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Experimental research on combined effects of flame retardant and warm mixture asphalt additive on asphalt binders and bituminous mixtures

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HIGHLIGHTS

- Viscosity reducing and the influence of Sasobit® on temperature susceptibility are related to the types of base asphalt.
- Flame retardant (ZIR®) and Sasobit® improve significantly high temperature performance of asphalt binders.
- ZIR® and Sasobit® have influences on indices of low temperature performance of bituminous mixtures for creep tests.
- Using $m(t)/S(t)$, t and $W_d(t)/W_s(t)$ to describe low temperature performance for creep tests can be more reasonable than only using m-value.

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ABSTRACT

The application of bituminous mixture modified by flame retardant (FR) and warm mixture asphalt additive (WMAA) (BMMFW) is becoming a trend in tunnel pavements. When FR improves the flame retardancy of asphalt binder and WMAA reduces the construction temperatures of bituminous mixture, the combined effects of the two additives on pavement performances need to be studied. In this study, two types of original asphalt, FR (ZIR®) and WMAA (Sasobit®) were chosen. Through conventional performance tests and high and low temperature rheological tests, the experimental research was focused on the pavement performance investigation of original asphalt binder, asphalt binder modified by FR and WMAA (ABMFW), base bituminous mixture and BMMFW. The results show that the used FR and WMAA can improve the high temperature properties of asphalt binder and bituminous mixture, reduce the low temperature performance but have slight negative effects on water stability. The effects of WMAA on reducing viscosity are related with asphalt species, and whether the influence of the FR and WMAA on the absolute value of viscosity–temperature index is increasing or decreasing also depends on the type of original asphalt. An interesting and important phenomenon is founded, which is that using the creep rate $m(t)$ to describe the low temperature performance of asphalt binder or bituminous mixture may sometimes be insufficient or unreliable, while employing the $m(t)/S(t)$ (ratio of creep rate to the stiffness at time t), stress-relaxation time and $W_d(t)/W_s(t)$ (ratio of dissipated energy to stored energy at time t) may be reasonable and accurate.

1. Introduction

With the continuous development of highway tunnel construction in China, higher requirements on comfort of tunnel pavement are put forward. Because of the advantages of comfort, low noise, less dust and easy maintenance and so on, asphalt pavement is becoming more and more a tunnel pavement [1]. But because of the tunnel space limitation, the safety problem caused by accidents and construction environment issue should be considered when utilized asphalt pavement. In this case, applying flame retardant (FR) and warm mixture asphalt additive (WMAA) into bituminous mixture at the same time to improve flame retardancy and reduce construction temperatures of bituminous mixture may be as a technical measure [2]. There are many species of FR and WMAA, and different kinds of FR and WMAA can have certainly different modifying effects. Moreover, when FR and WMAA are applied simultaneously, chemical reaction may be caused because of composing components. So, for the preparation of bituminous mixture modified by FR and WMAA (BMMFW), correct selection of FR and WMAA is very important. In addition, the using of the two additives must have effects on pavement performances of bituminous mixture, which needs to be investigated and mastered although the primary goal of using FR and WMAA is to avoid fire, save energy and protect environmental protection.

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The effects of FR on pavement performances of bituminous mixture were also studied by many researchers while they paid attention to the improvement of FR on fire-retardant property. Based on Marshall stability, retained Marshall stability, flow value, indirect tensile strength test (ITS) and rutting resistance index, some researchers studied the effect of mixed flame retardants formed by antimony trioxide, zinc borate and Antimony trioxide on bituminous mixture. Their conclusion is that it is feasible to prepare the excellent pavement of bituminous mixture using flame retardant modified asphalt binder compared with the control bituminous mixture and asphalt binder containing mixed flame retardants [3,4]. While flame retardant of magnesium hydroxide (MH) can increase the Marshall stability and decrease the loss of Marshall stability. Also, MH improves the initial and conditional ITS of the bituminous mixture, but results in slightly larger ITS loss, and it may not affect the moisture damage resistance of bituminous mixture [5]. The research results from Wu et al. [6] indicate that flame retardant modified asphalt binders show an increase in the complex modulus compared to the origin asphalt binder, especially at relatively low temperatures. Warm mix asphalt (WMA) technologies are hot engineering problems in recent years, which can be, basically, classified in three main groups [7–11]: organic additives, chemical additives and foaming technologies. Sasobit® is a typical kind of WMAA, and related researches and applications are the most common. Based on more than 230 research literatures, Jamshidi et al. [12] have finished a full review of Sasobit® on pavement performance of asphalt binder and bituminous mixture. Except the viscosity reducing effect of Sasobit®, the influence on some typical physical properties is as follows: Sasobit® increases $G^*/\sin\delta$ (where G^* is complex shear modulus and δ is the phase angle) and zero shear viscosity; it decreases the penetration number, Fraass breaking point irrespective of binder source and the creep compliance of asphalt binder for a given aging state and binder type and source [13–24]. Xiao F finds Sasobit® increases $G^*/\sin\delta$ and failure temperature and reduces creep compliance and phase angle more than other warm asphalt binder additives [14]. Akisetty et al. [25,26] observe that the WMA additives (Aspha-min® and Sasobit®) increase the high temperature performance values of the binders; Wang et al. [26] find in their study that, the three WMA additives (Sasobit®, RH® and Advera®) can all improve the CRM binders' resistance to rutting. However, because of the lower production temperatures, as well as moisture susceptibility of WMA, rutting may be mainly caused by the less ageing of the binder, and this can result in the premature rutting of the pavement surface [8]. Besides, due to the relative newness of WMA, field test sections are still few in number, and they also have a short life. For this reason, it is not as yet possible to talk about long term performance [8]. Lee et al. [27] study the recycled WMA binders (containing Sasobit® and Aspha-min® respectively), which are observed to have significantly lower resistance on low temperature cracking (measured by the BBR test). Zhao et al. [28] study WMA-high RAP mixtures, and they find that WMA shows lower rutting resistance than corresponding HMA mixtures regardless of WMA technology, RAP content and structural layer, and WMA-high RAP surface mixtures prove insusceptible to moisture regardless of WMA technology along with cracking and fatigue may not be a major concern when it comes to WMA-high RAP technology.

