



## Research paper

# Synthesis and characterization of triethoxyvinylsilane surface modified layered double hydroxides and application in improving UV aging resistance of bitumen



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## ABSTRACT

Layered double hydroxides (LDH) surface modified by triethoxyvinylsilane (TEVS) was prepared and characterized by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM) and Ultraviolet and visible (UV–Vis) spectrophotometry. FTIR and XRD showed that TEVS has been successfully grafted onto the surface of LDH. Compared with LDH, the agglomeration between the TEVS surface modified LDH (TEVS-LDH) particles was obviously weakened according to SEM. UV–Vis reflectance and absorbance analyses indicated that TEVS-LDH possessed better UV reflectance and absorbance than that of LDH. Then the effects of TEVS-LDH and LDH on the physical and anti-aging properties of bitumen were thoroughly evaluated. The storage stability test showed that TEVS-LDH existed more stably than LDH in bitumen, which was due to the surface modification of LDH which improved the compatibility between LDH and bitumen. The results of physical properties tests showed that TEVS-LDH improved high-temperature behavior and had a less pronounced effect on the low temperature ductility of bitumen in contrast with LDH. Aging indices (viscosity aging index, softening point increment and ductility retention rate) and FTIR showed that TEVS-LDH exhibited better improvement than LDH on anti-ultraviolet aging of bitumen.

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## 1. Introduction

Layered double hydroxides (LDH) was a kind of anionic clay, also known as hydrotalcite-like compounds with host-guest supramolecular structures. The general formula of LDH was often written as  $[M^{II}_1 - x M^{III}_1 - x(OH)_2]^{x+}(A^{n-})_{x/n} \cdot mH_2O$  ( $M^{II}$  and  $M^{III}$  are divalent and trivalent metal cations, respectively) (Ma et al., 2006; Chakraborty et al., 2015; Li et al., 2015). Owing to the versatility in chemical composition and unique physical structure of LDH, LDH has been intensively studied and widely used in different materials in recent years. For example, LDH has been applied in polymers to improve mechanical properties, thermal stability, flame-retardant properties and anti-ultraviolet aging performance (Vaccari, 2002; Carrado and Bergaya, 2007; Xue and Pinnavaia, 2007; Manzi-Nshuti et al., 2009; Kovanda et al., 2010; Zhang et al., 2010; Ay et al., 2011; Nogueira et al., 2011; Li et al., 2013; Basu et al., 2014; Omwoma et al., 2014).

Bitumen was widely used as an adhesive binder in the construction of flexible pavement due to its good viscoelastic properties, which played a prominent role in determining many aspects of bitumen-based materials (Galooyak et al., 2010; Zhang et al., 2011). However,

like other organics, bitumen was vulnerable to aging when exposed to heat, oxygen or ultraviolet (UV) light during its mixing, paving and compaction, as well as service life, which resulted in the age-hardening and sacrifice of desirable physical properties of bitumen (i.e. high-temperature rutting, low-temperature cracking) (Dehouche et al., 2012; Abu Al-Rub et al., 2013; de Sá Araujo et al., 2013), and ultimately failed to meet the performance requirements and subsequently limited its further application (Miró et al., 2015; Xiao et al., 2015). Therefore, it was of pragmatic significance to increase durability of pavement by improving the bitumen with respect to anti-aging-related properties. A series of experiments have been conducted to search for bitumen with preferable aging resistance. The most widely used methods were to add anti-aging additives in bitumen. For instance, carbon black has been used to improve the anti-ultraviolet aging performance of bitumen, and carbon black could enhance anti-ultraviolet aging ability of bitumen to some extent. However, carbon black could seriously deteriorate the low-temperature performance of bitumen (Cong et al., 2014).

In recent years, it has been reported that LDH could be used as anti-ultraviolet aging additives to improve UV aging resistance of bitumen, it was attributed to the multi-layered structure of LDH, which had excellent physical shielding and chemical absorption to UV light (Pang et al., 2014; Xu et al., 2015d). Unfortunately, LDH was a kind of inorganic

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materials with strong hydrophilicity, and had different physical properties (e.g., density and surface charge) from bitumen, which resulted in poor compatibility between LDH and bitumen (Dong et al., 2014). Because of this, LDH powder centralized in the upper layer of bitumen while mixing, the mixture in storage would have the tendency of phase separation which further led to weaken the anti-ultraviolet functions of LDH (Wang et al., 2005). Therefore, it was crucial to modify LDH to improve the compatibility between LDH and bitumen.

Some efforts have been made to improve the compatibility between LDH and bitumen. For example, some organics (i.e. 2-hydroxybenzoic acid, sodium dodecylbenzenesulfonate and 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid) were intercalated into LDH to synthesize organic LDH by anion exchange method (Xu et al., 2015a, b). However, organics were fixed into the interlayer of LDH, and the bitumen molecular chains could not enter the interlayer (Xu et al., 2015c). The improvement on the compatibility between LDH and bitumen was limited by organic intercalation. LDH possessed a multitude of free hydroxyls on the surface, which would lead to its hydrophilicity and the agglomeration between LDH particles. Hence, it could enhance the compatibility between LDH and bitumen by reducing the hydroxyl groups and increasing lipophilic group on the surface of LDH. Based on this, silane coupling agents, which were known as “molecular bridges” and frequently used to connect organic and inorganic materials by means of forming hydrogen bonds or chemical bonds (Li et al., 2008; Xie et al., 2010; Min et al., 2015; Xu et al., 2015d), could be applied to improve the compatibility between LDH and bitumen. Firstly, the hydrolysis of silane coupling agents would generate Si–OH, which could react with hydroxyl groups on the surface of LDH to form hydrogen bonds and covalent bonds. Then, the organic groups of silane coupling agents combined with bitumen through molecular entanglement. In this way, modified LDH could disperse homogeneously and exist stably in bitumen. Compared with the method of organic intercalation, silane coupling agents surface modification made the surface of LDH coated by organics, which could more efficiently resolve the compatibility between LDH and bitumen.

