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Investigation of sodium stearate organically modified LDHs effect on the anti aging properties of asphalt binder

Yuanyuan Li [a], Shaopeng Wu [a,*], Ying Dai [b], Ling Pang [a], Quantao Liu [a], Jun Xie [a], Dezhi Kong [a]

[a] State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, China
[b] State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China

HIGHLIGHTS

- Sodium stearate was used to organically modify the LDHs.
- Asphalt with SS-LDHs has better storage stability than that of LDHs/SBS MA.
- Aging resistances of LDHs/SBS MA and SS-LDHs/SBS MA are better than SBS MA.
- SS-LDHs has better improvement effect on aging resistance of asphalt than LDHs.

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ABSTRACT

The layered double hydroxides (LDHs) has been widely used as the anti-aging modifier to improve the anti aging performance of an asphalt binder, but for the different chemical and physical properties of LDHs with asphalt binder, the compatibility of them is not very well, therefore the best modification effect can not be obtained. The sodium stearate was designated to organically modify the LDHs to enhance its improvement effect on the anti aging performance of asphalt binder. The modern test instruments such as Fourier transform infrared spectroscopy, X-ray diffractometer, scanning electron microscopy, ultraviolet visible spectrophotometer were used to investigate the sodium stearate effect on the characteristics of LDHs. In addition, the LDHs modified styrene–butadienestyrene modified asphalt (LDHs/SBS MA) and sodium stearate organically modified LDHs (SS-LDHs) modified styrene–butadienestyrene modified asphalt (SS-LDHs/SBS MA) were prepared by the melt blending method, the thin film oven test and UV aging test were conducted to simulate the short term thermo-oxidative aging and UV aging of asphalt binder respectively. Finally, the chemical structure, viscosity, and complex modulus of LDHs/SBS MA and SS-LDHs/SBS MA before and after aging were tested respectively. The results show that the sodium stearate can improve the compatibility of LDHs with styrene–butadienestyrene modified asphalt binder (SBS MA). Both LDHs and SS-LDHs can improve the anti thermo-oxidative aging and anti UV aging performance of SBS MA. Notably, the improvement effect of SS-LDHs on the anti aging performance of SBS MA is much better than that of LDHs.

1. Introduction

The layered double hydroxides (LDHs) is a layered nanomaterial with the structure of being similar to brucite. LDHs can prolong the distance of oxygen to penetrate to the internal of an asphalt binder (winding path) by its barrier performance [1], therefore decrease the penetration rate of oxygen; meanwhile, the host layers and interlayer anion have a chemical absorption and physical shielding properties of UV light respectively, which can make LDHs to be a good kind of UV barrier material. For the above two reasons, LDHs can improve the anti thermo-oxidative aging and anti UV aging performance of asphalt binders. However, there are lots of unbridged hydroxyl groups on the surface of LDHs particles, it is easy to form a soft agglomeration by hydrogen bonds between adjacent particles, and form a hard agglomeration through surface hydroxyl condensation [2,3]. In addition, hydroxyl group is a kind of hydrophilic group, which can make the LDHs shows a strong hydrophily [4,5], decrease the compatibility of LDHs with asphalt binder, as well as decrease the modification effect of LDHs on the

* Corresponding author.
E-mail addresses: liyuanyuan@whut.edu.cn (Y. Li), wusp@whut.edu.cn (S. Wu), yingdai@whut.edu.cn (Y. Dai), lingpang@whut.edu.cn (L. Pang), liuqt@whut.edu.cn (Q. Liu), xiejun3970@whut.edu.cn (J. Xie), Kongdz@whut.edu.cn (D. Kong).

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anti aging performance of an asphalt binder [6]. So it is important to organically modify the LDHs to improve the compatibility between LDHs with asphalt binder.

At present, the methods of organic modification of LDHs are mainly divided into three methods, namely exfoliation [7], intercalation [8,9] and surface modification [4]. The charge density of LDHs is about 2–3 times larger than that of other layered structure materials such as laponite and montmorillonite, a strong electrostatic effect exists between the host layers and interlayer anion [10], it is difficult to form a complete exfoliation [7]. The distance between the host layers of LDHs is small (approximate 0.76 nm), and there is also a strong hydrogen bonding effect between the interlayer anions, which makes it difficult for the polymer's molecular chains or branches to insert into the LDHs layers [10]. While, the surface modification method is simple and easy to implement compared with the exfoliation and intercalation modification [11,12].

The chemical formulas of sodium stearate is $C_{17}H_{35}COONa$, there are contain 17 carbon atoms in its alkyl chain, the alkyl chain length of which is relative longer than the commonly used surface modification agents [13,14], this is good for sodium stearate to winding and form good miscibility with polymer molecules, so the sodium stearate may have a better modification effect [6,15]. Zhang [16] used the one step method to prepare the sodium stearate Organically-modified CoAl-LDHs, the results demonstrated that the storage modulus of the bio-nanocomposites with 5 wt% of sodium stearate Organically-modified Co-Al-LDHs was enhanced remarkably. The research work of Kotal M [17] indicated that stearate-intercalated could significantly improve the tensile strength and elongation at break of thermoplastic polyurethane (PU). Pradhan B [18] demonstrated that Stearate ion-modified Mg–Al-LDHs improve the thermal degradation temperature and melting and crystallization temperatures of silicone rubber. The previous research work about sodium stearate used for the surface modification of LDHs indicted that, the sodium stearate was successfully attached on the surface of LDHs, the layered structure of LDHs was not destroyed, and the hydrophilic of LDHs was improved significantly [15]. So it has technical feasibility to use the sodium stearate as the surface modifier for LDHs to improve its modification effect on the anti aging performance of asphalt binders.