Although there have been some application researches on flame retardant and WMA in China, system research reports about the combined effects that FR and WMAA act on pavement performance of bituminous mixture at the same time are fewer. Moreover, different FR and WMAA have different influences on asphalt binders and bituminous mixtures; it is necessary to give a system research for the used FR, WMAA and asphalt binders.

The primary object of this paper is to investigate the combined effect of the selected FR and WMAA on conventional properties of different binders, and systematically evaluate the pavement performance of asphalt binders modified by the FR and WMAA (ABMFW) and bituminous mixture modified by the FR and WMAA (BMMFW).

2. Materials and test programme

2.1. Materials

2.1.1. Binders

AH-70 petroleum asphalt modified by SBS (marked as B1) and rubber asphalt (marked as B2) were selected as original asphalts to develop ABMFWs. The rubber asphalt was prepared with the AH-70 petroleum asphalt and waste rubber powder of 60-mesh, and the quality proportion was 15% (the quality percentage of waste rubber powder to AH-70 petroleum asphalt). Conventional performance indices of the two binders are shown in Table 1.

2.1.2. Fire retardant and WMA additive

A common FR (its trademark is ZIR®) was selected, which is easy to be seen in market. It is white powder; its density is greater than 2.0 g/cm^3, and its grain size is less than 10 μm. The endothermic and decomposition temperature of the FR are larger than 250 and 270 °C, respectively. The FR dose used in this study is 4% of asphalt quality by mass.

The used WMA additive (WMAA) is Sasobit®. It is widely used as a viscosity reduction material. The dose of WMAA is determined as 3% of asphalt quality by mass in this study.

So, two kinds of ABMFW were obtained; their compositions were B1+4%FR+3%WMAA (marked as ZW-B1) and B2+4%FR+3%WMAA (marked as ZW-B2), respectively.

Whether FR and WMAA have good compatibilities in asphalt should be taken into consideration, because chemical reaction may occur between them in asphalt in high temperature condition, bring about dangers. The tentative tests in this research showed that there could be chemical reaction between some specific FRs and WMAAs (several kinds of FRs and WMAAs were prepared through many manufacturers) in asphalt media, and gases with intense irritant smell and a great deal of bubbles were produced, which made the volume of melting asphalt get bigger. This phenomenon can not only reduce the effect of FR and WMAA, but also cause hidden dangers to construction, which must be avoided. Fortunately, when heated original asphalt binder blended with ZIR® and Sasobit®, no chemical reaction was found, so the effect and safety could be guaranteed.

2.1.3. ABMFW preparation

The WMAA will rapidly melt when temperature is above 120 °C and can be well mixed with hot asphalt. Meanwhile, the adding of WMAA can improve the flow state of asphalt, making the FR be dispersed more easily. So, the WMAA should be firstly added into original asphalt in this research; then followed by the FR. The detailed preparation programs of asphalt modified by ZIR® and Sasobit® were as follows: took a certain quality asphalt to a heated state (about 150 °C); added the WMAA into it according to the determined dose (3%) and stirred 2 min by electric mixer with speed of 500 r/min, then added the FR with the quality by 4% dose and stirred 20 min, and in last the two additives were mixed uniformly with original asphalts and the ABMFW were prepared.

2.1.4. Design of BMMFW

The ZW-B1 bituminous mixture whose nominal maximum size of aggregate was 13 mm was used to investigate the effects of the FR and WMAA on pavement performance. The aggregate gradation was designed according to some ideas of Superpave system, and the optimum asphalt content was determined by Marshall Test method [29]. Being omitted the detailed design process of the mixtures, the design results were shown as follows: aggregate (basalt) gradation was shown in Fig. 1, and the optimum asphalt content was 4.8% (the FR and WMAA contents were not included).

The mixing and compaction temperature of BMMFW were 150 °C and 133 °C, respectively.

Table 1
Conventional properties of the two binders.

Binder type	Penetration number at 25 °C (0.1 mm)	Softening point/$T_{R\&B}$ (°C)	Ductility at 5 °C (5 cm/min) (cm)
B1	49.8	65.7	34.7
B2	55.2	54.2	7.3

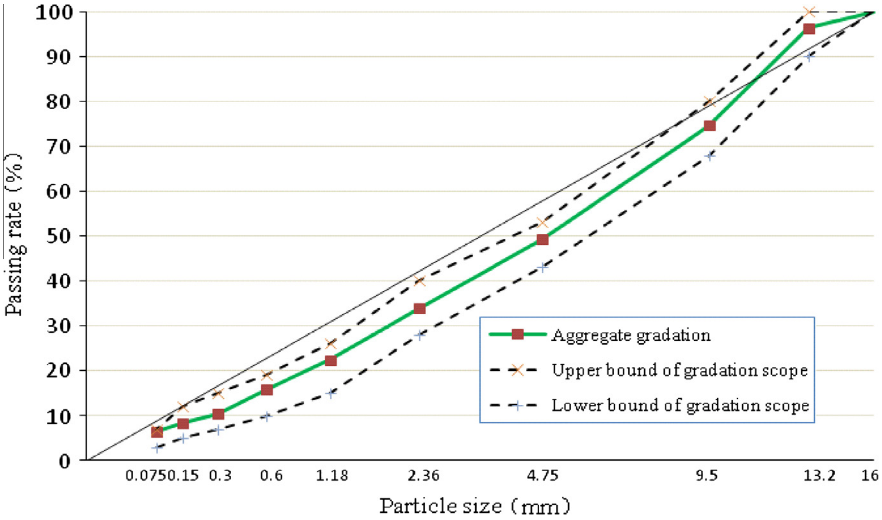

Fig. 1. Gradation design curve.

2.2. Test program

2.2.1. Limiting oxygen index (LOI) test

Limiting oxygen index (LOI) is the minimum concentration of oxygen expressed as volume percent in a mixture of oxygen and nitrogen that just supports flaming combustion of a material initially at room temperature. It is measured by passing a mixture of oxygen and nitrogen over a burning specimen [30]. The bigger LOI is, the more difficult material burn.

