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Effects of long-term aging on the properties of asphalt binder containing diatoms

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HIGHLIGHTS

- Diatomite modified asphalts were prepared by melt blending with diatomite.
- Honeycomb silica structure in modified asphalt can prevent the oxygen.
- Modified asphalt binder with diatomite has better aging resistance.
- The oxygen dispersion in hydrophobic diatomite modified asphalt was harder.

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ABSTRACT

Diatomite modified asphalts were prepared by melt blending with different contents of diatomite and hydrophobic diatomite. The effects of long-term aging on physical properties and chemical structure of the diatomite modified asphalts were investigated. The results show that the viscosity aging index and the increment in the softening point of the modified asphalts decrease significantly with increasing diatomite contents. The retained penetration increases after the introduction of diatomite. The chemical structures of the aged asphalts were evaluated using FTIR. The increased extent of the C=O and S=O bands areas of the modified asphalt is smaller than that of pure asphalt. The aging resistances of the modified asphalts are much better than that of pure asphalt, because they can efficiently prevent the oxygen, which attribute to the honeycomb silica structure in the modified asphalts. Compared with diatomite modified asphalt, the oxygen dispersion in hydrophobic diatomite modified asphalt was harder, which means the oxidative aging resistance of hydrophobic diatomite modified asphalt was better.

1. Introduction

Asphalts are a complex mixture of high molecular weight hydrocarbon molecules, many of which contain the heteroatom nitrogen, oxygen, sulfur and trace amounts of metals such as vanadium and nickel. Asphalt has been widely used as an adhesive material in many fields, especially in pavement construction [1–3]. The effects of oxidation on physical properties are important because oxidation is the major factor responsible for irreversible asphalt hardening and changes in rheological properties leading to the deterioration of many desirable asphalt performances [4–7]. The change of physical properties and chemical structures is called aging during the construction and the service life in the pavement.

The thin film oven test (TFOT) or the rolling thin film oven test (RTFOT) is used to simulate the thermal oxidative aging of asphalt during construction. The pressure aging vessel (PAV) is designed to simulate the thermal oxidative aging occurred in pavement application [8–10]. The aging properties of asphalts are normally characterized by measuring physical and rheological properties before and after aging in the laboratory. The age hardening is evaluated by observing how viscosity, penetration and softening point change with aging. Many researches on asphalt aging have been reported [11–13]. Most of them focus on the aging behaviors, only a few of them proposed the method for improving aging resistance, but, the effect was not significant. The major functional group types formed on oxidation have been identified as ketones and sulfoxides, with lesser amounts of dicarboxylic anhydrides and carboxylic acids being formed only during the latter stages of oxidation [14,15].

Diatomite is a type of mineral with low cost and abundance, which used in industry as filler, a filtering agent, an absorbent, a clarifier, and an insulator. The diatomite is a sedimentary rock,

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white to light yellow in color, composed of the fossilized skeletons of diatoms, one celled algae-like plants which accumulates in marine or lacustrine environments [16]. The skeletons are composed of amorphous silica (silicon dioxide, SiO_2), a very durable substance. Besides its amorphous silica content diatomite rocks commonly contain carbonate and clay minerals, quartz and feldspars. Diatoms skeletons are honeycomb silica structures that give diatomite useful characteristics such as high absorptive capacity and surface area, chemical stability, and low bulk density [17]. It is also called diatomaceous earth or organogenetic sedimentary rock. Diatomite was often used as the filler of polymer due to its lightweight, high void content, low density and strong sorption performance [18]. However, there have been few reports about the preparation of the modified asphalt with diatomite till now, especially the effect of diatomite on asphalt binder aging.

In this paper, the effects of long-term aging on physical properties of the diatomite modified asphalts were investigated, and the change of the chemical structure of diatomite modified asphalts before and after long-term aging was evaluated using FTIR spectroscopy.

2. Experimental

2.1. Materials

The asphalt type of AH-70 used was obtained from Panjin Petrochemical Industry, Liaoning Province of China, with penetration of 72 (0.1 mm at 25 °C, 100 g and 5 s), softening point of 45.9 °C, ductility of at least 150 cm (at 15 °C) and viscosity of 0.45 Pa·s (at 135 °C).

Diatomite was obtained from Shenzhen Jitong diatomite Co. Ltd., with bulk density 0.29 g/cm^3, maximum particle size 19 μm, PH 7.2.

Silane coupler KH-570 was obtained from Guanzhou Ouying Co. Ltd., with chromatography purity 99.4%, molecular weight 236.4 g/mol and chemical structure $CH_2{=}C(CH_3)COOCH_2CH_2CH_2Si(OCH_3)_3$.

2.2. Preparation of hydrophobic diatomite

The diatomite was calcined for 4 h at 450 °C in a muffle furnace, and let it cool to room temperature. A certain amount of calcined diatomite was added into the mixed solution of water and ethanol (1:1 vol ration), the acetic acid was employed to adjust the PH 4. Then the mixture was stirred 30 min at 70 °C and the silane couple KH-570 was added slowly to mixture and stir 2 h. Finally, the hydrophobic diatomite was obtained after the solvent recovery.

