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Influence of surface modification on physical and ultraviolet aging resistance of bitumen containing inorganic nanoparticles

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HIGHLIGHTS

- The compatibility between inorganic nanoparticles and bitumen is improved by surface modification.
- The UV aging resistance of bitumen can be effectively improved by inorganic nanoparticles.
- The improvement in UV aging resistance is significantly influenced by surface modification of inorganic nanoparticles.

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ABSTRACT

Three inorganic nanoparticles (nano-SiO_2, nano-TiO_2, nano-ZnO) were adopted to improve the ultraviolet (UV) aging properties of bitumen. Inorganic nanoparticles were surface modified by silane coupling agent. The compatibility between inorganic nanoparticles and bitumen was studied by high temperature storage stability test. More attention had been paid to the effect of surface modification on physical and UV aging properties of inorganic nanoparticles modified bitumens. The results showed that the compatibility between inorganic nanoparticles and bitumen was improved obviously by surface modification. Under the same content, the difference between the softening points as well as the nanoparticles contents in the upper and lower sections of the toothpaste tubes was decreased remarkably after surface modification. The penetration and ductility were decreased, while the softening point and viscosity of bitumen were increased with the addition of inorganic nanoparticles. However, this influence was weakened after surface modification of inorganic nanoparticles and the weakening effect of surface modified nano-ZnO was the most obvious. As a result of UV aging, softening point increment and viscosity aging index of bitumen were decreased by inorganic nanoparticles. The UV aging resistance of modified bitumen was further enhanced by surface modification of inorganic nanoparticles, and nano-ZnO modified bitumen showed the best UV aging resistance after surface modification.

1. Introduction

Bitumen pavement is the main type of high-grade highways because of its good road performance, which has been widely used in the world [1]. Bitumen pavement easily suffers from cracking, rutting, raveling and other diseases during their service life, which affects the performance and service life of bitumen pavement seriously. Many published researches showed that these damages were caused mainly by thermal aging and ultraviolet (UV) radiation aging occured by bitumen which suffers from temperature, oxygen, ultraviolet light, water and/or other external environmental factors [2,3]. Traditional bitumen has been unable to fully meet the requirements of high-grade highway, so more and more modified bitumen materials have been widely used in road construction.

Recently, nanomaterials have been used for modifying bitumen. Jahromi et al. [4] studied the effect of nanoclay on the rheological properties of bitumen binder, the results suggested that nanoclay added in bitumen changed rheological properties of bitumen, increased stiffness of bitumen, reduced phase angle of bitumen and enhanced aging resistance of bitumen. Galooyak et al. [5] studies showed that high temperature storage stability of styrene butadiene styrene (SBS) copolymer modified bitumen was improved by adding organic montmorillonite, which had no negative impact on other properties, and the uniformly dispersed degree of organic montmorillonite in the SBS modified bitumen was limited greatly by its amount. Polacco et al. [6] prepared bitumen/SBS/clay

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mixture through adding SBS and clay alone or blended to bitumen, rheological tests showed that blend modes had a great impact on the rheological properties of bitumen/SBS/clay mixture. Authors also have been working on studying inorganic nanomaterials modified bitumen, these researches indicated that montmorillonite added in bitumen can improve bitumen thermal aging resistance [7]. Compared with ordinary montmorillonite, organic montmorillonite showed the better compatibility with bitumen and the better dispersity in bitumen, thereby aging resistance of bitumen was improved more remarkably [8]. You [9] used nanotubes, nano-SiO_2 and other nanomaterials to modify bitumen, field emission scanning electron microscopy test showed that the nano-SiO_2 was well dispersed in bitumen.

Inorganic nanoparticles have also been used for modifying bitumen. Ye et al. [10] studied the performance of nano-SiO_2 modified bitumen and their mixture, which indicated that nano-SiO_2 added in bitumen can improve pavement performance of bitumen. Ma et al. [11] used nano-$CaCO_3$ to modify bitumen, the results showed that the temperature sensitivity and some pavement performance of bitumen could be improved by adding nano-$CaCO_3$. Amen et al. [12] studied the rheological properties of three bitumen binder sources containing various percentages of carbon nanoparticles, the results indicated that the addition of carbon nanoparticles was helpful in increasing the viscosity, failure temperature, complex modulus, and elastic modulus values as well as in improving rutting resistance of the binder. Xiao et al. [13] added nano-ZnO to SBS modified bitumen, the results indicated that the high or low temperature performance and thermo-oxidative aging resistance performance of SBS modified bitumen can be improved. Inorganic nanoparticles have huge surface area, extreme high surface energy and very easily agglomerate, which not only make uniform dispersion more difficult, but also may make their nanometer effect lost. In order to avoid these phenomenon, surface modification is necessary. However, there is few published research related to the effect of different inorganic nanoparticles and their surface modification on bitumen properties, especially ultraviolet aging properties.