2.2.2. Viscosity test

A Brrokfidd viscometer (made in the United States) was utilized to measure the viscosities of each ABMFW at 120, 135, 155 and 175 °C. The test was conducted by ASTM D4402. The 27th rotor was employed during the testing, and the rotational speed was 20 rpm.

2.2.3. Pavement performance test

2.2.3.1. For asphalt binders. The evaluation of combined effect of the used FR and WMAA on pavement performance of original asphalt binder was mainly based on viscosity–temperature index, softening point, low temperature ductility and high temperature shear rheological properties. All these tests were conducted according to the stand test methods [31].

2.2.3.2. For bituminous mixtures. By rutting test, water stability test, bending test and bending rheological test at low temperatures, the effects of the FR and WMAA on bituminous mixture pavement performance were investigated, and pavement performances of the BMMFW were evaluated. All these tests were conducted according to the stand test methods [31,32]. The rutting test temperature was 60 °C.

The dimension of beam specimens used in bending test at low temperatures is as follows: length × width × height equals to (250 ± 2 mm) × (30 ± 2 mm) × (35 ± 2 mm), and the span is 200 mm, as shown in Fig. 2. For dense gradation bituminous mixtures, the test temperature is −10 °C. The concentrated load is loaded on mid-span of the specimens; the loading rate is 50 mm/min. The maximum load is determined when a tested specimen is destroyed, and then the flexural tensile strength, bending tensile strain and bending stiffness modulus can be calculated by Eqs. (1) and (3), respectively.

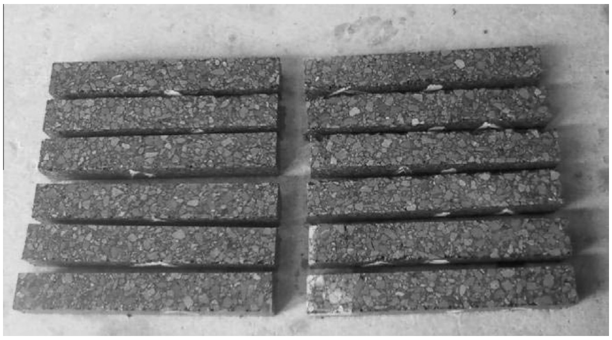

Fig. 2. Specimens for bending tests at low temperatures.

$$R_B = \frac{3LP_B}{2bh^2} \tag{1}$$

$$\varepsilon_B = \frac{6hd}{L^2} \tag{2}$$

$$S_B = \frac{R_B}{\varepsilon_B} \tag{3}$$

where R_B is flexural tensile strength when specimen damaged, MPa; ε_B is maximum bending tensile strain when specimen damaged; S_B is bending stiffness modulus when specimen damaged, MPa; b is width of mid-span section, mm; h is height of mid-span section, mm; L is span length of specimen, mm; P_B is the maximum load when specimen damaged, N; d is mid-span deflection when specimen damaged, mm.

The test temperature of bending creep test at low temperature was 0 °C, and the used specimen size was the same as that of bending test at low temperature. The time consuming of test was depended on the condition of specimen deformation, and the moment into failure stage was regarded as the deadline. When the test has lasted for 2 h (7200 s) but the specimen still undamaged, stop the test.

3. Results and analysis

3.1. The results and analysis of ABMFW

3.1.1. Limiting oxygen index

The oxygen index test was carried out in accordance with the stand test methods [30]; the results were shown in Table 2.

Compared with those of the original asphalt binders, the limited oxygen indices of the two kinds of ABMFW were improved obviously, as shown in Table 2, and the increasing degrees were 21.9% and 24%, respectively. The results showed that the addition of FR (in the presence of the WMAA) greatly reduced the burning capabilities of the two original binders.

3.1.2. Viscosity

To study viscosity–temperature relationship of asphalt binder is one of the basic contents of rheology. In a certain temperature

Table 2
Limiting oxygen indices of original binders and ABMFWs.

Types of binder	B1	ZW-B1	B2	ZW-B2
Oxygen index (%)	20.1	24.5	20.4	25.3

range, the trend that viscosity changes with temperatures directly reflects the temperature susceptibility of the binder. The regression line can be drawn according to the viscosity values at different temperatures in the double logarithm coordinate, the slope of which represents the temperature susceptibility of asphalt binder in the tested temperature range. The most widely used regression formula that describes the viscosity–temperature relationship is the Eq. (4) which Walther and Saal have established, and it is as follows [33].

$$\lg\lg(\eta) = N - M\lg(T + 273.13) \tag{4}$$

where η is viscosity, MPa s; T is centigrade degree (°C); M, N is regression coefficients; while M is the slope of the viscosity–temperature line, representing the temperature susceptibility, that is the viscosity temperature index.

According to the viscosity test results about the two original asphalt binders and two kinds of ABMFW at different temperatures, the regression line could be obtained as shown in Fig. 3, and the regression equations were shown in Table 3.

It can be seen from Fig. 3 that, in the presence of the FR the WMAA Sasobit® with the same dose brings about different effects on reducing viscosity for different asphalt binders. For binder B1, the decreased degrees of viscosity by Sasobit® at different temperatures are not large and different, being from 9% to 17.5% (the average value is 11.6%). The absolute value of viscosity–temperature index of the ABMFW (that is ZW-B1) increases, showing that the WMAA and FR have slightly negative effects on asphalt sensitive-temperature property of B1. However, compared with that of B1 the WMAA (in the presence of the FR) reduced the viscosity of B2 greatly in the whole experimental temperature range, being from 36.9% to 51.6% (average valve is 43.8%). The absolute value of viscosity–temperature index of the ABMFW (that is ZW-B2) reduces, reflecting that the WMAA and FR have some positive effects on asphalt sensitive-temperature property of B2.

3.1.3. Conventional properties

The conventional properties conclude needle penetration at 25 °C, softening point, and low temperature ductility at 5 °C which are shown in Table 4.