In this paper, the sodium stearate was designated to organically modify the Mg-Al-LDHs (SS-LDHs), the LDHs modified asphalt and SS-LDHs modified asphalt were prepared by melt blending method respectively, these three asphalt binders were aged at the same condition (short term thermo-oxidative aging and UV aging tests) to investigate whether the sodium stearate can enhance the modification effect of LDHs on the anti thermo-oxidative aging and anti UV aging performance of styrene-butadienestyrene modified asphalt binder (SBS MA).

2. Materials and experiments

2.1. Materials

2.1.1. Asphalt binder

A commonly used asphalt binder SBS MA with 4.5% dosage of SBS was selected, the PG grade of which was PG76-28. The technical information of SBS MA is shown in Table 1.

2.1.2. LDHS and sodium stearate organic modified LDHs

The sodium stearate modified LDHs (SS-LDHs) was produced by wet modified method [23]. First, weighed 30 g of LDHs and set in a beaker with the volume of 200 ml, and added 100 ml deionized water; second, a designed content (6% by weight of the LDHs) of sodium stearate was added into the beaker, set the beaker in a water bath and heated to a constant temperature of 80 °C, a shear apparatus was used to slowly stir for two hours, the shear rate was 500 r/min; finally, the sediment was drained and washed to neutral, set the filter cake into drying oven at the temperature of 80 °C until to a constant weight. The technical information of LDHs and SS-LDHs were shown in Table 2.

Table 1
Technical information of SBS MA.

Information	Units	Results	Methods
Penetration (25 °C, 100 g, 5 s)	0.1 mm	61.7	ASTM D5 [19]
Softening point	°C	78.2	ASTM D36 [20]
Ductility (5 °C)	cm	33.3	ASTM D113 [21]
Viscosity (135 °C)	Pa.s	1.337	ASTM D4402 [22]

Table 2
Information of LDHs and SS-LDHs.

Information	Units	LDHs	SS-LDHs
MgO/Al_2O_3	–	4 ± 0.2	4 ± 0.2
LDHs content	%	≥99.5	–
Mass loss (105 °C)	%	≤0.5	≤0.5
Specific surface area	m^2/g	1.34	17.4
Average particle size	<mu>m	5.267	2.334

2.2. Experimental methods

2.2.1. Preparation of modified asphalt binder

First, SBS MA was heated to 170 °C; then the 4.0% contents of the LDHs or SS-LDHs were added to the asphalt binder, and used the high speed shear mixing machine to shear for 40 min at the shear rate of 4000 r/min. For the comparison purposes, the SBS MA without LDHs and SS-LDHs were also gone through the same heating and shearing process as LDHs modified SBS MA (LDHs/SBS MA) and SS-LDHs modified SBS MA (SS-LDHs/SBS MA), so that the aging resistance performance of asphalt binders could be evaluate at the same condition.

2.2.2. Experiments of LDHs and SS-LDHs

The characterizations of LDHs and SS-LDHs were studied by the Fourier transform infrared spectroscopy (FTIR), X-ray diffractometer (XRD), scanning electron microscopy (SEM) and Ultraviolet visible spectrophotometer (UV–Vis) respectively. The FTIR was used to study the chemical structure of LDHs and SS-LDHs, the samples for FTIR were prepared by KBr pellet method, the mass ratio of LDHs to KBr was 1:100. The wave number range was from 4000 to 400 cm^{-1}, scan times was set to 64 times. The crystal structure of LDHs and SS-LDHs were studied by the XRD, the sample for XRD test was prepared by positive pressure method, the 2θ range was 1.5°–45° at scanning rate of 0.25°/s, the wavelength of the CuKα ray was 0.1506 nm. The SEM was used to investigate the surface micro structure of LDHs in the high vacuum, the voltage was 20KV and the resolution was 3 nm. The samples were dried at 80 °C for five hours before being tested. The Lambda 750 S UV–Vis produced by PerkinElmer was conducted to investigate the light barrier properties of LDHs and SS-LDHs, the bandwidth of which could be continuously adjusted at the range of 0.17–5.00 nm, the wave length range was 200–800 nm.

2.2.3. Experiments of asphalt binders

The segregation test was conducted to study the storage stabilities of asphalt binders, it could be used to evaluate weather sodium stearate can improve the compatibility of LDHs with SBS MA. The samples were poured into an aluminum tube with the diameter and height of 25 mm and 140 mm, respectively. Set the aluminum tube in an oven at the constant temperature of 163 °C for 48 h and then cooled in a refrigerator at 5 °C for more than 4 h. After that, the tubes were cut into three equal sections. The softening points of top and bottom sections were tested to calculate the softening point differences. The chemical structures of asphalt binders before and after aging were also tested by FTIR. First, prepared the 5 wt% of asphalt CS_2 solution, then used the glue dropper to drop three drops of solution on the KBr chip, finally, it could be tested after the CS_2 being volatilized completely. The wave number range was from 4000 cm^{-1} to 400 cm^{-1}, scan times was 64 times. The viscosity test was conducted to study the viscosity of asphalt binder before and after aging according to the criterion of ASTM D4402. The complex modulus of asphalt binders were tested by DSR at the temperature range of 52–82 °C, the rotor diameter was 25 mm, the sample thickness was 1.0 mm, the frequency and strain were 10 rad/s and 0.5%, respectively.

2.2.4. Aging simulating test of asphalt binder

The samples used for the ultraviolet aging were first aged by thin film oven test (TFOT). The sample mass of TFOT was 50 ± 0.5 g, diameter of the sample plate was 140 mm, asphalt film thickness was approximate 3.2 mm, test temperature was stable at 163 ± 0.5 °C for 5 h. After TFOT, the UV aging simulation test was performed by the straight pipe high pressure mercury lamp, the UV radiation intensity