urban road construction due to their good permeability, low noise, and excellent skid resistance (Barrett and Shaw 2007). Porous asphalt pavements are mainly composed of high-viscosity modified asphalt (HVMA) and coarse aggregate; HVMA is of critical importance in the drainage performance of asphalt pavements (Hu et al. 2014). However, due to its high viscosity, HVMA requires high construction temperatures in the production, mixing, and compaction stages, which results in high flue gas emissions and energy consumption and, therefore, environmental pollution and energy waste (Tian et al. 2020). In addition, the high construction temperatures of HVMA cause performance degradation and aging. Therefore, warm-mix technology has been applied to HVMA to decrease construction temperature, decelerate attenuation and aging, save energy, and reduce gas emissions (Chen and Dong 2021; Takahashi 2013).

The application of warm-mix technology is considered to be an effective solution for decreasing the viscosity of asphalt mixtures during construction. Warm-mix technology also reduces mixing and compacting temperatures, which enhances energy savings and reduces environmentally hazardous emissions compared with hot-mix asphalts (Behl and Chandra 2017; Chen et al. 2021). Currently, three types of warm-mix technology are available for asphalt, namely foam warm mix, wax warm-mix additives, and surface active warm-mix additives. Some researchers have studied warm-mix technology (Podolsky et al. 2017). Mansoori and Modarres (2020) analyzed the rheology and microstructure of asphalt binders with Sasobit and two chemical warm-mix additives (Zycothm and PAWMA). According to the research results, the three aforementioned warm-mix additives decreased the construction temperature of asphalt mixtures, but the two warm-mix additives had slight effects on the rheological properties and microstructure of asphalt binder. Zheng et al. (2019) studied the effects of warm-mix additives on the physical, rheological, and morphological properties of HVMA. Their results showed that warm-mix additives obviously reduced viscosity (135°C) and improved dynamic viscosity (60°C). Also, it was found that EC-120 could enhance the high- and low-temperature rheological properties of the system under different aging conditions. Zhang et al. (2018) explored the effects of adding black thermochromic powders to asphalt binder on the aging behavior of asphalt through three aging tests (thin film oven test, pressure aging vessel test, and ultraviolet radiation). Their results showed that the addition of black thermochromic powders, especially at a concentration of 4%, improved the high- and low-temperature performance of asphalt and strengthened its aging resistance.

Overall, several research works have been performed using different types of warm-mix technologies and HVMA (Table 1).

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Table 1. Effects of aging behavior on asphalt as observed by various researchers

References	Research object	Aging method	Related results
Wang et al. (2020)	SBS modified asphalt	TFOT and PAV	High-viscosity modifiers had good compatibility with SBSMA binder. High-viscosity modifiers can effectively improve the antiaging performance of SBSMA.
Zheng et al. (2019)	High-viscosity asphalt	TFOT and PAV	EC-120 improved the low-temperature creep behavior of asphalt binder after long-term aging.
Mansoori and Modarres (2020)	Bitumens and bituminous mastics	TFOT and PAV	Sasobit changed the microstructural properties analyzed by atomic force microscopy.
Hu et al. (2020b)	High-viscosity modified asphalt	TFOT and PAV	After long-term aging, the oxidation of base asphalt is the main reason for the changes in rheological property of high viscosity modified asphalt.
Hu et al. (2020a)	High-viscosity modified asphalt	Weathering aging	The strain recovery of HVMA decreased initially and then increased with the process of weathering aging.

Note: SBS = styrene-butadiene-styrene; and SBSMA = styrene butadiene styrene modified asphalt.

However, previous studies have generally focused on the influences of different types of warm-mix technologies on the performance of modified asphalt, and limited work has been conducted on of the effect of warm-mix technologies on the aging and microstructure of HVMA (Jamshidi et al. 2013). Therefore, it seemed necessary to explore the impacts of different warm-mix technologies on the performance and aging degree of HVMA (Antunes et al. 2015; Pasetto et al. 2017; Cheraghian et al. 2021).

This study aimed to evaluate the effects of different warm-mix technologies on the rheological properties and aging behaviors of HVMA and to introduce a new warm-mix additive and explore its applicability in HVMA. First, the effects of foam warm mix, wax warm-mix additive (SASOBIT), and surface active warm-mix additives (Evotherm and GLWBR) on the physical and rheological properties of HVMA were studied. Second, based on variations of asphalt aging index, this study analyzed the influence of warm-mix technology on the aging degree of HVMA after thin film oven test (TFOT) and pressure aging vessel (PAV) test. Last, correlation between carbon change and asphalt aging index in warm-mix HVMA was analyzed by Fourier transform infrared (FTIR) spectroscopy. Fig. 1 schematically illustrates the experimental techniques applied in this study (Cheraghian and Wistuba 2021).

Experiments

Materials

Asphalt

The 70# base asphalt produced by the SsangYong asphalt plant in Seoul, South Korea was adopted in this work and the physical properties of base asphalt are summarized in Table 2.

High-Viscosity Modifier

The high-viscosity modifier applied in this research was made in our laboratory. The main components of the high-viscosity modifier was styrene-butadiene-styrene (SBS), C9 petroleum resin, wax, and naphthenic white oil. Fig. 2 shows the high-viscosity modifier, and basic performance test results are given in Table 3.

Warm-Mix Additives

Currently, three types of warm-mix technology are available for asphalt, namely, foam warm mix, wax warm-mix additives, and surface active warm-mix additives. In this research, a representative foam warm mix, a wax warm-mix additive (Sasobit), a surface-active warm-mix additive (Evotherm), and a self-developed surface-active warm-mix additive (GLWBR) were adopted to explore the influence of different warm-mix technologies on the aging performance of HVMA (Dong et al. 2018). The warm-mix additives applied in the experiments included Sasobit, Evotherm, and GLWBR. Fig. 3 shows the aforementioned additives, and Fig. 4 presents their FTIR spectra. In addition, specific performance indexes are presented in Table 4. GLWBR is a new type of surfactant warm-mix additive; it was expected to produce warm-mix effects and antiaging properties similar to those of Evotherm.

As shown in Fig. 4, absorption peaks in the range of 2,850 to 2,925 cm^{-1} were assigned to the stretching vibrations of methylene ($-CH_2$) and methyl ($-CH_3$). The absorption peak at 1,740 cm^{-1} was due to the stretching vibration of the carbonyl group ($-C{=}O$), and those at 1,460 and 700 cm^{-1} corresponded to the bending vibrations of CH_2 and $C{=}C$, respectively. From the absorption bands on the spectra, it was evident that the main components of Sasobit were organic compounds containing C-H functional groups, and the main chemical functional groups of Evotherm and GLWBR were almost the same. The specific functional groups in the warm-mix additives directly affected their performance.

Methodology

Preparation of Foam Warm-Mix HVMA

An FM300-Digital shear machine (FLUKO, Shanghai, China) was used for the preparation of HVMA. An 8% high-viscosity modifier was added to the base asphalt and mixed in the shear machine at 175°C and 4,000 rpm for 40 min. Specifically, the HVMA was prepared by stirring for 2 h. Next, an FCF-1 reactor (Qiusuo Technology, Shanghai, China) was employed to fabricate foam warm-mix HVMA. During the experiments, foaming water content was 3% of the quality of HVMA (Dong et al. 2018).

Preparation of Warm-Mix HVMA with Warm-Mix Additives

In this study, HVMA was prepared according to the method mentioned previously. Briefly, three types of warm-mix additives were added to the HVMA and stirred with a stirrer for 1 h at 175°C to prepare different types of warm-mix HVMAs. According to the recommended dosages of warm-mix additives in the literature (Jamshidi et al. 2013), the volume fractions of Sasobit, Evotherm, and GLWBR used were 3%, 0.8%, and 0.8%, respectively (Podolsky et al. 2016; Ferrotti et al. 2017). The four warm-mix HVMAs were named WATER, SASOBIT, EVOTHERM, and GLWBR, respectively. The HVMA without warm-mix treatment was used as a blank sample.

