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Effect of Various Metal Hydroxide Flame Retardants on the Rheological Properties of Asphalt Binder

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Metal hydroxide has been widely used as flame retardant to reduce the hazards of tunnel fire, however, few researches investigate its effect on the rheological properties of asphalt binder systemically. This study explores and compares the effect and mechanisms of magnesium hydroxide (MH), aluminium hydroxide (ATH), hydrated lime (HL), and layer double hydroxides (LDHs) on the rutting resistance, anti-aging resistance, as well as the thermal cracking resistance of asphalt binder. Rotational viscosity (RV) test, dynamic shear rheometer (DSR) test, and bending beam rheometer (BBR) test are involved in the project. Test results indicate: (1) the addition of metal hydroxide generally improves the rutting resistance of asphalt binder during high temperatures due to the typical filler effect, while weakens the resistance to thermal cracking of binder at low temperatures because of the stress concentration; (2) HL and LDHs enhance the anti-aging resistance of asphalt binder; (3) LDHs modified binder, which is proved with better rheological properties, including great rutting resistance, anti-aging resistance and passable resistance to thermal cracking, is recommended for further use. However, the high procurement price is still a big obstacle for its wider application.

Keywords: asphalt binder, metal hydroxide, rheological properties, superpave.

1. INTRODUCTION

Asphalt concrete (AC) has been widely used for tunnel pavement nowadays due to its high driving comfort, low noise as well as the low construction cost [1]. However, the flammability of asphalt binder, an essential component of AC, becomes a tremendous obstacle to its application. As a viscoelastic material, asphalt binder melts and flows under a relatively low temperature, accelerating the spread of fire in combustion [2]. Also, asphalt binder decomposes in ignition, producing heat as well as toxic fumes, which negatively affects the rescue operation in fire [3, 4]. To improve the flame resistance of asphalt binder, flame retardant is applied to asphalt binder for modification.

Halogen based flame retardant modified binder shows effective flame resistance in previous researches [5, 6], however, it produces toxic fumes during ignition and probably causes environmental issues [7]. With the consideration of environment protection and safety issues, metal hydroxides, i.e. magnesium hydroxide (MH), aluminium hydroxide (ATH), hydrated lime (HL), and layer double hydroxides (LDHs), which have been proved with fantastic flame resistance as well as environmental friendly, gradually replace halogen flame retardant as more popular flame retardant in pavement design [8]. The thermal decomposition of these metal hydroxides during high temperatures play an important role in flame retardancy and smoke suppression for asphalt binder, and also be non-toxic. It is proved by differential scanning calorimetry (DSC) and thermogravimetry (TG) test that the flame retardancy of metal hydroxide modified binder is realized by the similar reaction temperatures of metal hydroxide and the chemical components of asphalt binder [9, 10]. Not only the decomposition of metal hydroxide is able to absorb the majority of heat and lower the temperature of asphalt mixture during combustion, but also the products from metal hydroxide decomposition, e.g. magnesium oxide, aluminium oxide, and calcium carbonate, can form a film to prevent the contact between oxygen and asphalt binder, effectively retarding the combustion [11]. Moreover, HL is capable of reducing the activity of some oxidation catalysts when performing as flame retardant, slowing the ignition rate and reducing the soot from the ignition of aromatics and resins [12].

To investigate the comprehensive effect of metal hydroxide, in addition to the flame characteristics of metal hydroxide modified binder, the pavement performance of modified binder should also be studied. From existing research, consistency tests show that the penetration of asphalt binder decreases with the addition of single metal hydroxide (MH, ATH) when metal hydroxide dosage is relatively high, while the softening point increases, and the ductility significantly decreases, suggesting the stiffening effect of metal hydroxide on asphalt binder [5, 13]. Moreover, from dynamic shear rheometer (DSR) test, it is observed that the addition of metal hydroxide (MH, ATH,

