Construction and Building Materials 75 (2015) 421–428

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Effect of ultraviolet aging on rheological properties of organic intercalated layered double hydroxides modified asphalt

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HIGHLIGHTS

- The organic intercalated layered double hydroxides (OLDHs) were used to modify asphalt.
- OLDHs restrain the increase of temperature susceptibility of asphalt caused by UV aging.
- DSR test results indicate OLDHs enhance the UV aging resistance of asphalt.

ARTICLE INFO

Article history:
Received 18 September 2014
Received in revised form 16 November 2014
Accepted 21 November 2014
Available online 5 December 2014

Keywords:
Asphalt
Organic intercalated layered double hydroxides
Ultraviolet aging
Temperature susceptibility
Rheological properties

ABSTRACT

The effect of ultraviolet (UV) aging on rheological properties of organic intercalated layered double hydroxides (OLDHs) modified asphalt was investigated by rotational viscosity and dynamic rheological properties. Experimental results indicate that the temperature susceptibility reflected by rotational viscosity and loss tangent of asphalt is decreased due to OLDHs, but the hardening caused by UV aging results in apparent decrease in temperature susceptibility of pristine asphalt, while the temperature susceptibility of OLDHs modified asphalt (OLMA) shows comparatively small decrement, exhibiting better UV aging resistance. Results of dynamic rheological properties show that the addition of OLDHs and LDHs increases the elastic component and rutting factor of asphalt binder at medium and high temperature, leading to a promotion of high temperature performance grade of asphalt. Owing to UV aging, the asphalt binder get stiffer and more brittle, which leads to more elastic behavior. However, the aging of OLMA are weakened for the smaller variations of rheological parameters as compared with pristine asphalt, which indicates excellent UV aging resistance of OLMA due to the introduction of OLDHs. Furthermore, OLDHs exhibit more effective modification than LDHs, which is due to the organic intercalation of LDHs that can improve the compatibility between LDHs and asphalt.

1. Introduction

Asphalt binder is widely used as adhesive materials for more than a century in the construction of flexible pavement due to its good viscoelastic properties, which plays a prominent role in determining many aspects of asphalt pavement [1]. However, asphalt is easily subjected to aging under the effect of traffic loading and complex environmental conditions, such as ultraviolet (UV) light, heat, and oxygen, during service life, which could lead to many types of failures, i.e. high temperature rutting, low thermal cracking, which can reduce the quality and performance of pavements and subsequently limit its further application [2].

Among those inducements causing the aging of asphalt, the UV irradiation is often ignored, not to mention the establishment of standard specification related to UV aging, which is due to the viewpoint that UV radiation only affects the upper layers of the asphalt pavement surfacing [3,4]. Nevertheless, more and more researchers pay attention to the UV aging performance of asphalt during the past few years, because the UV aging of asphalt binder is relevant in the areas that have high solar radiation intensity combined with high temperature, which are responsible for the decrease of the useful life of the asphalt pavement [5–7].

To improve the aging resistance of asphalt for long-life road applications, some attempts have been made by adding carbon black, montmorillonite, nanometer CeO_2, antioxidant and ultraviolet absorber, etc. into asphalt, and results show that asphalt binders with improved UV aging resistance could be obtained by adding these materials [8]. In recent years, layered double hydrox-

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http://dx.doi.org/10.1016/j.conbuildmat.2014.11.046
0950-0618/

ides (LDHs) are also employed as UV screening agent of asphalt, and the UV aging resistance of asphalt could indeed be improved due to the introduction of LDHs [9–11].

Asphalt is a viscoelastic material, which is highly depend on temperature and loading frequency at which the asphalt is used. Generally, the rheological characterization is a valid approach to evaluate the viscoelastic performance which is vital to the field application of asphalt binder. However, the previous researches about the effect of LDHs on properties of asphalt mainly focus on physical property evaluation, which is not enough to thoroughly understand the road performance of LDHs modified asphalt, especially for field application. On the other hand, LDHs possess strongly hydrophilic inorganic layers, the compatibility between LDHs and asphalt is very poor, that would lead to the difficult in obtaining homogeneous and stable dispersion of asphalt/LDHs composite. Therefore, it is very essential and significant to convert hydrophilic LDHs into more organophilic one [12–14].

In view of this point, the organophilic LDHs were synthesized and applied to modify asphalt for improving the UV aging resistance of asphalt. The viscosity-temperature profiles for pristine and modified asphalts before and after artificial accelerated UV aging were developed based on the rotational viscosity at different temperatures, and their temperature susceptibilities were analyzed. The rheological properties of all asphalt binders before and after UV aging have been investigated using dynamic shear rheometer. The temperature and frequency dependence of rheological properties were compared, and UV aging resistance for modified asphalt was evaluated.

2. Materials and experimental methods

2.1. Material

The 60/80 penetration-grade paving asphalt was used to prepare modified asphalt binder in this study, and the physical properties and chemical components of the pristine asphalt are listed in Table 1. The chemical components were determined by means of thin-layer chromatography with flame ionization detection (TLC-FID, Iatroscan MK-6 analyzer) as mentioned in the literature [15]. LDHs (MgAl–CO_3^{2-}–LDHs with Mg/Al = 2.0) were commercially available LDHs. Sodium dodecylbenzenesulfonate (SDBS) was analytically pure.