Aging Process

According to the ASTM D1754 (ASTM 2009) and ASTM D6521 (ASTM 2008a) standards, TFOT and PAV were used to simulate the short-term and long-term thermal oxidation aging processes of

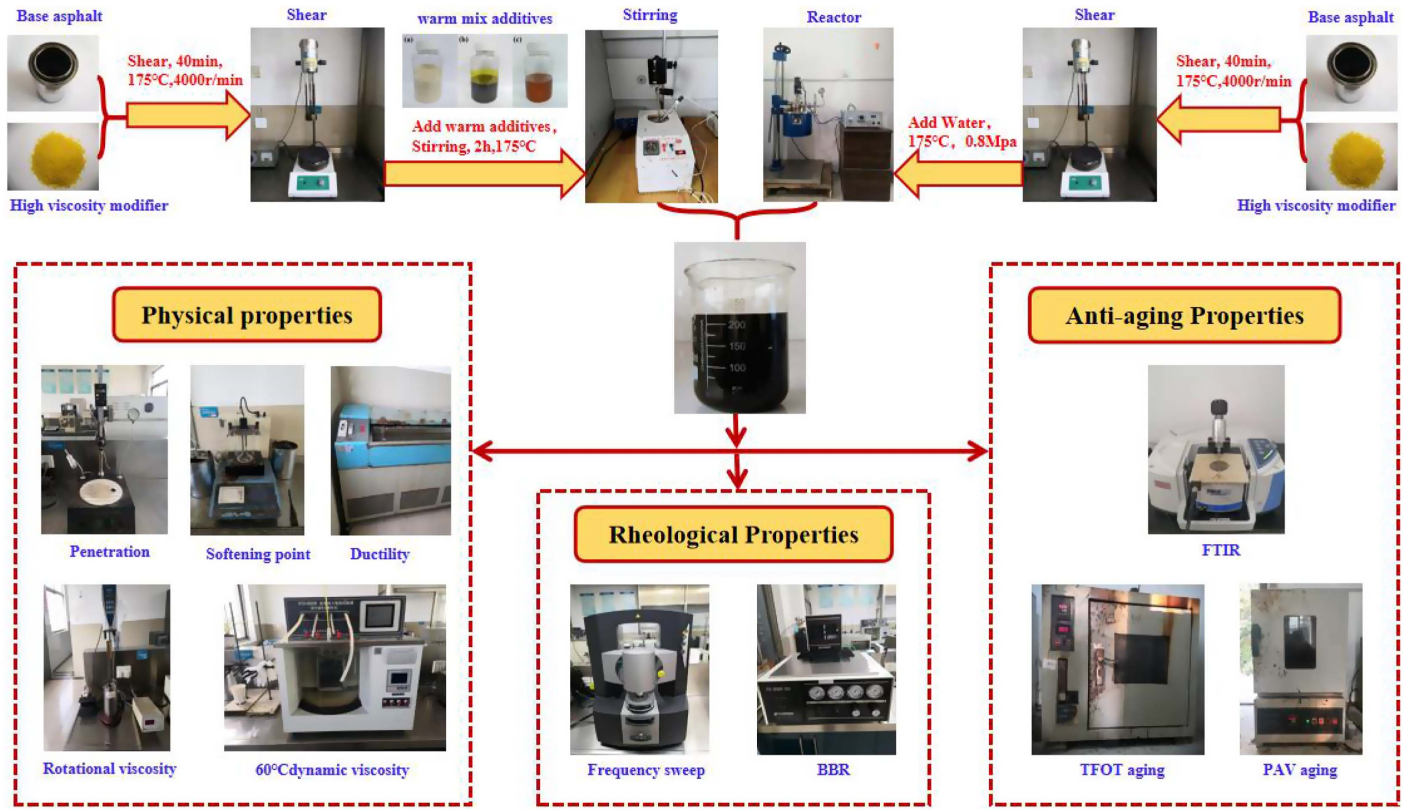

Fig. 1. Schematic overview of experimental techniques applied in this study.

Table 2. Physical properties of base asphalt

Property	Results
Penetration (0.1 mm)	68
Softening point (°C)	47.8
Ductility (15°C) (cm)	>100
Dynamic viscosity at 60°C (Pa · s)	167.5
Density (15°C) (g · cm^{-3})	1.047
Flash point (°C)	345

HVMA, respectively. Furthermore, the aging sensitivity of the warm-mix HVMAs was characterized by the variation of the physical and rheological properties of the materials before and after aging (Zhao et al. 2012); these properties are given in detail in Table 5 (Wang et al. 2020).

Characterization Methods

Physical Properties Tests

The penetration (25°C), softening point, and ductility (5°C) of the warm-mix HVMAs were tested according to the ASTM D36 (ASTM 2012a), ASTM D5 (ASTM 2013), ASTM D113 (ASTM 2007) standards, respectively. The temperature sensitivity of the asphalts was characterized based on penetration index (PI) such that higher values of PI indicated decreased sensitivity. Also, the asphalts had a gel structure at PI values of greater than or equal to 2 and a sol-gel structure at PI values of less than 2. PI was calculated according to Eq. (1):

$$\mathrm{PI} = \frac{1,952 - 500\,\log(P25) - 20Sp}{50\,\log(P25) - Sp - 120} \tag{1}$$

where Sp = softening point; and $P25$ = penetration at 25°C.

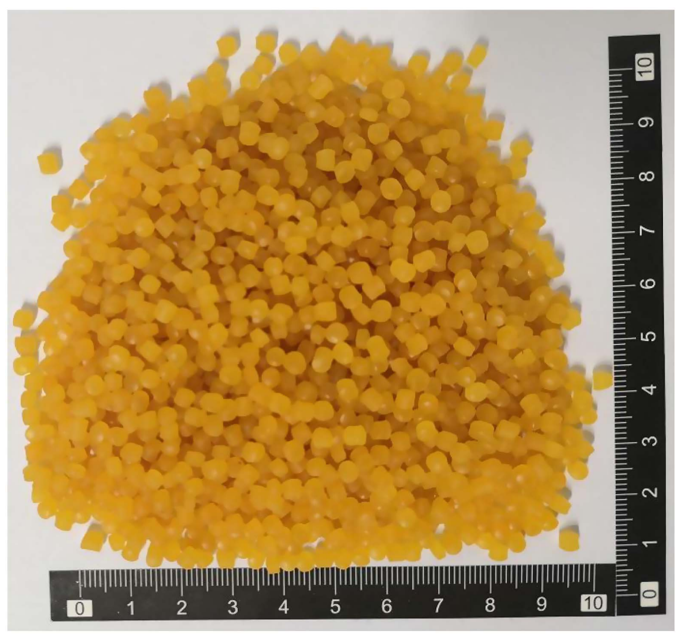

Fig. 2. Prepared high-viscosity modifier.

Rotational Viscosity and Absolute Viscosity Tests at 60°C

According to the ASTM D4402 (ASTM 2012b) standards, the viscosity of the warm-mix HVMAs was measured by rotating viscosity equipment in the temperature range of 135°C–175°C. An SYD-0620B asphalt absolute viscosity tester (Changji, Shanghai, China) was employed to analyze the 60°C absolute viscosity of the warm-mix HVMAs (Zhang et al. 2018).

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Table 3. Basic properties of high-viscosity modifier

Items	Results	Standard results	Test methods
Exterior	Yellow particles	Granular	Visual inspection
Melt index (g/10 min)	8	≥2.0	Melt
Individual particle quality (g)	0.12	≤0.5	Weighing
Dry mix dispersibility	No residue	No residue	Dry mix

Rheological Properties Tests

According to ASTM D7175 (AASHTO 1995), the rheological properties of HVMAs were studied using a TA-AR1500 dynamic shear rheometer (DSR) at experimental temperatures of 46°C (Netzsch, Selb, Germany), 52°C, 58°C, 64°C, 70°C, 76°C, and 82°C. The measured high-temperature rheological properties of the HVMAs included complex modulus G^*, phase angle δ, and rutting factor $G^*/\sin\delta$.

In order to explore the influence of different warm-mix technologies on the low-temperature performance of the HVMAs, bending beam rheometer (BBR) equipment fabricated by Cannon Instrument Company (Cangzhou, China) was applied at test temperatures of −6°C, −12°C, −18°C, and −24°C. The creep stiffness (S) and creep rate (m-value) of the warm-mix HVMAs were measured according to ASTM D6648 (ASTM 2008b) and AASHTO T 313 (AASHTO 2019; Jiang et al. 2017). Also, the performance grade (PG) of the warm-mix HVMAs was determined according to the high- and low-temperature rheological properties of the warm-mix HVMAs.

FTIR Tests

It is well known that the aging of asphalt binder in high-temperature environments is mainly caused by thermal oxidation. The number of carbonyl functional groups (C═O) in asphalt binders changes during thermal oxidation (Mouillet et al. 2007). Therefore, asphalt aging degree can be characterized by evaluating the carbonyl changes of warm-mix HVMA before and after aging. During the experiments, carbonyl index (CI) was characterized by carbonyl number, which is defined by Eq. (2) (Zhao et al. 2012):

$$\mathrm{CI} = \frac{A_{C=O}}{\sum A} \quad (2)$$

where $A_{C=O}$ = absorption peak area of the carbonyl functional groups (C═O) at 1,700 cm^{-1}; $\sum A$ = sum of absorption peak areas, that is $\sum A = \mathrm{A}^{-1}_{1700\,\mathrm{cm}} + \mathrm{A}^{-1}_{1600\,\mathrm{cm}} + \mathrm{A}^{-1}_{1460\,\mathrm{cm}} + \mathrm{A}^{-1}_{1376\,\mathrm{cm}} + \mathrm{A}^{-1}_{1030\,\mathrm{cm}} + \mathrm{A}^{-1}_{864\,\mathrm{cm}} + \mathrm{A}^{-1}_{814\,\mathrm{cm}} + \mathrm{A}^{-1}_{743\,\mathrm{cm}} + \mathrm{A}^{-1}_{724\,\mathrm{cm}}$. Higher differences in CI (ΔCI) before and after sample aging indicated a higher aging degree of the warm-mix HVMAs.