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HL) is able to improve the rutting parameter $G^*/\sin\delta$ at high temperatures due to the increased complex modulus and the decreased phase angle, which reveals the enhanced rutting resistance of metal hydroxide modified binder during high temperatures [2, 6, 14 – 16]. All of the above researches confirm the general hardening effect of single metal hydroxide on asphalt binder during high temperatures, but the effect of single metal hydroxide on the low temperature performance of asphalt binder still requires further study. Different from single metal hydroxide, LDHs are multi-nestification layered structure consisting of positively charged laminates and interlayer anions [17, 18]. Although LDHs modified binder shows similar consistency test results and high temperature performance to other unaged metal hydroxide modified binder [17 – 19], Pang et al. found that LDHs modified binder showed higher stiffness than base binder before aging, but lower stiffness after aging by shear creep test [19], which suggests that the inclusion of LDHs hardens unaged binder at low temperatures, weakening the binder resistance to thermal cracking, while provides a fantastic anti-aging resistance beneficial to the low temperature performance of binder after aging. Owing to the stronger alkalinity of HL and the well-established multi-layered structures of LDHs [12, 15, 17 – 18], different single metal hydroxides and LDHs are supposed to show discrepancy in the effect on the rheological properties of binder, especially after aging, which lacks particular investigation in previous researches.

The main objective of this study is to evaluate and compare the rheological properties of various metal hydroxide flame retardant modified binder systemically by Superpave tests. Not only the high temperature performance (resistance to rutting), low temperature performance (resistance to thermal cracking) and anti-aging resistance of MH, ATH, HL and LDHs modified binders are investigated, but the different mechanisms that how different metal hydroxides affect the rheological performance of binder are analysed as well. The grading system used in the paper is Superpave performance grading (PG), which involves with both climate conditions and aging stages of asphalt binder.

2. METHODOLOGY

2.1. Materials

The PG grade of base binder used in this research is PG 64-22, provided by Jiangsu Baoli International Investment Co., Ltd, China. Table 1 shows the basic properties of the base binder.

MH, ATH, HL, and LDHs are applied to base binder as flame retardant. ATH and HL are provided by Tianjin Bodi Chemical Industrial Co., Ltd., China, while MH is provided by aladdin, and LDHs are provided by Jingjiang Concord Plastics Technology Co., Ltd., China. All of these metal hydroxides are with high purity of 95 %. Other basic physical properties of these metal hydroxides are shown in Table 2.

Table 1. Basic properties of base binder

Penetration, d, mm	65.8
Softening point, °C	47.8
Viscosity at 60 °C, Pa·s	193
PEN Grade	60/70
AC Grade	AC 20

Table 2. Basic properties of metal hydroxide

	Molecular formula	Specific gravity, g/cm^3	Specific surface area, m^2/g	Median particle size, µm
MH	$Mg(OH)_2$	2.36	0.78	21.3
ATH	$Al(OH)_3$	2.42	0.83	13.5
HL	$Ca(OH)_2$	2.24	1.04	7.4
LDHs	$Mg_6Al_2CO_3(OH)_{16}\cdot 4H_2O$	2.10	3.25	–

To further study the different mechanisms of how these four metal hydroxides affect the rheological properties of asphalt binder, an SU8010 scanning electron microscope (SEM) supplied by Hitachi Company, Japan, is used to observe the particle of these four metal hydroxides.

In the experiment, there are five groups established, base binder, MH modified binder, ATH modified binder, HL modified binder, and LDHs modified binder. The dosage of all the metal hydroxide flame retardant is controlled as 25 wt.%, the usual used value to ensure flame retardancy based on existing researches [7, 8, 10].

The blending procedures of base binder and various metal hydroxides are performed as follows. Loading asphalt binder into a stainless steel can and heating it up to 160 ± 5 °C by an electric furnace. Then, stirring the asphalt at a rate of 1000 rpm by mixing machine for 15 minutes, while adding flame retardant to the asphalt and mixing the mixture in the initial 5 minutes. During this process, keeping the temperature at the range of 160 ± 5 °C. Then, with the same temperature, stirring the mixture by shear machine at a rate of 5000 rpm for 30 minutes to ensure the uniform distribution of metal hydroxide.

