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Investigation of the ultraviolet aging resistance of organic layered double hydroxides modified bitumen

Song Xu [a], Lian Li [b], Jianying Yu [a,*], Canlin Zhang [a], Ji Zhou [a], Yubin Sun [c]

[a] *State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, PR China*
[b] *Institute of Ethics, Wuhan University of Technology, Wuhan 430070, PR China*
[c] *Center for Materials Research and Analysis, Wuhan University of Technology, Wuhan 430070, PR China*

HIGHLIGHTS

- Two organic LDHs were synthesized to modify bitumen.
- Organic intercalation improves the compatibility between LDHs and bitumen.
- Organic LDHs further enhance the UV aging resistance of bitumen.

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ABSTRACT

Organic layered double hydroxides (LDHs) intercalated with sodium dodecyl sulfate (SDS) and sodium dodecyl sulfonate (SDSO) were synthesized by anion-exchange method and used to ameliorate the compatibility between LDHs and bitumen, the ultraviolet (UV) aging resistance of organic LDHs modified bitumens were investigated by physical and rheological properties. The results show that the organic intercalation improves the compatibility between LDHs and bitumen due to the conversion of LDHs from hydrophilic to organophilic. Compared with LDHs modified bitumen, organic LDHs modified bitumens exhibit enhanced cracking resistance and high temperature rutting resistance. After UV accelerating aging, the changes of physical and rheological properties of organic LDHs modified bitumens are smaller than that of LDHs modified bitumen, indicating that organic LDHs are superior to LDHs in enhancing UV aging resistance of bitumen.

1. Introduction

Bitumen is a black adhesive material produced from crude oil, and its most common use is as the binder in the construction of flexible pavement due to its good viscoelastic properties [1,2]. However, during the lifetime of pavement, bitumen is exposed to numerous external factors, such as ultraviolet (UV) light, high temperature, oxygen etc., associated with aging inducements, which will lead to hardening caused by bitumen aging and further result in deterioration of pavement structure, i.e. high temperature rutting, low thermal cracking, thus reducing service life and riding quality of pavements [3–5].

Several methods have been developed to simulate the aging of bitumen and predict bitumen behavior during application and service life. The thin film oven test (TFOT, ASTM D1754) or rolling thin film oven test (RTFOT, ASTM D2872) is used to replicate the short-term aging that occurs during the mixing and laying down operations, while the pressurized aging vessel (PAV, ASTM D6521) is adopted to reproduce the long-term aging which occurs during long-term, in-service oxidative aging in the field [6,7]. Those techniques mentioned above mainly simulate the bitumen aging associated with thermal-oxidative aging that is caused by heat and oxygen, but the photo-oxidative aging which is caused by the ultraviolet (UV) radiation and oxygen is often ignored, and there is no standardized test that allows quantitative evaluation of the photo-oxidative of bitumen [8–13]. However, it has been found that the UV radiation would induce the aging of the upper layers of wearing courses of pavement and further affect the intermediate and bottom layers [14,15]. Therefore, the method concerning the improvement of UV aging resistance of bitumen is needed to developed for the durable pavement.

To improve the UV aging resistance of bitumen, some attempts have been made. For examples, carbon black could block the UV rays and alleviate UV aging of bitumen, but the addition of carbon black has negative effect on low-temperature performance of

* Corresponding author.
E-mail address: jyyu@whut.edu.cn (J. Yu).

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bitumen [16,17]. Some UV absorbers can improve the UV aging performance of bitumen, but the effects largely depend on the type of UV absorber and the nature of bitumen [18,19]. Recently, layered double hydroxides (LDHs), also known as hydrotalcite-like compounds with host–guest supramolecular structures, were also used as UV screening agent of bitumen, and the results showed that enhanced UV aging resistance of bitumen could be obtained by adding LDHs, the main reason is that the multi-layered structure of LDHs can physically reflect the UV light by the interface of metal hydroxide sheets, which can protect bitumen from destruction of high-energy UV [20–22]. However, due to the high charge density and strong hydrophilicity of inorganic layers of LDHs, strong attractive forces exist between the LDHs particles dispersed in bitumen matrix, which leads to poor compatibility between LDHs and bitumen, and further resulting in difficulty in obtaining homogeneous and stable dispersion of bitumen/LDHs composite [23]. Fortunately, the intercalation modification of LDHs via organic anion-exchange can convert hydrophilic LDHs into more organophilic one, and increase the compatibility of LDHs with bitumen [24–26].