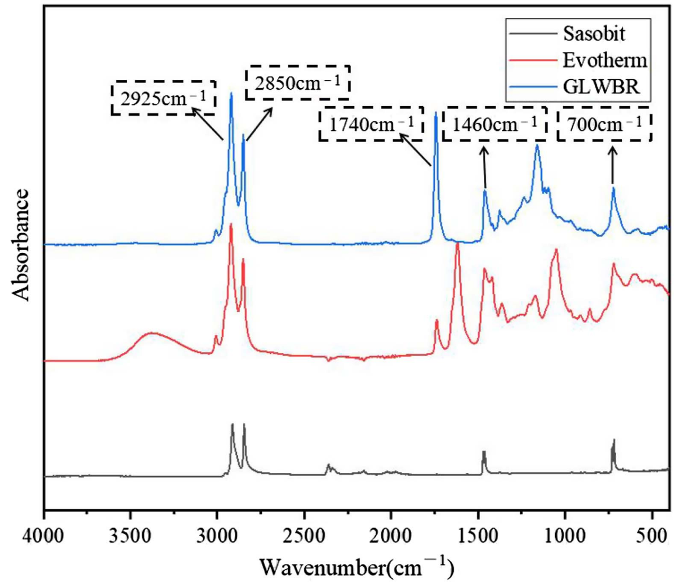

Fig. 4. FTIR spectra of the studied warm-mix additives.

Warm-mix HVMA was placed on an FTIR spectroscopy bench and compacted and fixed with a metal layer. Asphalt samples were scanned 32 times in the wave number range of 400–4,000 cm^{-1}. Spectral analyses were carried out using a computer program developed by MATLAB version R2017a software.

Results and Discussion

Analysis of Physical Properties

Analysis of Basic Properties

The effects of the different warm-mix additives on the physical properties of HVMA are presented in Fig. 5. Except for the foam warm mix (WATER), the warm-mix additives decreased the penetration of HVMA [Fig. 5(a)]. In addition, the different warm-mix additives exhibited different degrees of reduction, which was

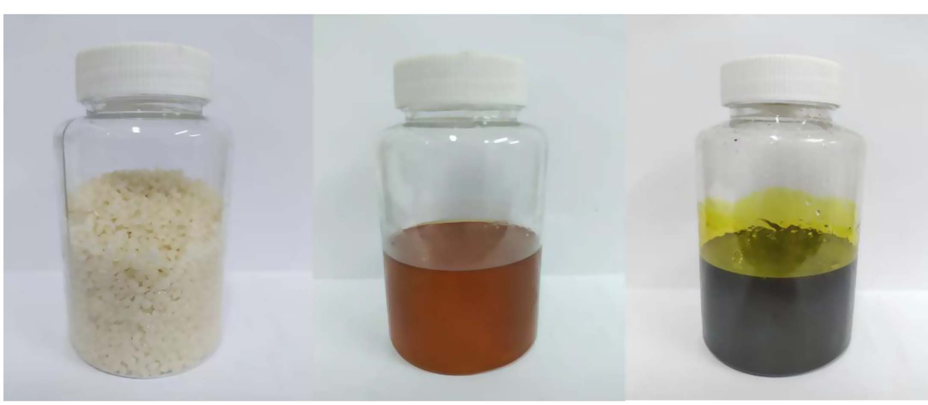

Fig. 3. The studied warm-mix additives: (a) Sasobit; (b) Evotherm; and (c) GLWBR.

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Table 4. Technical properties of the studied warm-mix additives

Property	Sasobit	Evotherm	GLWBR
Physical state	Solid	Liquid	Liquid
Odor	Odor-free	Odor-free	Odor-free
Color	Grayish-white	Pale yellow	Dark brown
Specific gravity	0.65	1.02	1.08
Viscosity (25°C) (MPa · s)	—	450–850	600–1,200
Amine value mg GKOH (g)	—	515–605	455–605
Surface tension (0.6 g/L, 25°C)	—	34.0–37.0	28.0–24.0

consistent with the results obtained by other researchers (Qin et al. 2014). As seen in Fig. 5(b), all four warm-mix technologies improved the softening point of HVMA. This improvement was more significant with the Sasobit warm-mix additive and might be due to the wax crystals in Sasobit. This result was consistent with the findings of previous studies (Sanchez-Alonso et al. 2011; Yu et al. 2016). Fig. 5(c) shows that Sasobit had a great influence on the low-temperature performance of HVMA, while the other three warm-mix technologies had only a small influence. According to Fig. 5(d), compared with the blank sample, the PI value

Table 5. Calculation methods of aging indices

Aging indices	Calculation methods
Softening point increment (SPI)	$\text{SPI} = \lvert \text{softening point}_{\text{aged}} - \text{softening point}_{\text{unaged}} \rvert$
Viscosity aging index (VAI)	$\text{VAI} = \lvert (\text{Viscosity}_{\text{aged}} - \text{Viscosity}_{\text{unaged}}) \rvert \times 100/\text{Viscosity}_{\text{unaged}}$
Complex modulus aging index (CAI)	$\text{CAI} = G^*_{\text{aged}}/G^*_{\text{unaged}}$
Phase angle aging index (PAI)	$\text{PAI} = \delta_{\text{aged}}/\delta_{\text{unaged}}$
Rutting factor aging index (RAI)	$\text{RAI} = \lvert (G^*/\sin\delta_{\text{aged}} - G^*/\sin\delta_{\text{unaged}}) \rvert \times 100/G^*/\sin\delta_{\text{unaged}}$

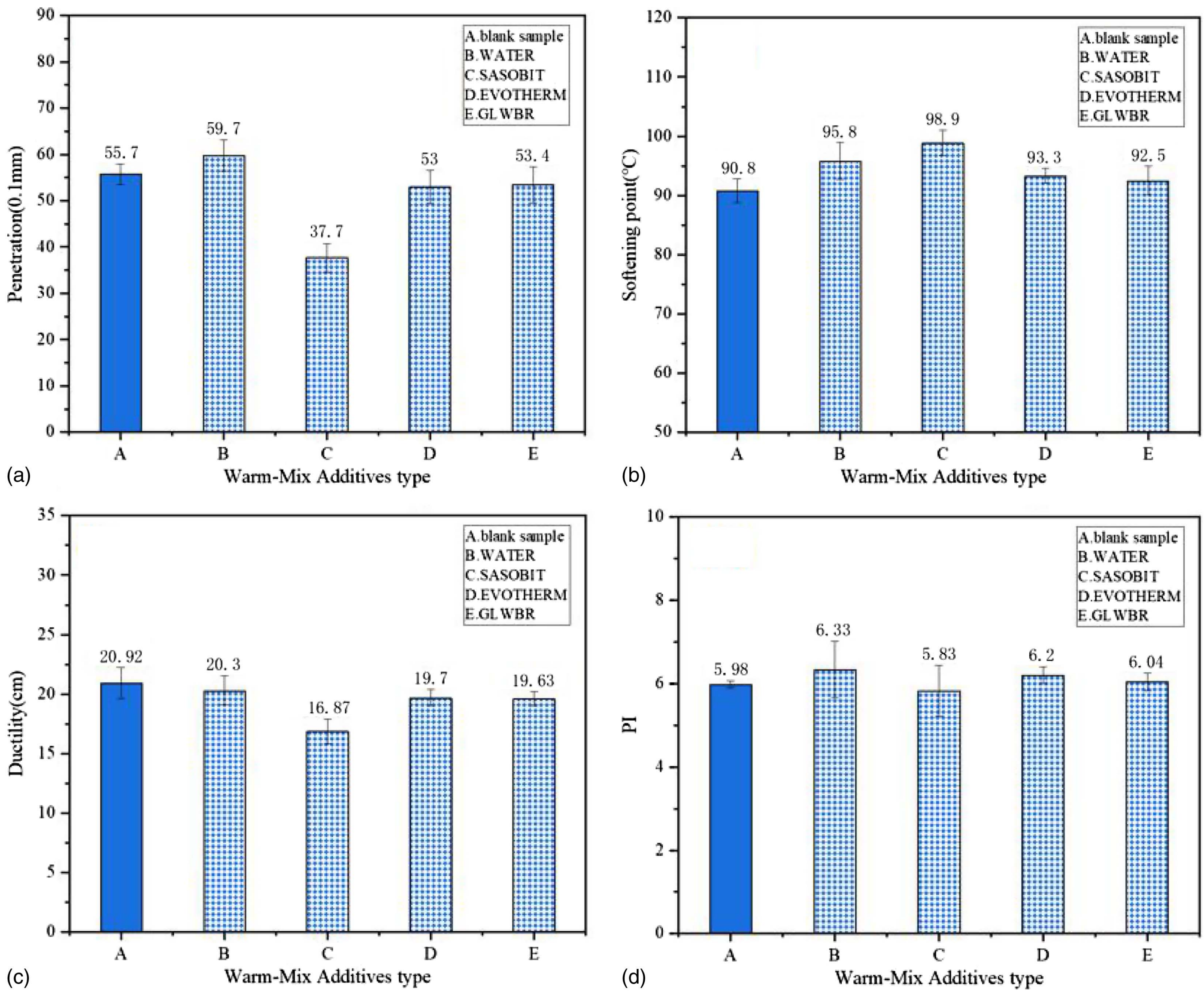

Fig. 5. Effects of different warm-mix additives on the physical properties of warm-mix HVMA: (a) penetration; (b) softening point; (c) ductility; and (d) PI.