2.2. Testing methods

After mixing, three Superpave tests are involved in this study to evaluate the rheological performance of asphalt binder, rotational viscosity (RV) test, dynamic shear rheometer(DSR) test and bending beam rheometer(BBR) test. Rolling thin film oven (RTFO) test and pressure aging vessel (PAV) test are aging tests used to simulate short-term aging and long-term aging process of asphalt binder.

In accordance with ASTM D4402, RV test is used to measure the rotational viscosity of asphalt binder at a specific temperature to evaluate the workability of binder at mixing and compaction temperatures. The test is performed using the NDJ-1F rotational viscometer provided by Shanghai Changji Geological Instrument Co., Ltd., China.

DSR test is based on ASTM D7175 to evaluate the rutting resistance of asphalt binder at high temperatures to represent the high temperature performance of binder, using the MCR102 DSR test system from Anton Paar GmbH, Austria, while BBR test is performed according to

ASTM D6648 to evaluate the resistance to thermal cracking of asphalt binder at low temperatures using the TE-BBR test system provided by CANNON Instrument Company, US. RTFO test is applied to asphalt binder to simulate mixing and compaction process to obtain short-term aged binder based on ASTM D2872, using the CS325-50 RTFO from James Cox & Sons Inc., US, while PAV test that simulates 7 – 10 years long-term aging process of binder is performed in accordance with ASTM D6521 using the PR9500 PAV test system supplied by PRENTEX Alloy Fabricators, Inc., US.

3. RESULTS

3.1. Rotational viscosity test

The rotational viscosity of binder from all the groups at 135 °C, the common temperature for asphalt binder mixing, is shown in Fig. 1 as follows. The mass of specimen used in test is around 11 g, and the resisting torque in test should be within the range of 10 % – 98 % of the instrument capacity to ensure the accuracy of test result. As Fig. 1 shows, all the modified binders have higher viscosity than base binder, but still satisfy the requirement for binder workability according to ASTM D4402 that the viscosity at 135 °C should be less than 3 Pa·s. The viscosity of HL modified binder and LDHs modified binder is relatively higher than that of other modified binders, suggesting the poorer fluidity at high temperatures of these two binders.

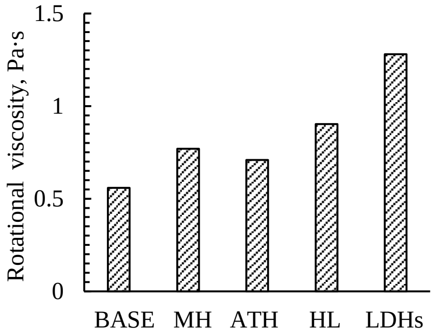


Fig. 1. Rotational viscosity of base binder and metal hydroxide binder at 135 °C

3.2. Dynamic shear rheometer test

Since most of the permanent deformation of asphalt binder occurs at early stages of pavement, Superpave specification requires to perform DSR test on virgin and RTFO binder at high temperatures to evaluate the rutting resistance of binder.

Loading frequency is set as 10 rad/s in DSR test based on ASTM D7175. As shown in Fig. 2, the rutting parameter $G^*/\sin\delta$ of all the metal hydroxide modified binders before aging is observed higher than that of base binder at the temperature range of 40 – 80 °C, while ATH and HL modified binders show similar curves of $G^*/\sin\delta$ which are slightly higher and steeper than that of MH modified binder but lower and gentler than that of LDHs modified binder. It indicates that the rutting resistance at high temperatures of asphalt binder is improved with the addition of metal hydroxide, while the effects of LDHs are the most significant. Also, it can be noticed that the beneficial influence of metal hydroxide on the rutting resistance is particularly considerable at relatively low temperatures, but the increase in $G^*/\sin\delta$ at higher temperature, e.g. 80 °C, is not so noticeable.