Comparing the results in Tables 4 and 1, it can be found that, for the two ABMFWs, the penetration number values at 25 °C are smaller and the softening point values are larger respectively than those of the original asphalt binders, and the ductility values at 5 °C reduce dramatically, demonstrating that the two additives can obviously improve the high temperature properties and drop the low temperature properties.

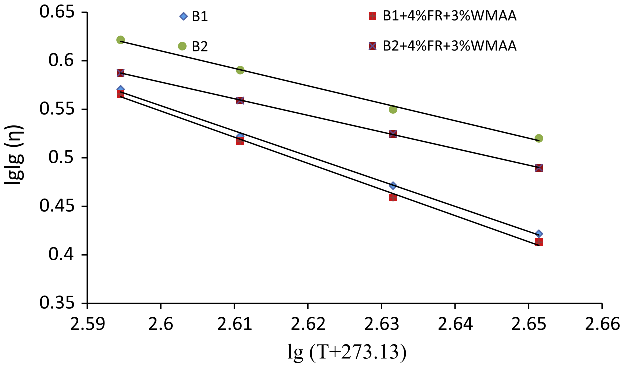

Fig. 3. Lines of viscosity–temperature relationship between original asphalt binders and ABMFWs.

Table 3
Viscosity–temperature equations of original asphalt and ABMFWs.

Binder	Regression equation	R^2	Viscosity–temperature index (M)
B1	$\lg\lg(\eta) = 7.2935 - 2.5922\lg(T + 273.13)$	0.9982	−2.5922
ZW-B1	$\lg\lg(\eta) = 7.5523 - 2.6939\lg(T + 273.13)$	0.9970	−2.6939
B2	$\lg\lg(\eta) = 5.2890 - 1.7995\lg(T + 273.13)$	0.9966	−1.7995
ZW-B2	$\lg\lg(\eta) = 5.0297 - 1.7122\lg(T + 273.13)$	0.9998	−1.7122

Table 4
Conventional properties of two base binders.

Binder species	Penetration number at 25 °C (0.1 mm)	Softening point TR&B (°C)	Ductility at 5 °C (5 cm/min) (cm)
ZW-B1	38.4	84.9	11.9
ZW-B2	36.6	72.3	4.1

3.1.4. Complex modulus and phase angle

In order to investigate the influence of the FR and WMAA on shear rheological properties of the binders at high temperatures, the shear tests for the binders of B1, B2, ZW-B1 and ZW-B2 at high temperatures were done by means of a dynamic shear rheometer (DSR). The DSR tests were performed in the light of the AASHTO T315-2 under the following test conditions: Mode of loading was controlled-strain (12%); temperatures were 46, 52, 58, 64, 70, 76, 82 and 88 °C, respectively, and test frequency was 1.59 Hz.

The test results of complex modulus G^* and phase angle δ are shown in Table 5. In order to observe the change trend intuitively, the curves are drawn in Figs. 4 and 5, and the results of rut factor $G^*/\sin\delta$ are shown in Fig. 6.

The complex modulus G^* can be considered as the total resistance of the binder to deformation when repeatedly sheared. The higher the G^* value, the stiffer and thus the more resistant to rutting the asphalt binder will be. But the G^* alone is not sufficient to characterize asphalt binders, and phase angle (δ) is also needed. For rutting resistance, a high complex modulus G^* value and low phase angle δ are both desirable [34].

From Table 5 or Figs. 4 and 5, it can be seen that the complex modulus G^* valves of four kinds asphalt binders decline nonlinearly with the increase of temperature, especially in the lower temperature range the decline rate is larger, which is the general character of asphalt binder. However, the changing trends of phase angles are different from those of complex modulus and are related to the types of binders. Meanwhile, it is clear to see that the complex modulus values of original asphalt binders are improved and the phase angles reduced dramatically by the FR and WMAA.

It can be seen from Fig. 6 that the changing trends of rut factor $G^*/\sin\delta$ are similar to those of the complex modulus G^*. The high temperature performances of asphalt binders are improved greatly by the used FR and WMAA; the degrees that the two additives improve at 64 °C are 112% for B1 and 62% for B2, respectively.

3.2. Results and analysis of BMMFW

3.2.1. High-temperature performance

The wheel tracking test is a basic test method for evaluating performance of bituminous mixture in high temperatures. The dynamic stability (DS) of bituminous mixture obtained by wheel tracking test can reflect the ability of resistance to rutting in high temperatures. The higher the DS value of bituminous mixture,

Table 5
Results of complex modulus G^* and phase angle δ at different temperatures.

Binder species	46 °C		52 °C		58 °C		64 °C	
	G^* (Pa)	δ (°)	G^* (Pa)	δ (°)	G^* (Pa)	δ (°)	G^* (Pa)	δ (°)
B1	33,700	63.45	16,230	63.04	8189	63.67	4300	65.13
ZW-B1	63,030	62.55	31,050	61.11	16,230	59.85	8675	59.72
B2	43,170	65.76	20,810	67.14	10,440	68.45	5642	69.73
ZW-B2	80,140	65.19	37,750	66.28	18,610	67.26	9273	67.96
	70 °C		76 °C		82 °C		88 °C	
B1	2381	66.67	1361	67.57	826.2	66.76	–	–
ZW-B1	4763	60.69	2702	62.13	1582	63.24	952.3	63.43
B2	3165	70.84	1881	71.43	1162	71.43	760.2	70.84
ZW-B2	4784	68.53	2589	68.65	1446	68.25	869.1	66.86

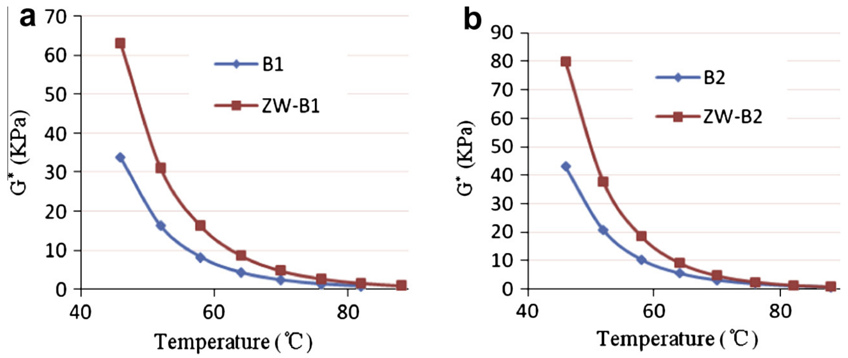

Fig. 4. Changing of complex modulus with temperatures.