dramatically increased for the foam warm mix, obviously decreased for SASOBIT, and did not change significantly for EVOTHERM and GLWBR. Also, the dosage of warm-mix additive affected the PI value of the warm-mix HVMAs. The increase of PI value for foam warm mix indicated an increase in the temperature sensitivity of the foam warm-mix HVMA due to the existence of residual water. The decrease of PI value for SASOBIT indicated that Sasobit reduced the temperature sensitivity of the HVMA, which could be due to the fact that in SASOBIT, wax absorbed a fraction of the heat. However, the PI values of EVOTHERM and GLWBR did not change significantly, which is the mechanism of action of surfactant warm mix additive.

Analysis of Rotational Viscosity and Dynamic Viscosity at 60°C

Based on Fig. 6, all four tested warm-mix technologies reduced the viscosity of HVMA and attained warm-mix effects. The foam warm mix and SASOBIT had the most outstanding effects. EVOTHERM and GLWBR did not have obvious warm-mix effects, because they used surfactant-based warm-mix additives. Surfactant-based warm-mix additives improve the contact between asphalt and aggregate, but their warm-mix effects on asphalt binder are not obvious (Cao and Ji 2011).

The dynamic viscosity test results of warm-mix HVMA at 60°C are shown in Fig. 7. Compared with the blank sample, the dynamic viscosity of the foam warm-mix HVMA was significantly decreased. The reason for this was that the water added to the foam warm-mix technology played a lubricating role among the asphalt molecules. This result was consistent with the findings of Dong et al. (2017). The addition of Sasobit, Evotherm, and GLWBR increased the dynamic viscosity of the warm-mix HVMAs. The addition of Sasobit increased the dynamic viscosity of HVMA, showing an increasing trend at first followed by a decreasing one, because Sasobit is a wax warm-mix additive with melting point of 90°C to 110°C (Silva et al. 2010). At a test temperature of 60°C, Sasobit acted as a physical stabilizer in HVMA. Kim et al. and Zheng et al. also found similar results (Kim et al. 2011; Zheng et al. 2019). The addition of EVOTHERM and GLWBR increased the dynamic viscosity of HVMAs, but the effect of increasing the dynamic viscosity was not obviously. This was because Evotherm and GLWBR are surface-active warm-mix additives. Under high-temperature conditions, a large number of surfactant micelles in warm-mix additive contacted with hot asphalt and water molecules outside the micelles evaporated. Therefore, lipophilic groups contacted the asphalt, improving the dynamic viscosity of the HVMAs.

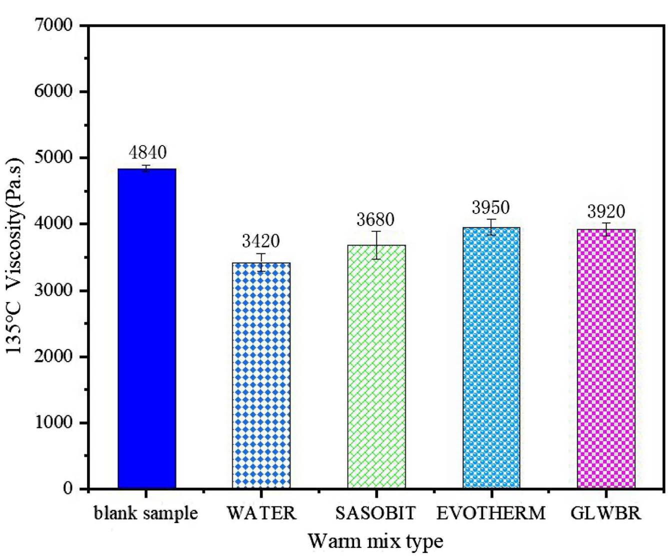

Fig. 6. Effect of warm-mix HVMA on viscosity.

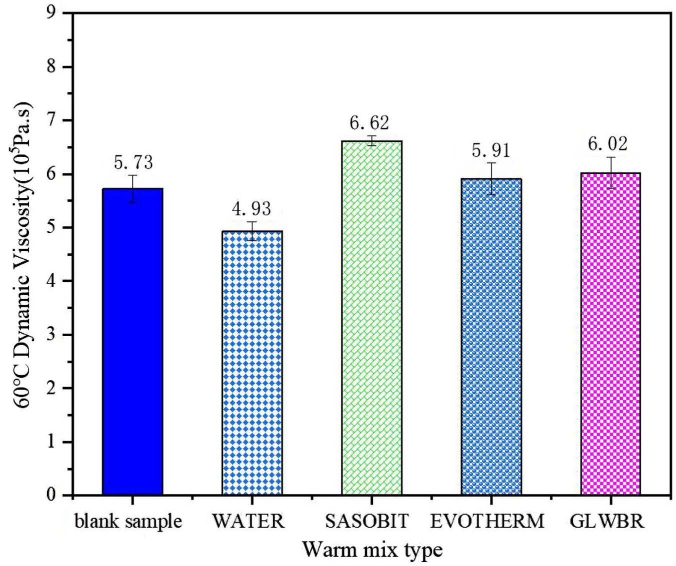

Fig. 7. Effect of warm-mix HVMA on dynamic viscosity (60°C).

Analysis of Rheological Properties

Complex Modulus and Phase Angle

The effects of the four warm-mix technologies on G^* and δ of HVMA for the temperature range of 42°C to 82°C are shown in Fig. 8. Compared with the blank sample, with the exception of the Evotherm warm-mix additive, the warm-mix additives increased the G^* value of the HVMAs; the effect was more significant for Sasobit. Fig. 8(b) shows that the four warm-mix technologies increased the δ value of HVMA, indicating that the warm-mix additives mainly affected the viscosity of the HVMAs and had little effect on their elasticity. It is worth noting that the newly developed GLWBR warm-mix additive increased the δ value of HVMA. Compared with Evotherm, GLWBR improved the high-temperature performance of HVMA. In comprehensively analyzing changes in G^* and δ values, the four warm-mix technologies were found to enhance the viscoelasticity and especially the viscosity of HVMA. The performance of the Sasobit warm-mix additive was the most prominent, because it was a wax additive. The wax crystals in Sasobit formed a lattice structure at temperatures below the melting point and were dispersed in the HVMA, thereby improving the elastic performance of the HVMA (Zheng et al. 2019).

Rutting Factor $G^*/\sin\delta$

The effects of the four warm-mix techniques on the $G^*/\sin\delta$ values of HVMA are shown in Fig. 9. The $G^*/\sin\delta$ values were found to be closely related to the rutting resistance of the asphalt mixtures. Greater $G^*/\sin\delta$ values indicated that the asphalt mixtures had better high-temperature resistance. Compared with the blank sample, with the exception of the Evotherm warm-mix additive, the other three warm-mix technologies, especially Sasobit, increased the $G^*/\sin\delta$ values of the HVMAs. The three warm-mix technologies enhanced the high-temperature stability of HVMA, and the Sasobit warm-mix additive significantly improved the rutting resistance of