The test result can be explained by the different mechanisms of these four metal hydroxides influencing the high temperature performance of asphalt binder. As solid particles, all of these metal hydroxides are able to destroy the bond between asphalt molecules and increase the internal friction of asphalt binder, restricting the flow of binder at high temperatures [2]. As a result, the rutting potential is reduced. Owing to the smaller particle size, ATH, HL, as well as LDHs modified binders have higher viscosity during 40 – 80 °C [2, 15], leading to a better rutting resistance comparing with MH modified binder. Apart from the typical filler effect, HL is also able to react chemically with the acidic components of asphalt binder to form insoluble calcium salts due to its stronger alkalinity and higher porosity [12, 14 – 16, 20]. Since the reactive components of asphalt binder, carboxylic acids and 2-quinolone types, belong to reactive pre-asphaltens or asphaltens that primarily provide rutting resistance at high temperatures [12], the decrease of these components increases the rutting risk of asphalt binder. Consequently, although the particle size of HL is slightly smaller than ATH, the rutting resistance of ATH modified binder and HL modified binder is observed similar.

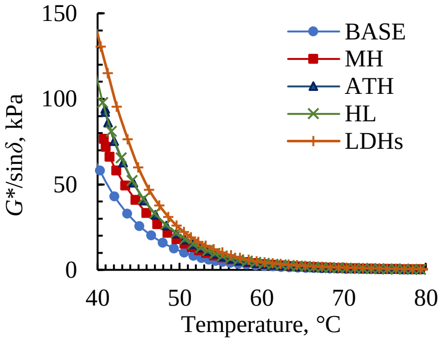
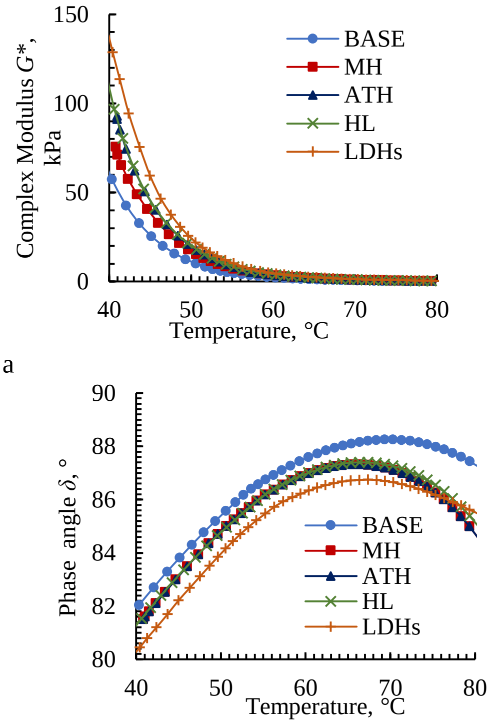


Fig. 2. Rutting parameter curves of base binder and metal hydroxide modified binders before aging

Fig. 3 shows the complex modulus and phase angle related to temperatures of base binder and metal hydroxide modified binders before aging. It can be noticed that the addition of metal hydroxide increases the complex modulus of asphalt binder but decreases the phase angle of asphalt binder. The stiffening effect of solid particles contributes to the improved complex modulus of all the metal hydroxide modified binders. Similar to the rutting parameter curves, the complex modulus curves of ATH and HL modified binder are close to each other, but higher than that of MH modified binder and lower than LDHs modified binder, which is due to the particle size effect as well as the irreversible chemical reaction of HL and asphalt binder. Furthermore, the lower phase angle of all the modified binders comparing with base binder within the temperatures of 40-80 °C reveal the more elastic portion of metal hydroxide modified binders, which results from the elasticity provided by the solid particles of metal hydroxide. During high temperatures, the phase angle of HL modified binder is observed slightly higher than ATH and MH modified binder, which is possibly because the reactive components of asphalt binder that HL irreversibly absorbs are mainly responsible for the elastic portion of binder [12]. Additionally, the major contributor to the increase in $G^*/\sin\delta$ of metal hydroxide modified binders is the complex modulus G^* considering the two affecting factors, complex modulus and phase angle. Comparing with base binder, the variation of $\sin\delta$ of metal hydroxide modified binders never exceeds 0.01, while the value of complex modulus changes in a much wider range.