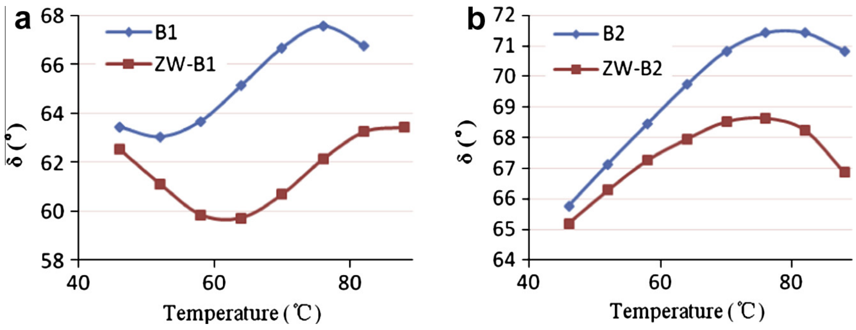

Fig. 5. Relationship between phase angle and temperature.

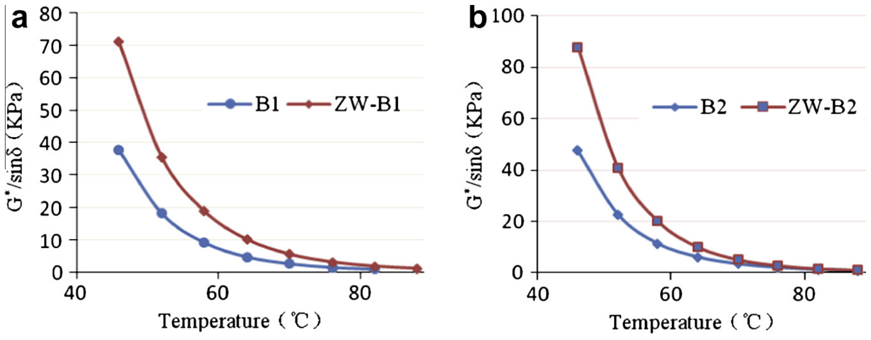

Fig. 6. Changing of $G^*/\sin\delta$ with temperatures.

the better the performance of resistance to permanent deformation in high temperatures will be. The test values of DS of three parallel specimens of BMMFW are 7258, 6738 and 7425 times/mm (the average value is 7140 times/mm), respectively, and the average value is higher than that (5935 times/mm) of base bituminous mixture by 20.3%.

3.2.2. Moisture stability performance

There are two indicators for evaluating moisture stability performance. One is the retained Marshall stability (MS) obtained from immersion Marshall Test, and the other is the indirect tensile strength ratio (TSR) obtained by freeze–thaw indirect tensile test. The test value of MS of the BMMFW is 88.4% (larger than the standard requirement of 85%), which is approximately 1.9% lower than that (90.1%) of mixtures of original asphalt binder. Meanwhile the test value of TSR of the BMMFW is 86.6% (larger than the standard requirement of 80%), which is approximately 2.7% lower than that (89.0%) of mixtures of original asphalt binder. Therefore the moisture stability performances of the bituminous mixtures can be slightly reduced by the FR and WMAA, but which still can meet the requirements of specifications.

3.2.3. Low-temperature bending performance

Low-temperature bending test is a common method used for evaluating bituminous mixture's performance of resistance to cracking in low temperatures. The parameters of flexural–tensile strength, strain and stiffness modulus can be obtained by low-temperature bending test and the flexural–tensile strain is the most concerned parameter among them. Generally speaking, the higher the flexural–tensile strain, the better the performance of resistance to cracking at low temperatures is. The test values of flexural–tensile strain of four parallel specimens of the BMMFW are 2681, 2739, 2870, 2873 and 2847 με, respectively, and the average value of them is 2847 με (it satisfies the criterion in Chinese specifications of no less than 2500 με), which decreases 12.3% compared to that (3195 με) of base bituminous mixture.

3.2.4. Low-temperature bending creep performance

The low-temperature bend test can only provide limited mechanics information. In order to investigate deeply the effect of the FR and WMAA on low-temperature performance, the low-temperature bending creep tests for three mixtures including original asphalt binder mixture, 3% WMAA mixture, and 4% FR together with 3% WMAA mixture were conducted.

3.2.4.1. Analysis of bending creep deformation. Bituminous mixture is a kind of viscoelastic–plastic material and has a typical creep deformation feature under a constant force. The creep deformation can be divided into three stages [35,36], which is shown in Fig. 7. The primary creep and secondary creep stages are important for analysis, which are usually described by Burgers model. Burgers model consists of a Maxwell model and a Kelven model in series, which is shown in Fig. 8.

The creep equation of Burgers model is as follow.

$$\varepsilon(t) = \sigma_0\left[\frac{1}{E_1} + \frac{t}{\eta_1} + \frac{1}{E_2}\left(1 - e^{-\frac{E_2}{\eta_2}t}\right)\right]$$

And the creep compliance equation can be written below.

$$J(t) = \frac{1}{E_1} + \frac{t}{\eta_1} + \frac{1}{E_2}\left(1 - e^{-\frac{E_2}{\eta_2}t}\right) \tag{5}$$

where E_1 and η_1 are instantaneous elastic modulus (MPa) and viscous coefficient (MPa s) of the Maxwell's model, respectively; E_2 and η_2 are elastic modulus (MPa) and viscous coefficient (MPa s) of the Kelven model, respectively.

3.2.4.2. Comparison of parameters of low-temperature performance.

3.2.4.2.1. Relaxation time. Relaxation time ($\tau_1 = \eta_1/E_1$) is an important material parameter. The longer relaxation time leads to a lower rate of stress relaxation, which goes against the rapid dissipation of stress [37]. Therefore, a short relaxation time means better low-temperature performance.