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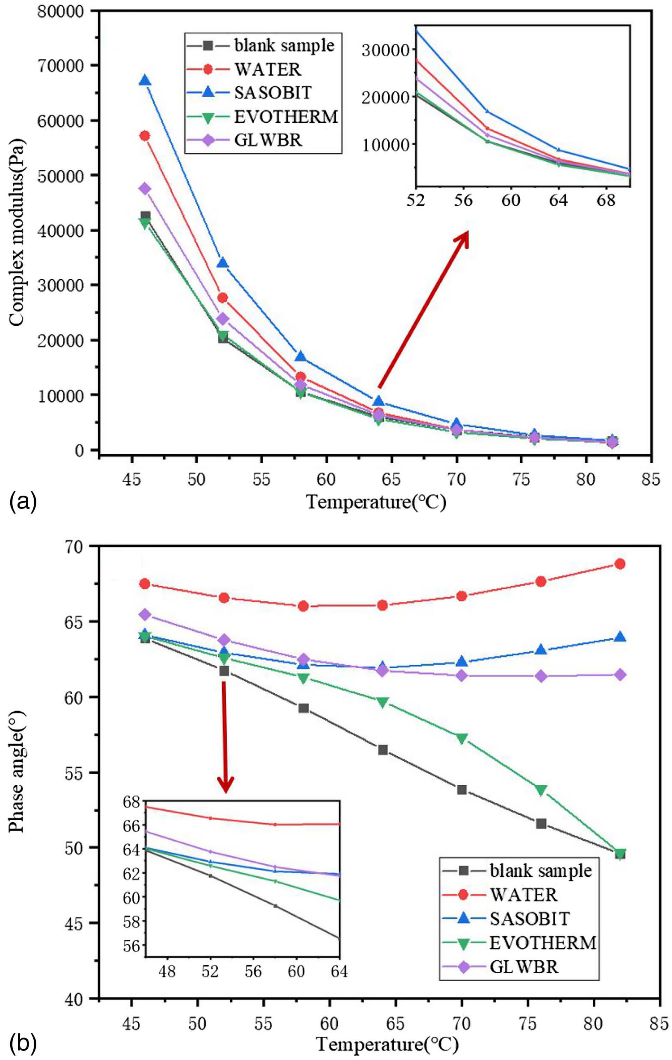

Fig. 8. The G^* and δ values of different warm-mix HVMAs in the temperature range of 42℃ to 82℃: (a) G^*; and (b) δ.

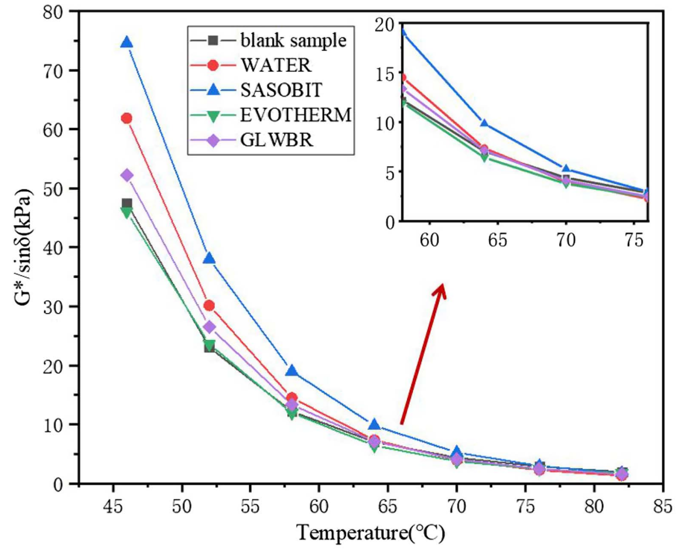

Fig. 9. The $G^*/\sin\delta$ values of different warm-mix HVMAs in the temperature range of 42℃ to 82℃.

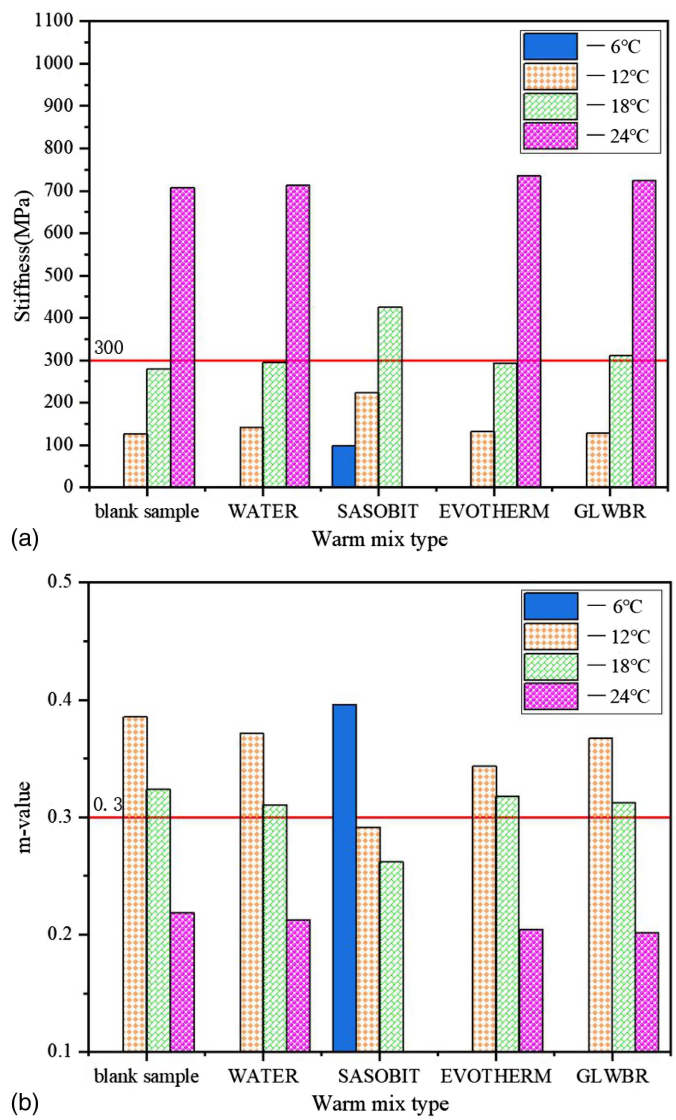

Fig. 10. Creep stiffness (S) and creep rate (m) of different warm-mix HVMAs at different temperatures: (a) S; and (b) m.

the asphalt mixture. This result was consistent with the results for G^* and δ values and the conclusions of Zheng et al. (2019).

Creep Stiffness and Creep Curve Slope

Fig. 10 shows the low-temperature creep behaviors of the HVMAs. The S and m values of the warm-mix HVMAs increased and decreased, respectively, with the decrease of temperature, respectively. Compared with the blank sample, the S and m values of the warm-mix HVMAs increased and decreased, respectively (Qin et al. 2014; Yu et al. 2017). This was especially obvious for SASOBIT. Adding the Sasobit warm-mix additive to HVMA was not conducive to the low-temperature performance of HVMA. However, WATER, EVOTHERM, and GLWBR had no significant effects on the low-temperature performance of the HVMAs.

Analysis of Performance Grade

Based on DSR and BBR test results, the study followed the requirements of modified asphalt PG classification method in the US SHRP (1994) specification ($G^*/\sin\delta \geq 1.0$ kPa for unaged asphalt, $G^*/\sin\delta \geq 2.2$ kPa for aged asphalt, $S \leq 300$ MPa, m value ≥ 0.3) (Zhang et al. 2019). PG grade of warm-mix HVMA is shown in Table 6. Compared with the blank sample, the warm-mix HVMAs in high- and low-temperature PG tests showed

Table 6. Continuous PG results of warm-mix HVMAs

Sample	Critical temperature ($G^*/\sin\delta \geq 1.0$ kPa for original HVMA, °C)	Critical temperature ($G^*/\sin\delta \geq 2.2$ kPa for TFOT residue, °C)	High continuous grading temperature	Critical temperature ($S \leq 300$ MPa)	Critical temperature (m value ≥ 0.3)	Low continuous grading temperature	Continuous grade
Blank sample	86.4	86.6	86.4	−26.9	−28.7	−26.9	86.4–26.9
WATER	83.3	85.1	83.3	−25.5	−27.5	−25.5	83.3–25.5
SASOBIT	84.4	84.8	84.4	−24	−23.5	−23.5	84.4–23.5
EVOTHERM	86.1	85	85	−25.6	−27	−25.6	85.0–25.6
GLWBR	84.6	85.9	84.6	−29.3	−26.4	−26.4	84.6–26.4

lower and higher continuous gradations, respectively, indicating that the warm-mix additives reduced rutting and protected the low-temperature resistance of asphalt to a certain extent. The introduction of warm-mix technology reduced the performance grade of the HVMAs. WATER had great influence on the high-temperature performance of HVMA, and Sasobit had obvious effects on the low-temperature performance of HVMA.