a

b

Fig. 3. Curves of two parameters about rutting resistance of base binder and metal hydroxide modified binders before aging: a – complex modulus; b – phase angle

The curves of $G^*/\sin\delta$ of base binder and modified binders after RTFO aging are shown in Fig. 4. The larger values of $G^*/\sin\delta$ of RTFO binders comparing with virgin binders indicate the improvement of thermal resistance which is owing to the increase of resins and asphaltens [21]. Although all the metal hydroxide modified binders still show larger $G^*/\sin\delta$ comparing with base binder, similar to the virgin situation, the trend of metal hydroxide modified binders varies. LDHs modified binder is observed with the largest value of $G^*/\sin\delta$ among all the modified binders, while MH modified binder shows the smallest value. It reveals the fantastic high temperature performance of LDHs modified binder after RTFO aging, which is not only related to the brilliant rutting resistance of LDHs modified binder, but also associated with its excellent anti-aging resistance [22].

Fig. 5 shows the complex modulus and phase angle related to temperatures of base binder and metal hydroxide modified binders after RTFO aging respectively. The curves of complex modulus are noticed similar to those of $G^*/\sin\delta$ in RTFO situation, but different from the complex modulus curves before aging.

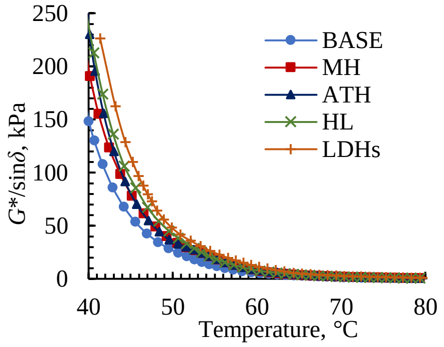
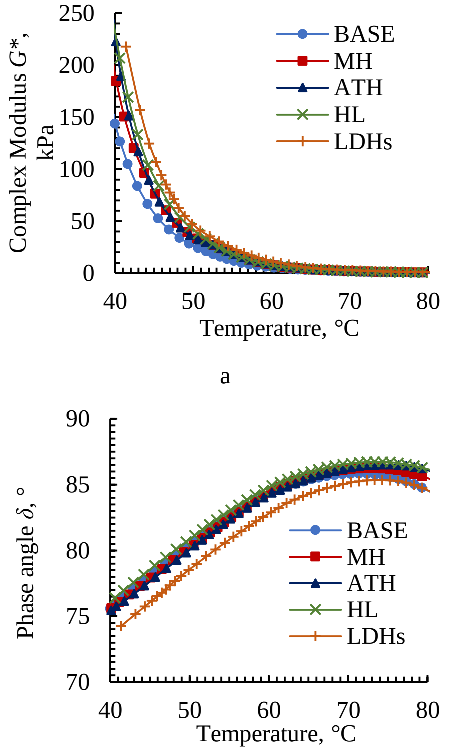


Fig. 4. Rutting parameter curves of base binder and metal hydroxide modified binders after RTFO aging

In addition, the phase angle curves in RTFO situation is also not compatible with those before aging. Instead of that all the metal hydroxide modified binders show smaller phase angle than base binder, the phase angle of HL modified binder is always larger than base binder during 40 – 80 °C, and ATH and MH modified binders also show larger phase angle than base binder at high temperatures. It is the different degrees of aging of various binders that result in the discrepancy. Moreover, similar to the virgin situation, it is the increase of complex modulus that primarily contributes to the increase of $G^*/\sin\delta$ for metal hydroxide modified binder.



a

b

Fig. 5. Curves of two parameters about rutting resistance of base binder and metal hydroxide modified binders after RTFO aging: a – complex modulus; b – phase angle