2.2. Synthesis of organic intercalated LDHs

The organic intercalated LDHs were prepared by the anion-exchange method. Firstly, to prepare LDHs slurry, MgAl–CO_3^{2}-LDHs powders (7.00 g) were suspended in the mixed solvent consisting of CO_2-free deionized water (100 mL) and anhydrous ethanol (100 mL) in a three-necked flask (1 L) under vigorous stirring at 70 °C. Secondly, SDBS solution (10.00 g SDBS dissolved in 400 mL CO_2-free deionized water) was directly added into LDHs slurry for the anion exchange, and the pH value of the mixture was adjusted to around 3 by adding adequate amount of HCl. Then, the anion exchange reaction took place by continuous stirring at 70 °C for 3 h under a nitrogen atmosphere. After reaction and cooling down, the white precipitation was separated and thoroughly washed with CO_2-free deionized water, and dried at 70 °C in a vacuum oven for 24 h. Finally, the organic intercalated LDHs (marked as OLDHs) were obtained after grounding to particle size of about 0.075 mm.

Table 1
Physical properties and chemical components of the pristine asphalt.

	Items	Measured values
Physical properties	Penetration (25 °C, 0.1 mm)	78
	Ductility (10 °C, cm)	17.1
	Softening point (°C)	44.4
	Viscosity (60 °C, Pa s)	227
	Viscosity (135 °C, Pa s)	0.49
Chemical components	Saturates (%)	13.26
	Aromatics (%)	45.82
	Resins (%)	31.27
	Asphaltenes (%)	9.65

2.3. Preparation of modified asphalt binders

All the modified asphalts were prepared by melt blending using a high shear mixer. Pristine asphalt was poured into an oil-bath heating iron container, and heated to a well fluid at around 140 °C. The weighed LDHs (3 wt.% by weight of asphalt) were added into the asphalt, and then, the blend was sheared for an hour at the shearing temperature of (140 ± 5) °C and shearing rate of 4000 rpm to ensure fully homogenous. Finally, the modified asphalt was poured into molds to conduct related experiments and tests. LDHs and OLDHs modified asphalt are denoted by LMA and OLMA, respectively. To compare with the modified asphalts preferably, the pristine asphalt was also treated under the same conditions.

2.4. UV aging procedures

The thin film oven test (TFOT) was employed to simulate short term thermal-oxidative aging of asphalt that occurs during the hot-mix process according to ASTM D1754. In the test, (50 ± 0.5) g melted asphalt sample was placed on a ∅ (140 ± 0.5) mm iron pan to form asphalt film which thickness was about 3.2 mm, and then the iron pan was put in the thin film oven to undergo thermal-oxidative aging at 163 °C for 5 h.

The UV accelerating aging treatment was carried out in an oven equipped with an UV lamp (the main wavelength was 365 nm) to simulate the photo-oxidative aging of asphalt in service life. As soon as the TFOT was over, the iron pans were transferred to an UV aging oven, the height from the pan to the lamp was adjustable to keep the average intensity of UV irradiation reaching to the asphalt's surface was about 1200 μW/cm^2. The asphalts underwent UV irradiation for 9 days at 60 °C in the oven.

2.5. X-ray diffraction (XRD) characterization

The X-ray diffraction (XRD) patterns of LDHs, OLDHs and modified asphalt were recorded using a D8 Advance diffractometer with Cu Kα radiation (λ = 0.15406 nm, 40 kV, 40 mA) at room temperature. The diffractive angle was scanned from 0.5° to 10° in the 2θ range of 0.02° steps, scanning rate was 2°/min.

2.6. Rotational viscometer test

Brookfield rotational viscometer (Model DV-II + Pro) was employed to measure the rotational viscosity (η) of the samples according to ASTM D4402. The testing temperatures varied from 60 °C, 90 °C, 120 °C, 135 °C to 150 °C in order to determine the viscosity-temperature profiles of the pristine and modified asphalt binders.

2.7. Dynamic shear rheometer (DSR) test

Dynamic rheological properties of the pristine and modified asphalts were measured by DSR (MCR102). Temperature sweep test, was performed under the strain-controlled mode at a constant frequency of 10 rad/s and with the temperature increment of 2 °C per minute, parallel plates with 8 mm diameter plate/2 mm gap between parallel plates and 25 mm diameter plate/1 mm gap between parallel plates were used for each sample in the temperature ranges of −20 to 30 °C and 30–80 °C, respectively. The principal rheological parameters, such as complex modulus (G^*), storage modulus (G'), loss modulus (G''), phase angle (δ) and complex viscosity (η^*), can be obtained from the DSR test.

Frequency sweep test was carried out from 400 to 0.1 rad/s. The testing temperatures were set at 0 °C and 10 °C. The plate used for frequency sweep test was 8 mm in diameter and the gap between the parallel plates was 2 mm.

3. Results and discussion

3.1. XRD analysis

The XRD curves for LDHs, OLDHs, LMA and OLMA are presented in Fig. 1. It could be easily observed that SDBS molecules are intercalated into the galleries of LDHs by anion-exchange, leading to the appearance of three diffraction peaks (003), (006) and (009) (corresponding 2θ values are 2.94°, 5.91° and 8.85°, respectively) in OLDHs, which is completely flat for LDHs in the investigated 2θ. The three diffraction peaks could be still observed in SLMB, whose 2θ angle positions are same with OLDHs. However, diffraction pattern of LMB is flat without any peaks as same as pattern of LDHs. The positions of diffraction peaks at low 2θ observed in SLMB and LMB are consistent with the corresponding OLDHs and LDHs cases, indicating that the OLDHs and LDHs are merely mixed in asphalt without destruction of lamellar structure due to the high