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Rheological examination of aging in bitumen with inorganic nanoparticles and organic expanded vermiculite

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HIGHLIGHTS

- This work evaluates a novel bitumen modification through the use of OEVMT and inorganic nanoparticles.
- The rheological properties of the binders were evaluated before and after TFOT, PAV and UV aging.
- The improvement in aging resistance of bitumen depends on the inorganic nanoparticles type.

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ABSTRACT

Bituminous pavement is vulnerable to aging during construction and service life, which can seriously affect the rheological properties of bitumen. A novel modified bitumen containing inorganic nanoparticles and organic expanded vermiculite (OEVMT) compound was evaluated in this paper. The binders were aged according to the thin film oven test (TFOT), pressure aging vessel (PAV) test and ultraviolet (UV) radiation, respectively. The effect of different inorganic nanoparticles (nano-titania, nano-zinc oxide, nano-silica) and OEVMT compound on rheological properties of bitumen before and after aging is investigated. The base bitumen is modified with 1% OEVMT by mass of bitumen and different content inorganic nanoparticles, which are 1%, 2% and 3% nano-titania (nano-TiO_2), 3% nano-zinc oxide (nano-ZnO) and 3% nano-silica (nano-SiO_2). The microstructures of expanded vermiculite (EVMT), OEVMT and separated OEVMT from modified bitumen were characterized by X-ray diffraction (XRD). The results show that the OEVMT/bitumen forms the exfoliated nanostructures. The complex modulus of bitumen is increased, while the phase angle is decreased with the introduction of compound modifiers. As a result of TFOT, PAV and UV aging, inorganic nanoparticles and OEVMT modified bitumens show the higher phase angle aging index and the lower complex modulus aging index in comparison with base bitumen, which indicates their good thermal oxidation and UV aging resistance. Moreover, based on nano-TiO_2 content effect, the recommended content is 1% OEVMT + 3% nano-TiO_2 by rheological properties results before and after three aging methods. Under the same compound modifiers content, the compound modified bitumen containing nano-ZnO shows better aging resistance than the compound modified bitumen containing nano-TiO_2 or nano-SiO_2.

1. Introduction

Bitumen pavement plays an increasingly important role in road construction at present due to its excellent road performance, such as driving comfort, safely, easy maintenance and so on [1,2]. However, bitumen is vulnerable to aging in the process of construction and service time because of the influence of temperature, oxygen, ultraviolet light or multiple factors. Based on different factors, the aging is usually divided into thermal oxidation aging and photo oxidation aging. In addition, the thermal oxidation aging can be further subdivided into short-term and long-term oxidation aging. The former occurs at high temperature (above 150 °C) when bitumen undergoes processing, mixing, paving and compaction procedures, while the latter occurs on service time. Many diseases of bitumen pavement may be caused by aging, e.g. cracking, raveling, distortion and so on, which will reduce the quality and performance of pavements seriously [3–7]. Therefore, for improving durability of bitumen pavement, it is of great importance to enhance aging resistance of bitumen.

Recently, various inorganic nanomaterials have been used to modify bitumen [8–19]. Especially for inorganic nanoparticles

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and layered silicates, many published researches showed that the performance of bitumen can be improved with the introduction of inorganic nanoparticles or layered silicates effectively. With respect to inorganic nanoparticles, Ye et al. [11] found that nano-SiO_2 or nano-TiO_2 added in bitumen could improve pavement performance of bitumen. Fini et al. [19] also investigated the effect of nano-SiO_2 on properties of bitumen, it could be concluded that addition of nano-SiO_2 in bitumen could improve the rheological properties and short-term thermal oxidation aging resistance of bitumen. Yang et al. [15] found that the right amount of TiO_2 could enhance the short-term thermal oxidation aging and ultraviolet (UV) aging resistance of bitumen. Xiao et al. [8] found that the high or low temperature performance and short-term thermal oxidation aging resistance performance of styrene–butadiene–styrene copolymer (SBS) modified bitumen could be improved by means of adding nano-zinc. Zhang et al. [17] further found that nano-ZnO can be effectively used as a modifier to improve the UV aging resistance of bitumen with 60/80 penetration grade.

In addition, in terms of layered silicates, Yu et al. [9] investigated the preparation and properties of montmorillonite (MMT) modified bitumen, the result indicated that the MMT and organic montmorillonite (OMMT) modified bitumen displayed enhanced viscoelastic properties, which improved its resistance to rutting at high temperatures. Jahromi et al. [12] studied that the effect of nanoclay on the rheological properties of bitumen binder, and found that nanoclay modification could increase the stiffness of bitumen, decrease the phase angle and improve the thermal oxidation aging resistance compare with unmodified bitumen. Zhang et al. [16] draw a conclusion that expanded vermiculite (EVMT) and organic expanded vermiculite (OEVMT) clays could improve the long-term thermal oxidation and UV aging resistance of bitumen binders effectively. Moreover, it could be found that the improved aging resistance of the bitumen by EVMT/OEVMT after long-term thermal oxidation aging was more obvious than that after UV aging. In order to compare the influence of nano-ZnO and OEVMT on the long-term thermal oxidation and ultraviolet aging of bitumen, Zhang et al. [18] found that OEVMT could improve thermal oxidation aging significantly, while nano-ZnO had little influence on the long-term thermal oxidation aging properties of bitumen. On the contrary, nano-ZnO could enhance UV aging effectively, while OEVMT had little influence on UV aging.