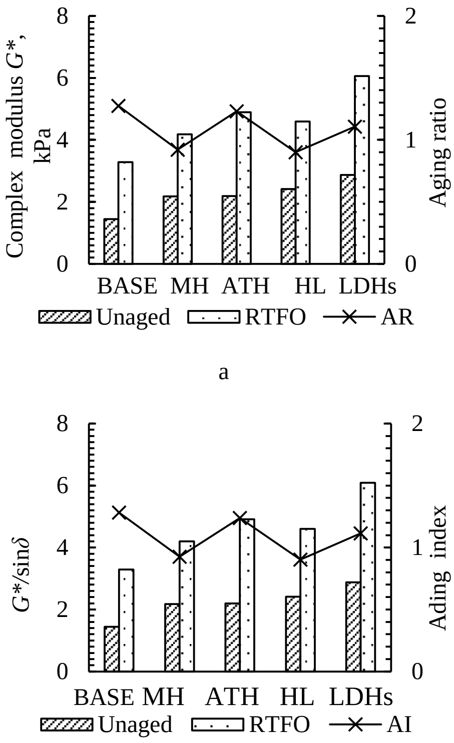
To evaluate the aging magnitude of binder, aging ratio (*AR*) and aging index (*AI*) of different binders are calculated and compared using Eq. 1 and Eq. 2 respectively [14, 16], and both G^* *and* δ can be obtained from DSR test.

$$AR = \frac{|G^*|(\omega,T)_{aged}}{|G^*|(\omega,T)_{unaged}}; \tag{1}$$

$$AI = \frac{(G^*/sin\delta)_{aged}}{(G^*/sin\delta)_{unaged}}, \tag{2}$$

where T refers to testing temperature, and ω refers to loading frequency.

Since the high temperature grade of base binder, 64 °C, is a significant temperature in DSR test to compare G^* and $G^*/\sin\delta$ of base binder and modified binders, it is selected for *AR* and *AI* calculation. The comparison of complex modulus and $G^*/\sin\delta$ at 64 °C before and after RTFO aging for base binder and metal hydroxide modified binders is shown in Fig. 6.

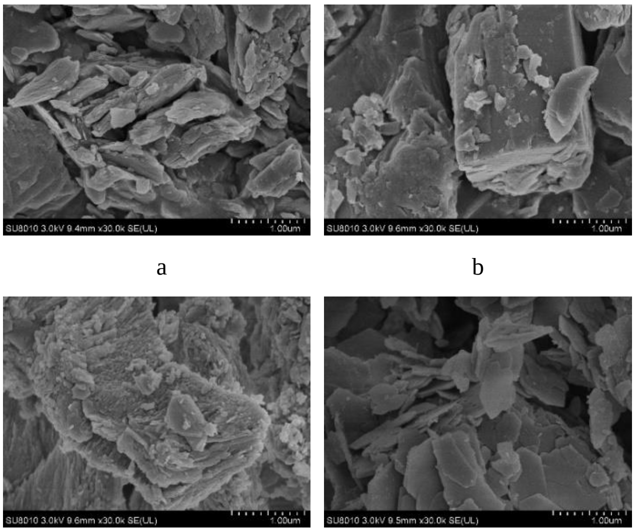


a

b

Fig. 6. Aging ratio and aging index for different binders at 64 °C: a – aging ratio; b – aging index