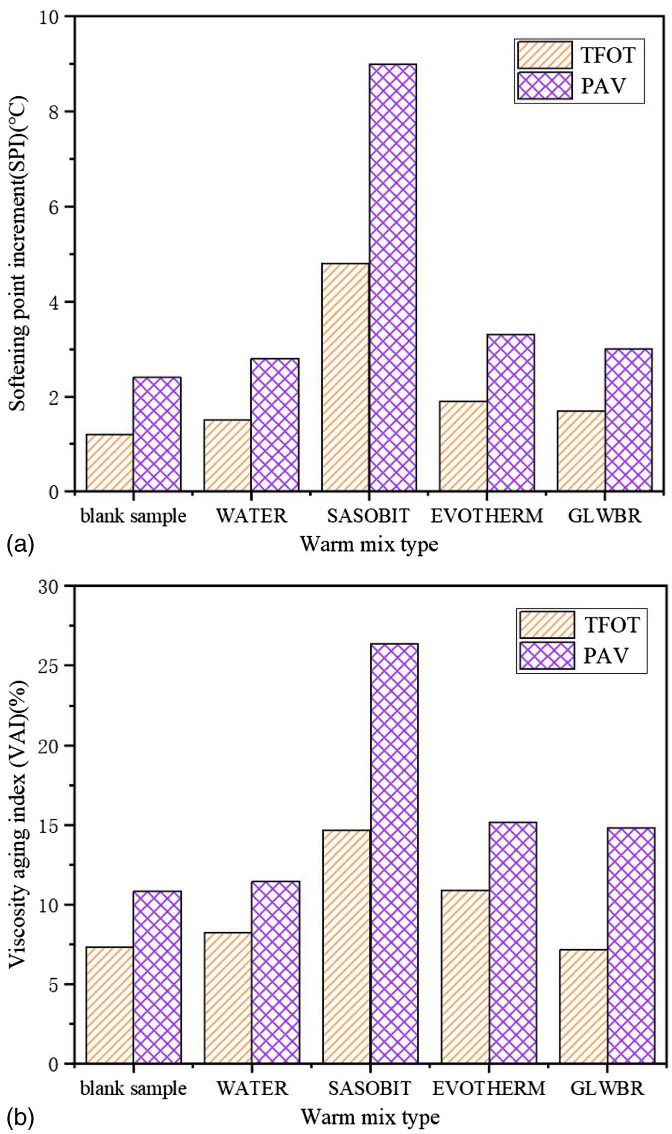

Fig. 11. SPI and VAI values of different warm-mix HVMAs after TFOT and PAV: (a) SPI; and (b) VAI.

Analysis of Antiaging Properties

Evaluation of Physical Aging Indices

Fig. 11 shows the variations of physical aging indexes [softening point increment (SPI) and viscosity aging index (VAI)] after applying two aging methods on the HVMAs. Compared with the blank samples, the SPI and VAI values of the HVMAs mixed at different temperatures after TFOT were higher. The PAV results were consistent with the TFOT results. From TFOT to PAV, SPI and VAI of warm mix HVMA increased. In addition, after TFOT and PAV,

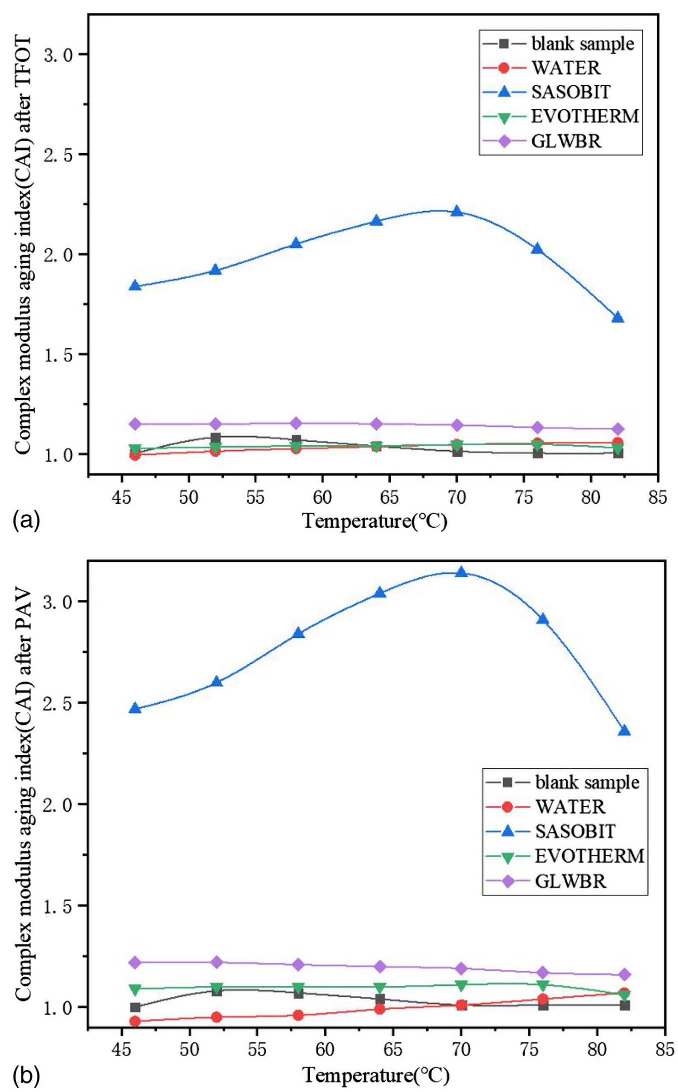

Fig. 12. CAI values of different warm-mix HVMAs after: (a) TFOT; and (b) PAV.

the SPI aging indices of the different warm-mix HVMAs were SASOBIT > EVOTHERM > GLWBR > WATER, and the VAI aging indices of the different warm-mix HVMAs were SASOBIT > EVOTHERM > GLWBR > WATER. The changes in the SPI and VAI aging indices indicated that the addition of Sasobit warm-mix additive significantly increased the short-term and long-term aging of warm-mix HVMA. This was due to the wax crystals in Sasobit, which increased the heat absorption rate of HVMA during the short- and long-term aging processes (Rodríguez-Alloza et al. 2017). For WATER, the majority of the foaming water was volatilized due to high temperatures during foaming, and small amounts of residual water had no obvious effect on the aging behavior of HVMA during thermal oxidation (Dong et al. 2018). For Evotherm and GLWBR, which are surfactant-based warm-mix additives, water molecules in the periphery of the warm-mix additive surfactant micelles evaporated rapidly in the process of thermal aging such that the lipophilic groups contacted the asphalt, increasing asphalt aging degree (Wan et al. 2020).

Analysis of Rheological Aging Indices

Fig. 12 shows complex modulus aging index (CAI) changes in the different warm-mix HVMAs after TFOT and PAV aging. The CAI value of the different warm-mix HVMAs increased with PAV compare to TFOT. This indicates that, compared with short-term thermal aging, long-term thermal aging has a greater effect on the G^* value of HVMA. According to Fig. 12(a), compared with the blank sample, the CAI values of the Sasobit- and GLWBR-treated HVMAs were significantly higher, in the temperature range of 46°C to 82°C; this effect was especially strong for the SASOBIT mix. Between 46°C and 64°C, the CAI values of the water- and Evotherm-treated HVMAs were smaller than that of the blank sample. Also, between 64°C and 82°C, the CAI values of the water- and Evotherm-treated HVMAs were higher than that of the blank sample. According to Fig. 12(b), with the exception of WATER, the CAI values of the warm-mix HVMAs were greater than that of the blank sample in the temperature range of 46°C to 82°C; this was especially true for SASOBIT. At a temperature of less than 70°C, the CAI value of WATER was lower than that of the blank sample. When the temperature was higher than 70°C, however, the CAI value of WATER was higher than that of the blank sample.

Figs. 13(a and b) show the phase angle aging index (PAI) changes of different warm-mix HVMAs after TFOT and PAV aging, respectively. According to Fig. 13, after TFOT or PAV aging, the PAI values of the different warm-mix HVMAs were significantly

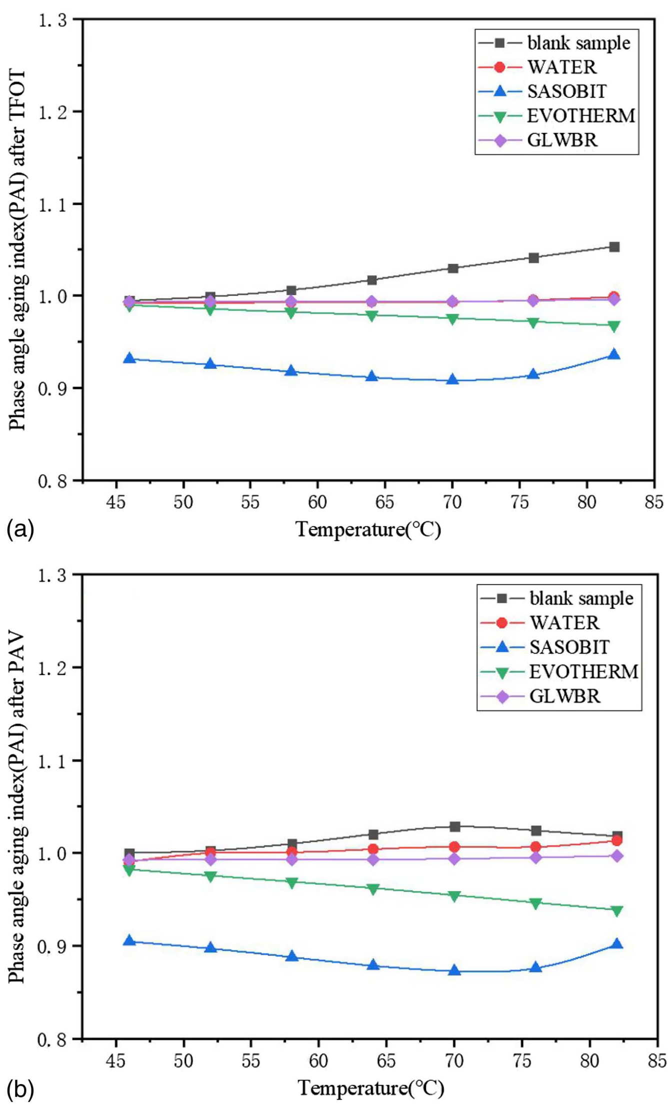

Fig. 13. PAI values of different warm-mix HVMAs after: (a) TFOT; and (b) PAV.