3.2.4.2.2. Creep rate. The rheological curve at low-temperature of asphalt binders is analyzed in Superpave system according to the situation shown in Fig. 9 [34].

In Fig. 9 the m-value is the slope of log creep stiffness versus log time curve when t is 60 s, which reflects the rate of change of creep deformation. Superpave system thinks that the larger m-value means a better low-temperature performance. For asphalt binder the m-value can be output automatically from the test system. By means of the evaluation idea the m-value is also applied in analysis of low-temperature performance of bituminous mixtures, but it should be mentioned that the m is actually a function of time t and should be written as $m(t)$. The $m(t)$ cannot be obtained directly in the creep test, but can be calculated by using the Eq. (8) below.

3.2.4.2.3. Ratio of $m(t)$ to $S(t)$. Usually, the low-temperature performances of binder materials can be compared to each other only by m-values, but it is difficult to evaluate the low-temperature performances for such binders with higher m-values and larger stiffness modulus. Therefore, based on the characteristics of the bending creep deformation, Liu et al. [35] established a physical Eq. (6) and proposed a new method which combined $m(t)$-value and

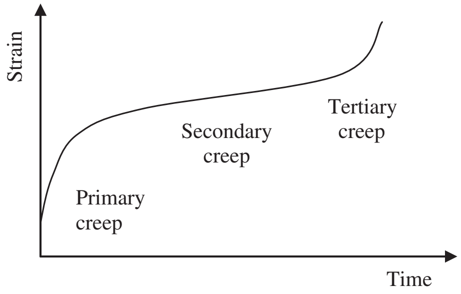

Fig. 7. Schematic diagram of creep deformation.

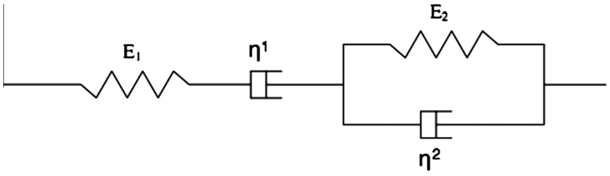

Fig. 8. Burgers model.

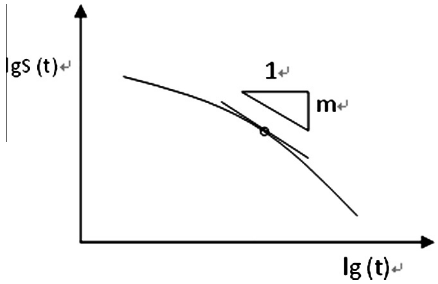

Fig. 9. Bending creep curve in double logarithmic coordinates.

$S(t)$-value synthetically and considered the binder with larger ratio of $m(t)$ to $S(t)$ as material of good low-temperature property.

$$\frac{m(t)}{S(t)}\times\frac{1}{t}=\frac{1}{\eta_1}+\frac{1}{\eta_2}e^{-\frac{E_2}{\eta_2}t} \tag{6}$$

Or

$$\frac{m(t)}{S(t)}\times\frac{1}{t}=J'(t) \tag{7}$$

$$m(t)=t\times J'(t)/J(t) \tag{8}$$

The right sides of Eqs. (6) and (7) have clear physical and geometric meanings, which is the changing rate of creep compliance at a certain time or the slope of compliance curve. The larger $J'(t)$ means the better deformation ability and lower temperature performance. While the larger $m(t)$ and smaller $S(t)$ means that the material has a good low-temperature performance, therefore when the ratio of $m(t)/S(t)$ is large at time t, the low-temperature performance of material is certainly good. Therefore, the Eq. (6) is not only the result of mathematical derivation, but also has a real physical basis. It is more reasonable to compare the different materials' low-temperature performances using the ratio of $m(t)/S(t)$ at the same time t.

3.2.4.2.4. Dissipated energy ratio. Based on the four parameters of Burgers model, the following Eqs. (9) and (10) used for calculation of dissipated energy and storage energy were obtained [35,38]. Dissipated energy ratio is the ratio of dissipated energy to storage energy (i.e. $W_d(t)/W_s(t)$), and it can reflect the low-temperature performances of materials. The larger dissipated energy ratio means the better low-temperature performance.

$$W_d(t)=\sigma_0^2\left[\frac{t}{\eta_1}+\frac{1}{2E_2}\left(1-e^{-\frac{2E_2}{\eta_2}\cdot t}\right)\right]$$

$$W_s(t)=\sigma_0^2\left[\frac{1}{E_1}+\frac{1}{2E_2}\left(1-2e^{-\frac{E_2}{\eta_2}\cdot t}+e^{-\frac{2E_2}{\eta_2}\cdot t}\right)\right] \tag{9}$$

3.2.4.3. Analysis of low-temperature bending creep performance. Based on the deformation data from bending creep tests, four parameters of Burgers model of 10 specimens were obtained using the nonlinear regression analysis and the results are shown in Table 6. In Table 6, Nos. 1-2 is base bituminous mixture, Nos. 3-6 is WMA with 3% of WMAA, and Nos. 7-10 is BMMFW with 4% of FR and 3% of WMAA.

The average values of each parameter are shown in Table 7.

It can be seen from Table 7 that, four Burgers model parameters of the WMA and BMMFW are all increased in different degrees compared with those of the base bituminous mixture. The parameters including the relaxation time τ_1, $m(t)$, $m(t)/S(t)$ and dissipated energy ratio which can all reflect the low temperature performance are calculated at when t is 3600 s (Specimens started to failure after 2 h. We used this moment from steady stage 2 in calculation), and the results are shown in Table 8.In accordance with the order of base bituminous mixture, WMA and BMMFW, it can be seen from Table 8 that τ_1 increases gradually while $m(t)/S(t)$ and $W_d(t)/W_s(t)$ decrease obviously. The trends of the three parameters indicate that WMAA can reduce the low temperature performance of bituminous mixture, while the FR has the same effect on WMA as the WMAA on base bituminous mixture. The combined adverse effects of the two additives on low temperature performance of base bituminous mixture are dramatically and the declined extents in the light of τ_1, $m(t)/S(t)$ and $W_d(t)/W_s(t)$ are 69.8%, 53.1% and 15.0%, respectively. It should be noted here that although the three parameters of the low temperature performance indicate the consistent changing trend, the change extents or the rates are significantly different (the values of $m(t)/S(t)$ and $W_d(t)/W_s(t)$ are related to t). This is an interesting fact, which needs to be studied to determine which parameter can exactly reflect the reduction magnitude of low temperature performance.