Both *AI* and *AR* of HL modified binder are observed particularly lower than others, suggesting that the transformation from saturates and aromatics to resins and asphaltens in HL modified binder during RTFO test is much weaker than others. This phenomenon can be explained by the greater anti-aging resistance of HL modified binder. Fig. 7. shows the SEM images of MH, ATH, HL, and LDHs particles, where the HL particles are observed with rougher surface and larger surface area comparing with MH and ATH, and LDHs particles shows evident layered structures. As a metal hydroxide with relatively strong alkalinity and large surface area, HL serves as a powerful chemical base to absorb the acidic reactive components in asphalt, reducing the amount of preasphaltens and asphaltens that are prone to aging. The removal of these components reduces the rate of oxidation of asphalt binder, as well as lowers the viscosity sensitivity caused by oxidation products, thus improving the anti-aging resistance of asphalt binder [12, 20]. The high temperature during RTFO test accelerates and intensifies the neutralization, therefore, the great anti-aging resistance of HL modified binder is more significant as Fig. 6 shows. LDHs are also noticed with lower *AI* and *AR* than base binder. By the well-established multilayered structures depicted in Fig. 7 d, LDHs obstruct the infiltration of oxygen to asphalt chains, thus retarding the aging process of asphalt binder.



a

b

c

d

Fig. 7. SEM image of different metal hydroxide particles: a – MH; b – ATH; c – HL; d – LDHs

3.3. Bending beam rheometer test

According to ASTM D6648, time-temperature superposition should be applied to BBR test for testing time reduction. Time-temperature superposition means that the creep stiffness $S(t)$ and m-value measured at the temperature with 60 second loading time are the same as those at the temperature 10 °C lower with 2 hours loading time, a usual loading time for in-service HMA pavements for cracking resistance measurement. Since the low temperature grade of base binder is –22 °C, BBR test is performed on PAV binder at – 12 °C. Considering the stiffness as well as m-value of asphalt binder, the thermal cracking resistance of binder at low temperatures can be evaluated. The time domain relaxation modulus is also calculated based on Hopkins and Hamming's algorithm [24] to show the low temperature performance of binder, which can be numerically solved by convolution integral

$$\int_0^t E(t)D(t-\tau)d\tau = t, \tag{3}$$

where $E(t)$ is the relaxation modulus, and $D(t)$ refers to the creep compliance, $D(t) = 1/S(t)$.

As Fig. 8 shows, all the metal hydroxide modified binders show higher creep stiffness than base binder. Therefore, from the perspective of stiffness, the presence of metal hydroxide particles hardens asphalt binder, detrimental to the resistance to thermal cracking. The same conclusion can also be drawn from the comparison of relaxation modulus in Fig. 9. Although the decreasing rate of relaxation modulus along time is noticed slightly lower than stiffness, the magnitude relation between the relaxation modulus of various binders are the same as the creep stiffness.

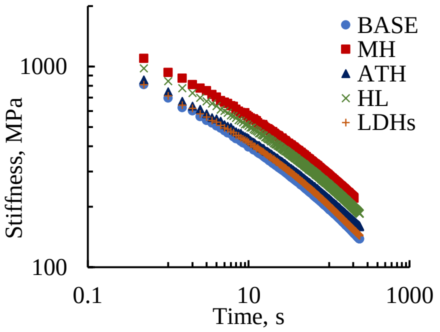


Fig. 8. Stiffness curves of base binder and metal hydroxide modified binders at – 12 °C

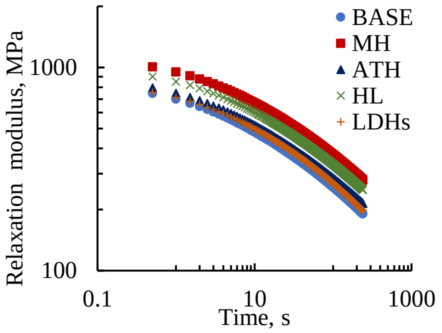


Fig. 9. Relaxation modulus curves of base binder and metal hydroxide modified binders at – 12 °C