In this paper, triethoxyvinylsilane (TEVS) was selected to modify LDH to enhance the compatibility between LDH and bitumen, and the structures and properties of TEVS-LDH were characterized by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), Scanning electron microscopy (SEM), Ultraviolet and visible (UV–Vis) spectrophotometry, and the effects of TEVS-LDH on the UV aging resistance properties of bitumen were also investigated by UV exposure test.

## 2. Experimental

### 2.1. Materials

LDH ( $\text{MgAl}-\text{CO}_3^{2-}$ -LDH with  $\text{Mg}/\text{Al} = 2.0$ ) were commercially provided by Beijing Tech-layer Co., Ltd., Beijing, China. Triethoxyvinylsilane (TEVS) was purchased from Aladdin Industrial Co., Shanghai, China. Acetic acid and Anhydrous ethanol were purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. Bitumen, SK-70 paving bitumen which was produced by the SK Petroleum Asphalt Factory, Southern Korea. The physical properties of pristine bitumen were listed in Table 1.

**Table 1**  
Physical properties of bitumen.

Physical properties	Measured values
Penetration (25 °C, 0.1 mm)	73
Ductility (10 °C, cm)	16.5
Softening point (°C)	48.8
Viscosity (135 °C, Pa·s)	0.49

### 2.2. Preparation of TEVS surface modified LDH

Firstly, LDH powders (50 g) were suspended in the deionized water (475 mL) and anhydrous ethanol (25 mL) in a three-necked flask (1 L) under vigorous stirring at 50 °C for 30 min. TEVS solution was also obtained by dissolving TEVS (1.5 g) in the mixture of ethanol (45 mL) and deionized water (5 mL). After mixing the two solutions, acetic acid was added dropwise with vigorous stirring at 50 °C until the pH of the reaction system reached to 3.5. Then the resulting slurry was reacted at 50 °C for 3 h with vigorous stirring in  $\text{N}_2$  stream and the reaction subsequently continued at 70 °C under  $\text{N}_2$  protection for about 30 min. Finally, the white precipitate was filtered, and washed completely with  $\text{CO}_2$ -free deionized water for six times and dried at 80 °C in a vacuum oven for 24 h. The dried product was then grinded to obtain TEVS surface modified LDH (denoted as TEVS-LDH) with a particle size of 200 mesh.

### 2.3. Preparation of modified bitumen

The modified bitumen was prepared using a high shear mixer. Firstly, pristine bitumen was heated to a well fluid at around 140 °C in an oil-bath heating iron container, and then the proper amount of TEVS-LDH or LDH (1%, 2%, 3% and 4% by weight of bitumen) was added to the bitumen. Then, the blend was sheared for an hour at the shearing temperature of 170 °C and a rate of 4000 rpm. Finally, the modified bitumen was poured into molds for later experiments. Pristine bitumen, TEVS-LDH and LDH modified bitumen are denoted as PB, TEVS-LMB and LMB for simplicity, respectively. The pristine bitumen was also prepared under the same conditions in order to keep consistency with the modified bitumen.

### 2.4. Aging procedures

The aging of bitumen was performed using thin film oven test (TFOT) and UV radiation, respectively. TFOT was executed during 5 h at 163 °C in air as described in the standard ASTM D1754, to simulate the thermal-oxidative aging of bitumen that occurs in processing of mixing with aggregates and compaction. The residue from the TFOT was UV-aged for 7 days in a draft oven with an UV lamp of 500 W (the main wavelength is 365 nm) to simulate the photo-oxidative aging of bitumen in service life. The intensity of UV radiation was 2000  $\mu\text{W}/\text{cm}^2$ . The melted bitumen was placed on a  $\emptyset$  (140  $\pm$  0.5) mm iron pan which was placed at the bottom of the chamber, and the thickness of bitumen film was about 3.2 mm. The vertical distance from the pan to the lamp was 500 mm. The working temperature was controlled at (60  $\pm$  3) °C.

### 2.5. Characterization

A FTIR spectrometer (Nexus, Thermo Nicolet Corp., U.S.) was used to obtain the spectra of solid powder TEVS-LDH, LDH and modified binders before and after aging by using the KBr disk method (1 mg of LDH in 100 mg of KBr) and liquid film method (three drops of bitumen solution with 5 wt.% concentration), respectively. All spectra were recorded in wavenumber ranging from 4000 to 400  $\text{cm}^{-1}$ . The number of scan was 64, and the spectral resolution was 4  $\text{cm}^{-1}$ .

The X-ray diffraction (XRD) patterns of TEVS-LDH and LDH were recorded using a D8 Advance diffractometer (Bruker Corporation, Germany) with  $\text{Cu}-\text{K}\alpha$  radiation ( $\lambda = 0.15406$  nm). The test was executed under the tube voltage of 40 kV and current of 40 mA at room temperature, and the  $2\theta$  diffractive angle was scanned from 1.5° to 60° with the step size of 0.02° steps, scanning rate was 2°/min.

Morphological features of the samples were studied using a field emission scanning electron microscope scanning electron microscopy (Quanta 250 FEG, FEI, Oregon, USA). SEM images were taken on the microscope at a voltage of 20 kV.

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