From all above, it can be concluded that inorganic nanoparticles as well as layered silicates can improve performance of bitumen. However, based on their different characteristics, inorganic nanoparticles are more efficient in improving UV aging of bitumen, while layered silicates show better improvement in thermal oxidation aging resistance of bitumen. Accordingly, the compound agents containing inorganic nanoparticles and layered silicates can be promising modifiers to improve both thermal oxidation aging and UV aging resistance of bitumen simultaneously. Additionally, given that the types of inorganic nanoparticles and layered silicates are diversity, it is necessary to seek an optimal inorganic nanoparticle or layered silicate.

In this paper, the effect of nano-TiO_2 content on the rheological properties of modified bitumen containing nano-TiO_2 and OEVMT was investigated. The base bitumen was modified with 1% OEVMT by mass of bitumen and different content inorganic nanoparticles, which are 1%, 2% and 3% nano-titania (nano-TiO_2). Moreover, in order to compare the effectiveness of nano-TiO_2 and nano-SiO_2 and nano-ZnO, 1% OEVMT + 3% nano-SiO_2 and 1% OEVMT + 3% nano-ZnO were also used to modify bitumen.

2. Experimental

2.1. Materials

The base bitumen (SK-70) with 60/80 penetration grade was used in this research. The physical properties of the base bitumen are as follows: penetration, 65 dmm at 25 °C; softening point, 47.4 °C; ductility, exceed 150.0 cm at 15 °C; viscosity, 449 mPa · s at 135 °C. Cetyltrimethyl ammonium bromide (CTAB, chemically pure) used as intercalation agent for cation exchange to modify expanded vermiculite (EVMT, 300 mesh) interlayer surface. Nano-titania, nano-silica and nano-zinc oxide, 15–25 nm, were surfaced modified by γ-(2, 3-epoxypropoxy) propyltrimethoxysilane.

To prepare OEVMT, a 500 mL round-bottom, three-necked flask with a mechanical stirrer, thermometer, and condenser with a drying tube was used as a reactor. Firstly, EVMT (30 g) was added in three-necked flask. Then concentrated hydrochloric acid (9 g) and sodium chloride (9 g) were blended, the mixture was also added in three-necked flask together with a certain amount of deionized water, the temperature is controlled at 60 °C by temperature indicator-controller, and the resultant suspension is vigorously stirred at fitted speed for 2 h through electric mixer. The treated EVMT is repeatedly washed with deionized water by circulating water vacuum pump. The filtrate is titrated with 0.1 mol/L $AgNO_3$ until no precipitate is formed. Next, the EVMT filter cake was dissolved in a certain amount of deionized water again, CTAB (9 g) was also added into this solution. The mixture was poured in three-necked flask, the temperature is controlled at 80 °C. The next process was same with the above process. Finally, the filter cake is then placed in electric heating oven to dry at 105 °C for 6 h, the dried cake is then ground to obtain OEVMT with a particle size of 300 mesh.

2.2. Surface modification of inorganic nanoparticles

In this paper, γ-(2, 3-epoxypropoxy) propyltrimethoxysilane was used as a silane coupling agent to modify the surface of three inorganic nanoparticles. The inorganic nanoparticles (nano-SiO_2, nano-TiO_2 or nano-ZnO) were mixed into toluene containing γ-(2, 3-epoxypropoxy) propyltrimethoxysilane by using a paddle agitator, the pH value of the suspension was controlled at 6.0 using acetic acid (95 wt%). Then the suspension was stirred for 5 h. After that, toluene was removed by filtration process. The treated inorganic nanoparticles were repeatedly washed with acetone. The washed inorganic nanoparticles were then placed in a vacuum oven to dry at 120 °C for 4 h [20].

2.3. Preparation of inorganic nanoparticles and OEVMT modified bitumen

Bitumen was heated to 150 ± 5 °C in an oil-bath heating container until it flowed fully. Then a certain amount of inorganic nanoparticles and OEVMT were added into bitumen, and the mixture was blended at 4000 rpm for 60 min by using a high shear mixer. After that, the mixture was blended using a paddle agitator at a rotation speed of 2000 rpm at 150 °C for about 90 min to produce inorganic nanoparticles and OEVMT compound modified bitumen. The same process was also performed on the base bitumen in order to achieve blank sample.

2.4. Rheological properties test

The rheological properties of the bitumens were evaluated by complex modulus (G^*) and phase angle (δ). Dynamic shear rheometer (DSR) was performed to determine G^* and δ of the bitumens according to ASTM D 7175 [21]. The temperature sweep test was performed for temperature ranges of 40–80 °C with 5 °C intervals.

2.5. Aging procedures

The thin-film oven test (TFOT) (ASTM D 1754 [22]) was employed to simulated short-term thermal oxidation aging of the bitumen, and the pressure aging vessel (PAV) (ASTM D 6521 [23]) aging was used to simulate long-term thermal oxidation aging. The thin-film oven test is executed in an oven with a plat and an axis, and the rotation of plat is carried out by the axis. The bitumen, which is put on the plats, was heated for 5 h at 163 °C. The PAV apparatus consisted of the pressure aging vessel and temperature chamber. A cylinder of dry and clean compressed air is provided by a pressure regulator with air pressure. The standard aging procedure of 100 °C, 2.1 MPa, and 20 h for the PAV was used.

Photo oxidation aging was simulated by using ultraviolet (UV) radiation. The residue from the TFOT was UV-aged for 6 days in a draft oven ($850 \times 600 \times 600$ mm, together with fresh air) with an UV lamp of 500 W. The intensity of UV radiation was about 8 w/m^2. The melted bitumens were placed on a Φ140 ± 0.5 mm iron pan, which was placed at the bottom of the chamber, and the thickness of bitumen film was about 2.0 mm. The vertical distance from the pan to the lamp was 450 mm. The working temperature was controlled at 60 °C.