Table 6
Parameters of Burgers model.

No.	E_1 (MPa)	η_1 (MPa s)	E_2 (MPa)	η_2 (MPa s)
1	6.176	6794.144	1.159	416.600
2	6.556	7304.923	1.250	556.661
3	7.885	13238.126	2.895	1987.955
4	7.642	11555.011	2.513	1278.154
5	7.690	13029.996	2.525	1259.158
6	8.534	12870.445	3.137	1868.908
7	8.174	14918.803	2.812	2532.929
8	8.648	16170.831	2.857	2006.694
9	9.368	18104.931	2.646	2437.320
10	8.531	16136.160	2.650	1797.715

Table 7
Average values of Burgers model parameters.

Material type	E_1 (MPa)	η_1 (MPa s)	E_2 (MPa)	η_2 (MPa s)
Base bituminous mixture (Of B1)	6.4	7049.5	1.2	486.6
WMA (Of Sasobit® and B1)	7.9	12673.4	2.8	1598.5
BMMFW (Of ZW-B1)	8.7	16332.7	2.7	2193.7

Table 8
Indicators of low-temperature performance based on Burgers model parameters.

Material type	τ_1/s	$m(t)$	$m(t)/S(t)$	$W_d(t)/W_s(t)$
Base bituminous mixture (Of B1)	1107	0.342	0.513	1.618
WMA (Of Sasobit®and B1)	1598	0.374	0.290	1.521
BMMFW (Of ZW-B1)	1880	0.345	0.241	1.376

However, according to the m-value, the low temperature performance of WMA is better than that of the base bituminous mixture, while the low temperature performance of BMMFW is not less than or very close to that of the base bituminous mixture, which indicate the WMAA can improve low temperature performance and the combined effects of the FR and WMAA does not lead to the degradation in low temperature performance. This is contradicted with the trend reflected by the other three indicators and the conclusion obtained from m-value in this research is incredible. In terms of experimental data of low-temperature bending creep tests in the paper, the conclusions drawn from the stress relaxation time, $m(t)/S(t)$ and dissipated energy ratio are preferable.

4. Conclusions

On the basis of large number of the laboratory tests, the combined effects of the FR (ZIR®) and WMAA (Sasobit®) on pavement performances of asphalt binders and bituminous mixtures were mainly researched, and the main conclusions and recommendations are summarized as follows.

The results of softening point and low temperature ductility tests show that the FR and WMAA improve significantly high temperature performance of asphalt binders and have adverse effects on low-temperature performance.

The results of viscosity–temperature tests show that the same kind of the WMAA with the same dose can have a different viscosity reduction for different asphalt binders, and the effect of WMAA on temperature susceptibility of asphalt binder is also related to the types of asphalt and it may be positive or negative.

The results of rutting test show that the used FR and WMAA can improve significantly the high temperature performance of asphalt binders, while the moisture stability tests indicate that the two additives have a slight negative impact on moisture stability. Considering the road tunnel does not usually undergo the influence of water, the slight decreasing in moisture stability may be neglected.

The results of low-temperature bending test also show that the two additives have an adverse effect on low temperature property of binder.The results of low temperature bending creep tests of three kinds of bituminous mixtures show profoundly that the FR and WMAA increase the stress relaxation time and decrease the $m(t)/S(t)$ and dissipated energy ratio, which means that the two agents can deteriorate the low temperature performance of base bituminous mixture. And the low temperature elasticity and viscosity of bituminous mixture are increased by the two additives.

Two interesting and important phenomenon were found in this paper: (1) Using the m-value alone to describe low temperature performance of asphalt binder or bituminous mixture may sometimes be insufficient, while using $m(t)/S(t)$, relaxation time and dissipated energy ratio can be more reasonable. (2) In this study, three parameters of relaxation time, $m(t)/S(t)$ and dissipated energy ratio can give the consistent reduction trends of low temperature performance, but the reduction extents of them are significantly different. And then further study will be needed to determine which parameter can actually reflect the reduction magnitude of low temperature performance.

In short, the used FR and WMAA can improve high-temperature performance of asphalt binders and bituminous mixtures, decrease low temperature performance and have slightly negative impacts on moisture stability of bituminous mixtures.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.conbuildmat.2013.12.058.

References

[1] Qin Xiantao, Zhu Siyue, Chen Shuanfa, Deng Kunyao. The mechanism of flame and smoke retardancy of asphalt mortar containing composite flame retardant material. Constr Build Mater 2013;41:852–6.

[2] Wen-sheng YU, Chan LI. Performance and application of antiflaming and warm-mix bituminous mixture. Highway 2010;1:129–34.

[3] Cong Peiliang, Jianying Yu, Shaopeng Wu, Luo Xiaofeng. Laboratory investigation of the properties of asphalt and its mixtures modified with flame retardant. Constr Build Mater 2008;22:1037–42.

[4] Wu Shaopeng, Cong Peiliang, Yu Jianying, Luo Xiaofeng, Mo Liantong. Experimental investigation of related properties of asphalt binders containing various flame retardants. Fuel 2006;85:1298–304.

[5] Xu Tao, Huang Xiaoming, Zhao Yongli. Investigation into the properties of Bituminous mixtures containing magnesium hydroxide flame retardant. Fire Saf J 2011;46:330–4.

[6] Wu Shaopeng, Mo Liantong, Cong Peiliang, Yu Jianying, Luo Xiaofeng. Flammability and rheological behavior of mixed flame retardant modified asphalt binders. Fuel 2008;87:120–4.

[7] Capitão SD, Picado-Santos LG, Martinho F. Pavement engineering materials: review on the use of warm-mix asphalt. Constr Build Mater 2012;36:1016–24.