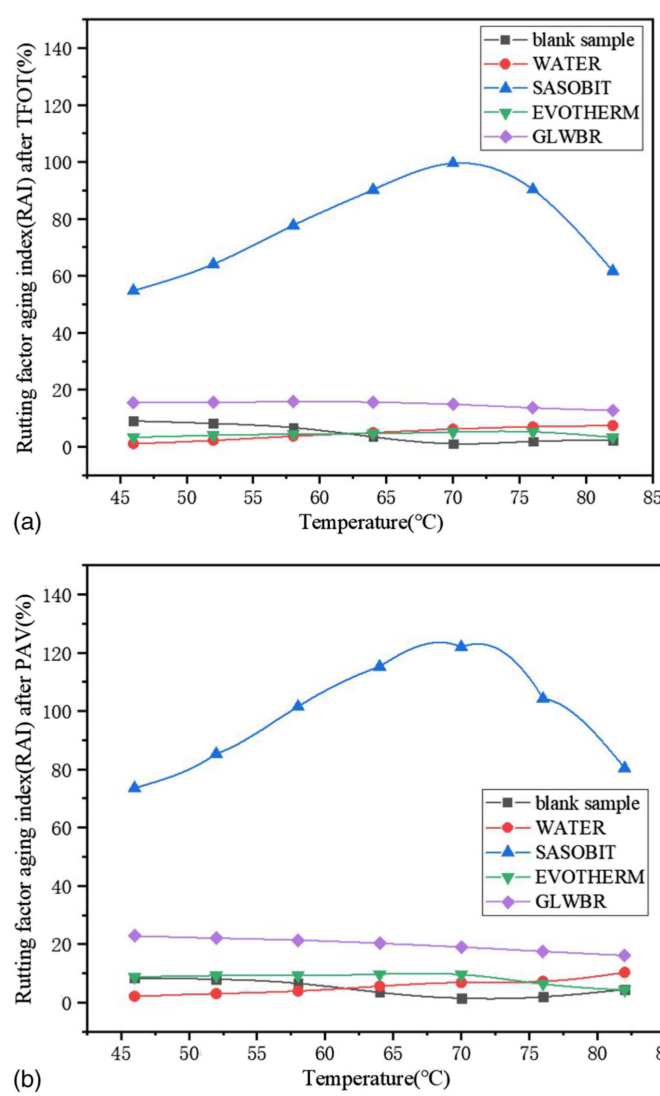

Fig. 14. RAI of different warm-mix HVMAs after: (a) TFOT; and (b) PAV.

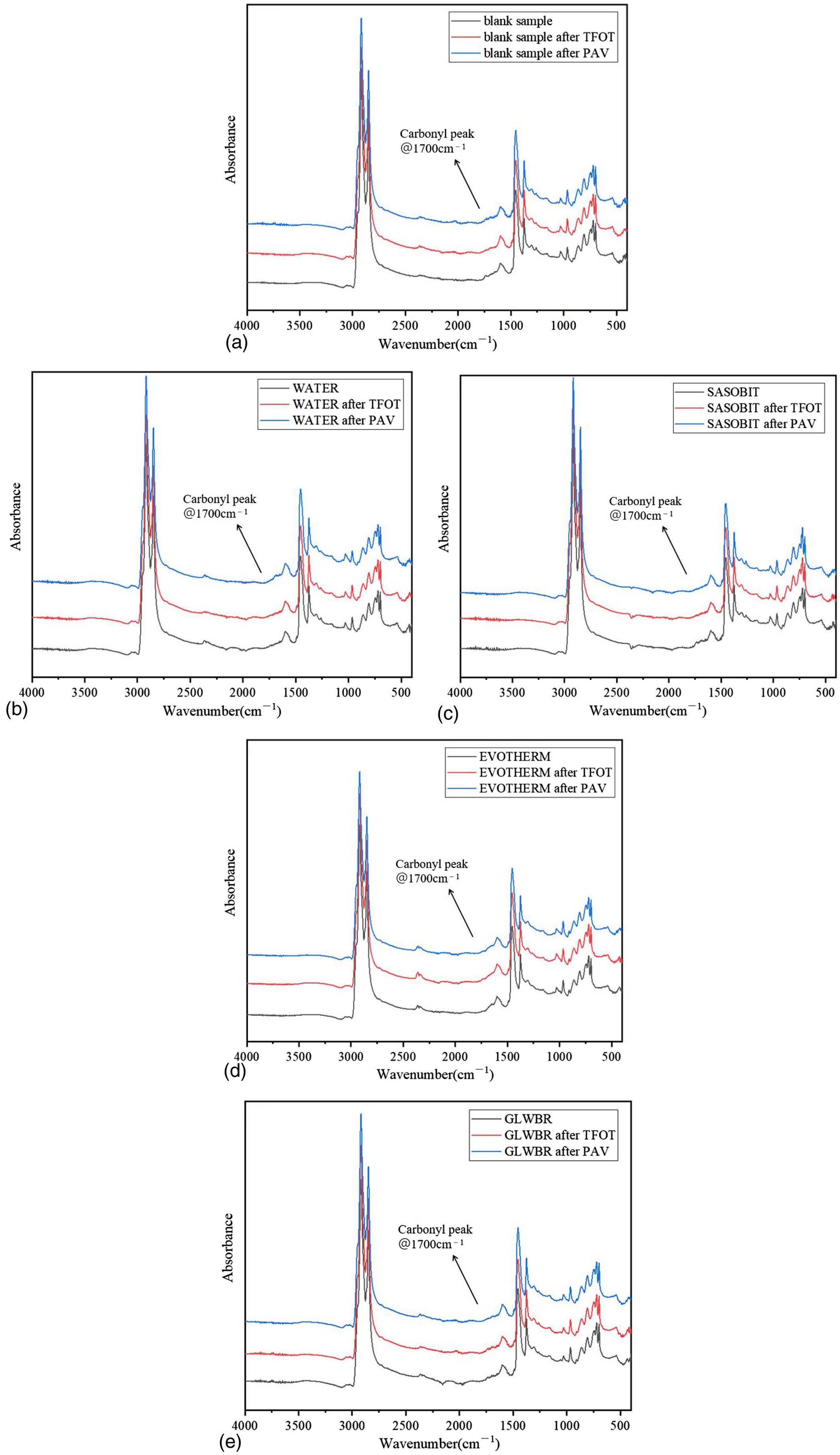

Fig. 15. FTIR spectra of binders before and after two aging methods: (a) blank sample; (b) WATER; (c) SASOBIT; (d) EVOTHERM; and (e) GLWBR.

Table 7. Carbonyl index of binders before and after two aging methods

Sample	Blank sample		WATER		SASOBIT		EVOTHERM		GLWBR	
	CI	ΔCI	CI	ΔCI	CI	ΔCI	CI	ΔCI	CI	ΔCI
Unaged	0.0521	—	0.0521	—	0.0494	—	0.0506	—	0.0506	—
TFOT aging	0.0527	0.0005	0.0523	0.0012	0.0534	0.004	0.0526	0.002	0.0526	0.002
PAV aging	0.0539	0.0018	0.0581	0.0035	0.0561	0.0067	0.0541	0.0061	0.0545	0.004

lower than that of the blank sample. In addition, the PAI values of the HVMAs mixed at different temperatures were as follows: SASOBIT > EVOTHERM > GLWBR > WATER. This corresponded to the CAI values. This result was due to the different effects of the main components of the different warm-mix additives on the aging of HVMA (Rodríguez-Alloza et al. 2017). In addition, it was particularly shown that, unlike Evotherm, GLWBR had no obvious effect on the antiaging performance of HVMA.