In the present work, in order to improve the compatibility between LDHs and bitumen and further enhance the UV aging resistance of bitumen, two organic anionic compounds, sodium dodecyl sulfate (SDS) and sodium dodecyl sulfonate (SDSO), were intercalated into LDHs to prepare organic LDHs by anion-exchange method. The effects of organic LDHs on the properties of bitumen, including compatibility, physical and rheological properties, were investigated, and the UV aging performance of organic LDHs modified bitumen was also evaluated.

2. Materials and experimental methods

2.1. Material

The bitumen used in this study was 60/80 penetration grade, the physical properties and chemical components of the pristine bitumen 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, Iatron Laboratories Inc., Tokyo, Japan) as mentioned in the literature [27].

LDHs (MgAl-CO_3^{2-}-LDHs with Mg/Al = 2.0) were commercially available LDHs, and the properties of LDHs are listed in Table 2. Sodium dodecyl sulfate (SDS) and sodium dodecyl sulfonate (SDSO), analytically pure, were used as organic modifiers.

2.2. Synthesis of organic LDHs

The organic 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, organic anionic compound solution (8.30 g SDS or 7.80 g SDSO 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 an 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 LDHs (OLDHs, SDS and SDSO intercalated LDHs are marked as SDS-LDHs and SDSO-LDHs, respectively) were obtained after grounding to particle size of 200 mesh.

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

	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

Table 2
The properties of LDHs.

Items	Results
Appearance	White powder
Whiteness (%)	⩾90
Bulk density (g/cm^3)	0.38
Moisture content (105 °C for 1 h, wt.%)	⩽0.5
Specific surface area (m^2/g)	34

2.3. Preparation of modified bitumen

All the modified bitumens were prepared by melt blending using a high shear mixer. Pristine bitumen was poured into an oil-bath heating iron container, and heated to a well fluid at around 140 °C. The weighed LDHs or OLDHs (3 wt.% by weight of bitumen as determined by our previous study [28]) were added into the bitumen, 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 bitumen was poured into molds to conduct related experiments and tests. LDHs, SDS-LDHs and SDSO-LDHs modified bitumen are denoted by LMB, SDS-LMB and SDSO-LMB, respectively. To compare with the modified bitumens preferably, the pristine bitumen was also treated under the same conditions.

2.4. High temperature storage stability test

Hot storage test is used to evaluate the high temperature storage stability of modified bitumen. The storage stability test for modified bitumen was conducted as following procedure: the tested bitumen sample was heated to a well fluid and poured into an aluminum foil tube with a diameter of 25.4 mm and a length of 140.0 mm. After sealing the tube without air enclosure, it was placed vertically in an oven for 48 h at (163 ± 5) °C. Then the tube containing bitumen sample was took out of the oven and cooled in a refrigerator at −5 °C. Once cooled, the tube was cut horizontally into three equal sections, and the softening points (Ring and Ball test, ASTM D36) of the top and the bottom sections were tested. The difference of softening point (ΔS) between the top and the bottom sections was used to determine the high temperature storage stability of modified bitumen. If the ΔS was less than 2.2 °C, the sample could be regarded as storage stable blend, and the smaller the ΔS was, the better storage stability the sample had.

2.5. UV aging procedures

The thin film oven test (TFOT) was employed to simulate short term thermal-oxidative aging of bitumen that occurred during the hot-mix process according to ASTM D1754. In the test, (50 ± 0.5) g melted bitumen sample was placed on a ∅ (140 ± 0.5) mm iron pan to form bitumen 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 (power: 500 W, main wavelength: 365 nm) to simulate the photo-oxidative aging of bitumen 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 radiation reaching to the bitumen's surface was about 1200 $\mu W/cm^2$. The bitumens underwent UV radiation for 9 days at 60 °C in the oven.

2.6. X-ray diffraction (XRD) characterization

The X-ray diffraction (XRD) patterns of LDHs, OLDHs and their modified bitumen were recorded using a D8 Advance diffractometer (Bruker Corporation, Germany) with Cu-Kα radiation (λ = 0.15406 nm, 40 kV, 40 mA) at room temperature. The diffractive angle was scanned from 0.5° to 15° in the 2θ range of 0.02° steps, scanning rate was 2°/min.

2.7. Physical properties test

The physical properties of pristine and modified bitumen, including softening point, penetration (25 °C) and ductility (10 °C), were tested according to ASTM D36, ASTM D5 and ASTM D113, respectively.

Brookfield rotational viscometer (Model DV-II + Pro, Brookfield Engineering Inc., USA) was employed to measure the rotational viscosity (135 °C) of the samples according to ASTM D4402.