[8] Carmen Rubio M, Martínez Germán, Baena Luis, Moreno Fernando. Warm mix asphalt: an overview. J Cleaner Prod 2012;24:76–84.

[9] EAPA. The use of warm mix asphalt – EAPA position paper. Brussels: European Asphalt Pavement Association; 2010.

[10] Zaumanis M. Warm mix asphalt investigation. Master of science thesis. Kgs. Lyngby: Technical University of Denmark in cooperation with the Danish Road Institute, Department of Civil Engineering; 2010.

[11] Prowell B, Hurley G, Frank B. Warm-mix asphalt: best practices. Lanham (MD): NAPA – national asphalt pavement association; 2011.

[12] Jamshidi Ali, Hamzah Meor Othman, You Zhanping. Performance of warm mix asphalt containing Sasobit: State-of-the-art. Constr Build Mater 2013;38:530–53.

[13] Wasiuddin NM, Saha R, King W Jr. Effects of a wax-based warm mix additive on lower Compaction Temperatures. Advances in geotechnical engineering proceedings of the geo-frontiers. USA: ASCE; 2011. p. 4614–23.

[14] Xiao F, Amirkhanian SN, Zhang R. Influence of short-term aging on rheological characteristics of non-foaming WMA binders. J Performan Constr Facilities; 2011. <http://dx.doi.org/10.1061/(ASCE)CF.1943-5509.0000223>.

[15] Xiao F, Punith VS, Amirkhanian SN. Effects of non-foaming WMA additives on asphalt binders at high performance temperatures. Fuel 2012;94:144–55.

[16] Biro S, Gandhi T, Amirkhanian SN. Midrange temperature rheological properties of warm asphalt binders. J Mater Civ Eng 2009;21(7):316–23.

[17] Edwards Y, Tasdemir Y, Isacsson U. Rheological effects of commercial waxes and polyphosphoric acid in bitumen 160/220-high and medium temperature performance. Constr Build Mater 2007;21(10):1899–908.

[18] Cao D, Ji J. Evaluation of long-term aged properties of Sasobit® modified asphalt. Int J Pavement Res Technol 2011;4(6):384–91.

[19] Jamshidi A, Hamzah MO, Aman MY. Effects of Sasobit content on the rheological characteristics of unaged and aged asphalt binders at high and intermediate temperatures. Mater Res 2012;15(4):1–11.

[20] Liu J, Saboundjian S, Li P, Connor B, Brunette B. Laboratory evaluation of Sasobit®-modified warm-mix asphalt for Alaskan conditions. J Mater Civ Eng 2011;23(11):1498–505.

[21] Biro S, Gandhi T, Amirkhanian SN. Determination of zero shear viscosity of warm asphalt binders. Constr Build Mater 2009;23(5):2080–6.

[22] Buss AF. Investigation of warm-mix asphalt using Iowa aggregates. USA: Iowa State University: Institute for Transportation Iowa State University Iowa; 2011.

[23] Hossain Z, Bhudhala A, Zaman M, O'Rear E, Cross S, Lewis S. Evaluation of the use of warm mix asphalt as a viable paving material in the United States. USA: Federal Highway Administration Turner-Fairbank Highway Research Center; 2009.

[24] Akisetty Chandra, Xiao Feipeng, Gandhi Tejash, Amirkhanian Serji. Estimating correlations between rheological and engineering properties of rubberized asphalt concrete mixtures containing warm mix asphalt additive. Constr Build Mater 2011;25:950–6.

[25] Akisetty Chandra K, Lee Soon-Jae, Amirkhanian Serji N. High temperature properties of rubberized binders containing warm asphalt additives. Constr Build Mater 2009;23:565–73.

[26] Wang Hainian, Dang Zhengxia, You Zhanping, Cao Dongwei. Effect of warm mixture asphalt (WMA) additives on high failure temperature properties for crumb rubber modified (CRM) binders. Constr Build Mater 2012;35:281–8.

[27] Lee Soon-Jae, Amirkhanian Serji N, Park Nam-Won, Kim Kwang W. Characterization of warm mix asphalt binders containing artificially long-term aged binders. Constr Build Mater 2009;23:2371–9.

[28] Zhao Sheng, Huang Baoshan, Shu Xiang, Woods Mark. Comparative evaluation of warm mix asphalt containing high percentages of reclaimed asphalt pavement. Constr Build Mater 2013;44:92–100.

[29] The Ministry of Communications of the People's Republic of China. Standard specification for construction and acceptance of highway asphalt pavement JTG F40–2004. Beijing: China Communication Press; 2004.

[30] National Energy Board. Measure of oxygen index burning property by oxygen index method (NB/NH/T 0815–2010)[S]. Beijing: China Petrochemical Press; 2010.

[31] The Ministry of Communications of the People's Republic of China. Standard test methods of bitumen and bituminous mixtures for highway engineering. JTG E20–2011. Beijing: China Communication Press; 2011.

[32] ASTM D6648–1. Standard test method for determining the flexural creep stiffness of asphalt binder using the Bending Beam Rheometer (BBR); 2008.

[33] Shen Jinan. Pavement performance of bitumen and bituminous mixtures. 1st ed. Beijing: China Communications Press; 2001.

[34] Roberts Freddy L, Kandhal Prithvi S, Brown E Ray. Hot mix asphalt materials, mixture design and construction. 2nd ed. NAPA research and education foundation; 1996.

[35] Liu Shutang, Cao Weidong, Shang Shujie. Analysis and application of relationships between low-temperature rheological performance parameters of asphalt binders. Constr Build Mater 2010;24:471–8.

[36] Zhang Xiaoning. The principles and applications of viscoelastic mechanics in asphalt and Bituminous mixture [M]. Beijing: People's Communications Press; 2006.

[37] Dazhi Guo. Mechanics of layered viscoelastic system. 1st ed. Harbin: Harbin Industrial University Press; 2001. p. 6–23 [original in Chinese].

[38] Johansson Lars S, Isacsson Ulf. Effect of filler on low temperature physical hardening of bitumen[J]. Constr Building Mater 1998;12:463–70.