As shown in Fig. 14, after TFOT and PAV aging, the rutting factor aging index (RAI) values of HVMA at different temperatures were different. This was because the RAI values of HVMA mixed at different temperatures were higher with PAV than with TFOT aging, especially for SASOBIT. Aging by TFOT or PAV had a great influence on changes in RAI values for the different warm-mix HVMAs, indicating that the thermal oxidation method had a great influence on the rutting resistance of the different warm-mix HVMAs. The RAI values of the Sasobit- and GLWBR-treated HVMAs were significantly higher than that of the blank sample; this was especially obvious for SASOBIT. The RAI values of the WATER and EVOTHERM warm-mix HVMAs changed with aging mode, but these changes were not large, indicating that the mixing temperature of WATER and EVOTHERM had little effect on the aging resistance of HVMA.

FTIR Analysis

Fig. 15 shows the FTIR spectra of warm-mix HVMAs before and after the application of the two aging methods. Compared with the blank sample, the absorption peak areas of the EVOTHERM and GLWBR warm-mix HVMAs at 2,925, 1,460, and 700 cm^{-1} were higher. This was due to the overlapping of the absorption peaks of HVMA with those of the Evotherm and GLWBR warm-mix additives. The absorption peak areas of the WATER and SASOBIT warm-mix HVMAs showed no obvious changes, because the WATER warm-mix additive was mainly composed of water and the SASOBIT mix's additive was mainly composed of wax. The aging degree of the warm-mix HVMAs gradually deepened and the areas of the absorption peaks of the warm-mix HVMAs at 1,700 cm^{-1} were higher with PAV compared to TFOT. This phenomenon was consistent with the change rule of ΔCI.

The carbonyl indices of the warm-mix HVMAs before and after the application of the two aging methods are summarized in Table 7. The CI and ΔCI value of the different warm-mix HVMAs increased with PAV compare to TFOT. Compared with the blank sample, the CI and ΔCI values of the different warm-mix HVMAs were significantly higher. After TFOT aging, the △CI order of the asphalt binders was found to be SASOBIT > EVOTHERM > GLWBR > WATER > blank sample; the corresponding order for PAV aging was SASOBIT > EVOTHERM > GLWBR > WATER > blank sample. Increased values of CI and ΔCI for the asphalt binders resulted in higher aging degrees of the warm-mix HVMAs. The △CI value of SASOBIT was the largest, indicating that Sasobit had the most serious aging effect on HVMA. This was because the main ingredient of Sasobit was wax, a light ingredient. Therefore, when the light component was thermally oxidized, it was volatilized or transformed into a relatively recombined component under the influence of heat, leading to the aging of the asphalt binder (Sol-Sánchez et al. 2018).

Correlation Analysis

In previous studies, increase of carbonyl index has been considered as an important indicator of asphalt aging degree (Morian et al. 2015). Therefore, △CI index can be applied to characterize asphalt aging degree. The date sets used for correlation analysis with △CI included aging index (SPI, VAI, and CAI at 70°C, PAI at 70°C, and RAI at 70°C), as shown in Fig. 16 and Table 8. According to Table 8, compared with the blank sample, the △CI indices of the warm-mix HVMAs after TFOT or PAV aging increased

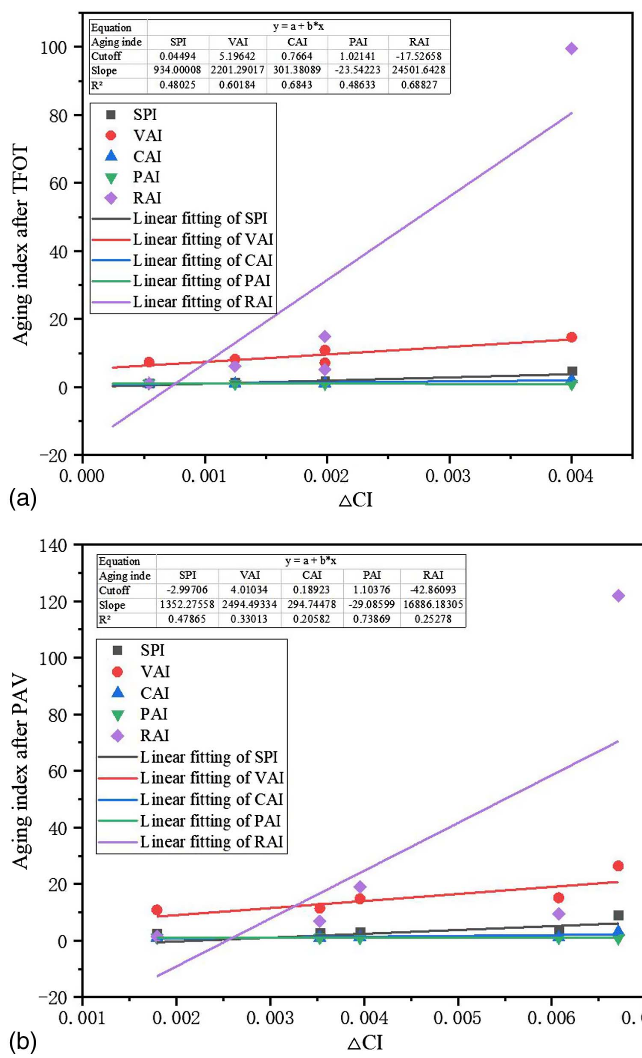

Fig. 16. Correlation analysis of ΔCI with the overall results of the physical and rheological tests: (a) TFOT; and (b) PAV.

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Table 8. Data sets for correlation analysis

Sample name	ΔCI	SPI	VAI	CAI	PAI	RAI
Blank sample + TFOT	0.000541	1.2	7.34	1.01	1.03	1.04
WATER + TFOT	0.001246	1.5	8.21	1.05	0.99	6.19
SASOBIT + TFOT	0.004002	4.8	14.67	2.21	0.91	99.52
EVOTHERM + TFOT	0.001982	1.9	10.89	1.05	0.98	5.17
GLWBR + TFOT	0.001982	1.7	7.14	1.15	0.99	14.91
Blank sample + PAV	0.001793	2.4	10.84	1.01	1.03	1.53
WATER + PAV	0.003527	2.8	11.44	1.01	1.01	6.82
SASOBIT + PAV	0.006710	9	26.36	3.14	0.87	121.92
EVOTHERM + PAV	0.006074	3.3	15.14	1.11	0.95	9.57
GLWBR + PAV	0.003956	3	14.8	1.19	0.99	19.04

significantly, which was consistent with changes in asphalt aging indices. After TFOT aging, ΔCI index had a certain correlation with the aging index of asphalt, and the correlation coefficient R^2 was between 0.48 and 0.7. After PAV aging, PAI was strongly correlated with ΔCI index ($R^2 = 0.739$), SPI ($R^2 = 0.479$), VAI ($R^2 = 0.330$), CAI ($R^2 = 0.206$), and RAI ($R^2 = 0.253$). Based on the aforementioned analyses, the relationships among VAI, CAI at 70°C, and RAI at 70°C and the ΔCI indices of the warm-mix HVMAs after TFOT aging were closer than those of SPI and PAI at 70°C. Therefore, the aging degrees of the warm-mix HVMAs aged by TFOT can be predicted by VAI, CAI at 70°C, and RAI at 70°C. The relationship between PAI at 70°C and the ΔCI index of the warm-mix HVMAs after PAV aging was closer than the relationships among SPI, VAI, CAI at 70°C, and RAI at 70°C and the ΔCI index. Therefore, the aging degree of warm-mix HVMAs aged by PAV can be predicted by PAI at 70°C. The correlations found between aging indices and the ΔCI index of the warm-mix HVMAs were consistent with the findings of Wang et al. (2020).

Conclusions

This paper evaluated the effects of warm-mix additives on the aging and chemical and rheological properties of HVMA. The following conclusions can be drawn:

1. Warm-mix technologies reduced the viscosity of HVMA so as to achieve a warm-mix effect. Warm-mix technologies improved the softening point and G^*, $G^*/\sin\delta$, and S values of the HVMAs; they also reduced penetration, ductility, and m value.
2. The influence of the warm-mix technologies on the aging performance of the HVMAs in order of strength was Sasobit > Evotherm > GLWBR > water > blank sample.
3. Foam warm-mix technology had the strongest effect on warm-mix HVMA but had little effect on the antiaging ability of HVMA. Therefore, foam warm-mix technology is recommended to be used in the warm mixing technology of HVMA.
4. Aging indices VAI, CAI at 70°C, and RAI at 70°C and PAI can be used to predict the aging degree of warm-mix HVMAs after short-term and long-term thermal oxidation, respectively.
5. Compared with Evotherm, GLWBR had warm-mix effect on HVMA but had less effect on HVMA aging.

Data Availability Statement

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgments

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