Apart from stiffness, m-value, which represents the change rate of stiffness, should also be considered to evaluate the low temperature performance of asphalt. According to Superpave specification, the relationship between the creep stiffness and time in log scale can be expressed as a quadratic function

$$\log[S(t)] = A + B[\log(t)] + C[\log(t)]^2, \tag{4}$$

where *t* represents the loading time, *S(t)* refers to the stiffness corresponding to a specific *t*, and regression coefficients *A*, *B*, *C* are determined using the stiffness values at 8, 15, 30, 60, 120 and 240 second. m-value is defined as the derivative of creep stiffness in log scale at 60 second, that is, $m = \log(t) + 2C\log(t)$, where *t* is equal to 60. Larger m-value corresponds to the more elasticity of asphalt binder. That is, the binder with larger m-value is able to relax better under loading, thus releasing more energy. Fig. 9 displays the stiffness and m-value of base binder and metal hydroxide modified binders at the loading time of 60 second. It is noticed that the stiffness of MH, ATH and HL binders is observed higher than base binder, while their m-values are lower than base binder, revealing the significant adverse effect of MH, ATH and HL on the low temperature performance of asphalt binder. It is not only due to the stress concentration in the defective structure of asphalt binder caused by metal hydroxide particles [2], but also because of the high brittleness of modified binder resulting from the large amount of resins and asphaltens produced during aging [23]. However, different from other modified binders, LDHs modified binder shows similar value of creep stiffness to base binder, and its m-value even exceeds that of base binder, indicating the great rebounding ability of LDHs modified binder. It is likely owing to the fantastic anti-aging resistance of LDHs modified binder, which helps remain more elastic portion of asphalt binder after PAV aging. Therefore, although the addition of metal hydroxide is detrimental to the low temperature performance of asphalt binder generally, the adverse effect of LDHs is very slight and even can be neglected.

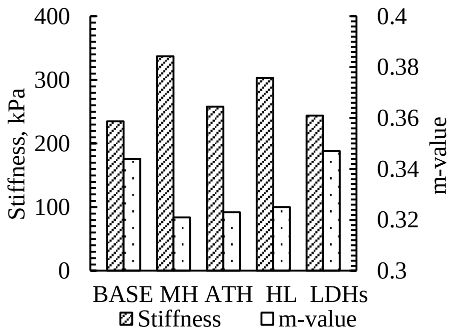


Fig. 10. Stiffness and m-value of base binder and metal hydroxide modified binders at the loading time of 60 second at – 12 °C

4. COST COMPARISON

To further investigate the feasibility of the application of these metal hydroxide flame retardancy in field, material cost should also be considered. Observed from the cost comparison in Table 3, HL is the most economical choice as the flame retardancy for asphalt, while the procurement price becomes a significant issue for the field application of LDHs.

Table 3. Cost comparison for metal hydroxides

Metal hydroxide	Cost, RMB/kg
MH	5.3
ATH	5.0
HL	0.5
LDHs	25.0

5. CONCLUSIONS

On the basis of the results of RV test, DSR test, as well as BBR test, the effect of MH, ATH, HL and LDHs on the rheological properties of asphalt binder are investigated, with the main conclusions as follows.

1. The rotational viscosity of asphalt binder increases with the addition of metal hydroxide, but still satisfies the requirement of flow capacity for mixing with the modifier dosage as 25 wt.%.
2. Metal hydroxide is able to improve the rutting resistance of asphalt binder at high temperatures due to the typical filler effect. However, the irreversible reaction between HL and acidic components in asphalt weakens the improvement.
3. HL and LDHs enhance the anti-aging resistance of asphalt binder by different mechanisms. HL is able to irreversibly absorb the acidic components in asphalt that is prone to aging, while the well-established multi-

layered structures of LDHs obstruct the infiltration of oxygen to asphalt, reducing the oxidation rate.

4. Although metal hydroxide is found to be detrimental to the resistance to thermal cracking of asphalt binder at low temperatures owing to the stress concentration, the negative effect of LDHs is particularly slight and can be neglected.
5. LDHs modified binder, which has been proved with fantastic flame resistance and environmental friendly from existing researches, as well as been found with great rutting resistance, anti-aging resistance and passable resistance to thermal cracking in this study, is recommended as the best choice of asphalt flame retardancy. However, the high procurement price of LDHs is still a big obstacle for its wider application.

Acknowledgments

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