In the paper, silane coupling agent is used for surface modifying inorganic nanoparticles. The effects of inorganic nanoparticles and their surface modification on the compatibility between inorganic nanoparticles and bitumen, physical properties of bitumen and UV aging properties of bitumen were investigated.

2. Experimental

2.1. Materials

The physical properties of the base bitumen with 70 penetration grade are listed in Table 1. The diameter of nano-SiO_2, nano-TiO_2 and nano-ZnO was 20–50 nm. γ-(2,3-epoxypropoxy) propyltrimethoxysilane was used as a silane coupling agent to modify three inorganic nanoparticles surfaces.

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 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 [14].

Table 1
Physical properties of the base bitumen.

Physical properties	Measured values
Penetration (25 °C, 0.1 mm)	73
Softening point (°C)	48.5
Ductility (15 °C/10 °C, cm)	145.0/16.1
Viscosity (60 °C, Pa s)	258

2.3. Preparation of inorganic nanoparticles modified bitumen

Base bitumen was heated to 150 ± 5 °C in an oil-bath heating container by closed electrothermal furnace, then the right amount of inorganic nanoparticles or surface modified inorganic nanoparticles was added into bitumen. First, the mixture was blended at 4000 rpm for 1 h by using a high shear mixer. Then the mixture was blended at a rotation speed of 2000 rpm for 1.5 h at 150 °C by using a paddle agitator to produce inorganic nanoparticles modified bitumen. The same process was also performed on the base bitumen in order to obtain blank sample.

2.4. High-temperature storage stability test

A toothpaste-shaped aluminum tube with a diameter of 25.4 mm and a length of 140.0 mm was filled with the binders. Then the tube was placed vertically in an oven for 48 h at 163 °C. Once cooled, the tube was cut into three sections, and the top and bottom portions were used as samples. The samples were reheated at 150 °C to conduct the ring-and-ball (R&B) softening point test. The stability of the binders was evaluated by researching the difference in softening point as well as the nanoparticles content between the top and bottom samples. The nanoparticles content in binders was determined by combustion method in muffle at 500 °C.

2.5. Physical properties test

The physical properties of the bitumens, such as penetration (25 °C), softening point and ductility (10 °C, 15 °C), were tested according to ASTM D 5, ASTM D 36, and ASTM D 113, respectively [15–17]. The rotational viscosity of the bitumens at 60 °C and 135 °C was measured by Brookfield viscometer according to ASTM D 4402 [18].

2.6. Ultraviolet aging procedure

UV radiation was utilized to simulate photo oxidation aging of the bitumens. First, the melted bitumens about 30 ± 0.5 g was poured in a Φ140 ± 0.5 mm iron pan, this sample was used to conduct TFOT according to ASTM D 1754 [19]. Then the iron pan containing the residue from the TFOT was placed in UV-aged draft oven for 12 days, and the working temperature was controlled at 60 °C. An UV lamp of 500 W was adopted in the UV-aged draft oven, and the intensity of UV radiation was about 8 w/m^2. The iron pan was placed at the bottom of the chamber. The aged bitumens were evaluated by measuring physical properties such as viscosity (60 °C) and softening point.

2.7. Measurements

A UV–visible spectrometer (UV2550, Shimadzu, Japan) was utilized to measure the absorbance of wavelength range from 200 to 800 nm of three inorganic nanoparticles power. $BaSO_4$ was used as a reflectance standard in the UV–vis diffuse reflectance experiments.

3. Results and discussion

3.1. High-temperature storage properties of inorganic nanoparticles modified bitumen

The uniform dispersion degree of inorganic particles in bitumen is evaluated by high-temperature storage stability test. In order to investigate the compatibility between different inorganic nanoparticles before and after surface modified by γ-(2,3-epoxypropoxy) propyltrimethoxysilane and bitumen, the fixed 2 wt% inorganic nanoparticles content are used in this research. For different inorganic nanoparticles modified bitumen, the difference between the softening points in the upper and lower sections of the toothpaste tubes is shown in Fig. 1, and the corresponding inorganic particles contents in the two sections are shown in Table 2. As shown in Fig. 1, before surface modification, the differences between the softening points (ΔS) of nano-SiO_2, nano-TiO_2 and nano-ZnO modified bitumen are 0.9 °C, 1.2 °C and 1.3 °C, respectively, which indicate that the compatibility between nano-SiO_2 and bitumen is better than that between nano-TiO_2 or nano-ZnO and bitumen. After surface modification, the compatibility between inorganic nanoparticles and bitumen is improved obviously compared with