2.3. Preparation of diatomite modified asphalts

The modified asphalts were prepared using a high shear mixer. Asphalt was first heated until it becomes sufficiently fluid at around 150 °C in the mixer for 5 min. After that, diatomite or hydrophobic diatomite was added into asphalts, and the mixtures were blended at 3000 r/min rotation speed about 120 min to ensure the well dispersion of diatomite or hydrophobic diatomite in the asphalt binders.

2.4. Long-term aging processes

Firstly, the 35 g asphalt sample is placed in a glass bottle, which has a narrow top opening. The glass containers are placed in a carriage such that the axis of revolution is horizontal and the container opening is facing a jet of air. The oven is kept at 163 °C and the carriage is rotated in the oven at a rate of 15 rpm for 85 min. As soon as the time of RTFOT of asphalt was over, 50 ± 0.5 g of each asphalt sample was immediately poured into two marked pans and placed together in the same PAV for a single test run. When the temperature inside the PAV was within ± 2 °C of the aging temperature (109 °C), an air pressure of 2.1 ± 0.1 MPa was applied and maintained for 20 h ± 10 min. After 20 h, the pans were removed and poured into different containers for different physical and chemical tests.

2.5. Physical properties test

The physical properties of pure asphalt and diatomite modified asphalts, including softening point, penetration (25 °C) and ductility (5 °C and 15 °C), were tested according to ASTM D36, ASTM D5 and ASTM D113-86, respectively.

Brookfield viscometer (Model DV-II+, Brookfield Engineering Inc., USA) was employed to measure the viscosity of the modified asphalts according to ASTM D4402. Approximately 30 g of asphalt are heated in an oven so that it is sufficiently fluid to pour into the sample chamber. The amounts of asphalt used vary with the different sizes of the spindles. The sample chamber containing the asphalt sample is then placed in the thermo container. After the desired temperature is stabilized for about 30 min, the spindle is lowered into the chamber to test the viscosity.

2.6. Fourier Transform Infrared (FTIR) analysis

The infrared spectra were recorded with a Perkin Elmer Paragon-400 spectrometer. Asphalt samples, dissolved in dichloromethane (30 g/l) are laid on a potassium bromide (KBr) thin plate. The solvent is evaporated under a nitrogen flow to avoid interference in the obtained spectra. The band areas are measured from valley to valley [2].

3. Results and discussion

3.1. Hydrophobic diatomite properties

Fig. 1 showed the FTIR spectra analysis curves of diatomite and hydrophobic diatomite. The results showed that the strong peaks of 470 cm^{-1} are the antisymmetric bending vibration absorption bands of the O—Si—O in $[SiO_4]^{4-}$. The characteristic absorption peak of 801 cm^{-1} and 1096 cm^{-1} is attributed to Si—O stretching vibrations in $[SiO_4]^{4-}$. The O—H bending vibration is observed at 1632 cm^{-1} and the stretching vibration absorption peak of Si—O—H and O—H is observed at 3422 cm^{-1}. The results indicated that the diatomite is the amorphous oxygen-silicon tetrahedron structure and there is a little water on the face of diatomite. The positions of absorption bands in the curves of hydrophobic diatomite are likely the diatomite. The only difference is the stretching vibration absorption bands of the alkyl (C—H) appear at 2928 cm^{-1} and 2856 cm^{-1}. Besides this, no other new peak appears in the infrared absorption spectrum of hydrophobic diatomite. Therefore, it can be confirmed from the analysis of the infrared absorption spectrum that the chemisorption is occurred between silane couple KH-570 and diatomite. The characteristic absorption peak of C=O is not appear in the infrared absorption spectrum. It maybe attribute to the content of silane couple KH-570 in hydrophobic diatomite is less and the absorption peak of C=O has not appeared.

3.2. Hydrophobic surface modification mechanism

Silane couple KH-570 reacts with water (hydrolysis) to form silanol groups, and oligomers are formed through partial condensation. Then the silanol oligomers react with hydrogen bond to the surface of the diatomite. Finally, the diatomite put through a drying process and robust chemical bonds are formed through a dehydration condensation reaction. The Mechanisms of hydrophobic

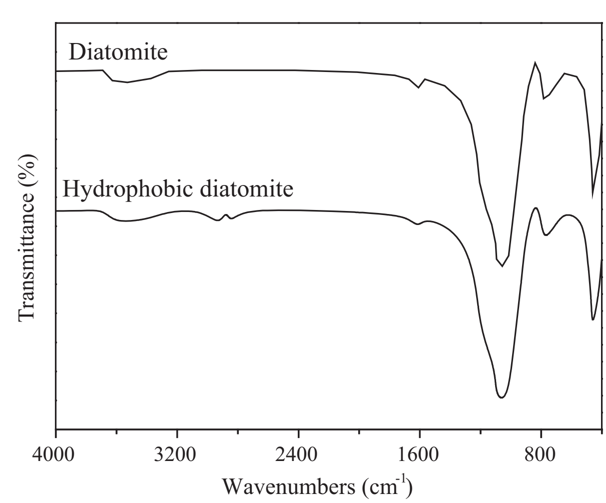

Fig. 1. FTIR spectra of diatomite and hydrophobic diatomite.