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Investigation of the graphene oxide and asphalt interaction and its effect on asphalt pavement performance

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HIGHLIGHTS

- Graphene oxide (GO) was used to modify two kinds of asphalt binders.
- No chemical reaction between GO and asphalt, the CO₂ was produced by the decomposition of GO.
- Lamella structure of GO was completely stripped, and scattered to a single layer in asphalt binders.
- GO modified asphalt binders have better performance at both low and high temperatures.
- GO could improve the anti-fatigue performance of the asphalt.

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ABSTRACT

Previous studies have shown that graphene oxide (GO) could improve the mechanical and rheological properties of polymers. However, the GO-asphalt interaction and the effects of GO on asphalt pavement performance are still not clear. In this paper, the GO modified asphalt binders were prepared by melt blending method, the GO-asphalt interaction was investigated by the Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and gas chromatography-mass spectrometer (GC-MS). The GO effects on the pavement performance of asphalt binders were also studied. In addition, the thermal properties of GO modified asphalt were studied by thermo gravimetric (TG) analysis. The results show that the gas released from GO modified asphalt is the CO₂. There is no chemical reaction between the GO and asphalt binders, as much as the CO₂ is produced by the decomposition of GO. After being mixed with asphalt binders, the lamella structure of GO is completely stripped, and scattered to a single layer in asphalt binders. The GO has many advantages in improving the pavement performance.

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1. Introduction

Graphene oxide (GO), is a precursor for grapheme, which has atomic thickness and two dimensional sizes in the tens of micrometer range or larger [1,2]. Its molecular structure is roughly the same as the grapheme, and also has the excellent gas and liquid blocking performance [3,4] and certain conductivity [5,6]. Currently, GO, for its excellent structure and functional properties, is widely used in many areas such as gas sensors, carbon-based electronics, impermeable membrane and polymeric composite materials. For instance, Yoo B M [7] demonstrated that GO layered structures could maximize the gas-diffusion path length and, as a result, significantly decrease the gas flux through layered composite films. Tang Z [8] found that the modulus and tensile strength of

the butadiene-styrene-vinyl pyridine rubber could be significantly improved by utilization of GO. Jeffrey T. P [9] and Bai H [10] demonstrated that GO had a extremely large specific surface area, high modulus, in conjunction with water solubility and versatile surface chemistry, so it could be used as the reinforcing additives for various polymers. The research work conducted by Yongjin [11] showed that GO could improve the conductivity, thermal, mechanical, and rheological properties of poly (methylmethacrylate). What's more, Wenbo Zeng [12] and Wu S [13] used the GO as anti-aging modifier for asphalt, the results showed that GO improved the anti thermo-oxidative aging and anti UV aging performance of asphalt binders.

Other researchers have indicated the effects of aging of asphalt binder on the performance of the flexible pavement [14,15]. For the special molecular structure, it is possible for GO to improve the anti-aging performance of asphalt by several ways: a) GO can prevent the contact of oxygen with the asphalt, and reduce the reac-

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tant concentration (O₂) of the oxidation reaction in the aging process and b) GO can retard the volatilization of light components in asphalt due to its excellent blocking performance. Previous studies have shown that GO can improve the anti-aging performance of asphalt [12,13], but how the GO interacts with asphalt is not known in great detail. The Young's modulus and strength of graphene are both intrinsically high, but it is strongly influenced by the van der Waals force [16], that may cause the GO be more difficult to disperse in the polymer evenly and weaken its improvement effect sharply. In order to maximize the improvement effect of GO on asphalt, it is important to study the interfacial interaction between asphalt and GO accurately. In this paper, two types of asphalt binders (base asphalt of 80/100 penetration grade and SBS modified asphalt) and two levels of GO contents (1% and 3% by weight of asphalt binder) are designed to prepare the GO modified asphalt (GO MA) by melt blending method. First, the Fourier transform infrared spectroscopy analysis (FTIR), X-ray diffraction analysis (XRD) and gas chromatography-mass spectrometer (GC-MS) were used to study the interaction between GO with asphalt. In addition, the thermo gravimetric (TG) analysis was performed to study the thermal properties of GO and its effect on the thermal properties of asphalt binders.

The main purpose of the previous studies on GO was the investigation of the anti-aging performance of the asphalt [12,13]. However, whether GO will significantly improve or damage the pavement performance of asphalt before aging is still not clear, therefore, it is necessary to study the pavement performance of GO MA. The GO effect on the physical performance of asphalt binders were studied, in this research project, by the penetration test, softening point test, ductility test and viscosity test; then the rheological properties of asphalt were tested by the dynamic shearing rheometer (DSR) at a temperature range from –10 °C to 80 °C; finally, the bending beam rheometer (BBR) was used to study the low temperature (–12 °C and –18 °C) of rheological properties of the asphalt binders.

2. Material and experiments

2.1. Materials

2.1.1. Asphalt binders

Two types of asphalt binders were used in this research, namely base asphalt with 80/100 penetration grade (simply refer to as 90A) and styrene–butadiene–styrene modified asphalt (simply refer to as SBS MA). They were obtained from Inner Mongolia Xindalu Asphalt CO., LTD (Inner Mongolia, China). Technical information of these two asphalts are shown in Table 1.

2.1.2. Graphene oxide

The GO with 5 to 10 layers was provided by the Suzhou Heng Ball Graphene Technology CO., LTD (Suzhou, China). Purity of GO was higher than 95% with a specific surface area of about 100–

300 m²/g, the structure crystallite and the lamella diameter of the GO were 43.2 and about 10 μm to 50 μm, respectively.

2.2. Characterization of GO

2.2.1. Fourier transform infrared spectroscopy test

Fourier transform infrared spectroscopy (FTIR) was used to test the characteristic functional group changes of the asphalt binders before and after being mixed with GO, the preparation procedures of asphalt sample for FTIR are described briefly next. First, the asphalt CS₂ solution with 5 wt% concentration of asphalt was prepared, then 2 drops of asphalt CS₂ solution were placed on the KBr chip by the glue dropper, so the thin film asphalt sample could be obtained after the CS₂ being fully volatilized. The scan wave number range was from 4000 cm⁻¹ to 400 cm⁻¹, scan times were 64 times.

2.2.2. X-ray diffraction test

The layer-to-layer distances (d) of the pure GO and the GO in GO MA were conducted by the Smart Lab X-ray diffraction (XRD) at scanning rate of 0.25°/s, the 2θ range was from 5° to 40°, the wavelength of the CuKα ray was 0.1506 nm. The GO samples used for XRD were prepared by the positive pressure method; the GO MA samples were prepared by hot casting method, GO modified 90A (GO/90A) and GO modified SBS MA (GO/SBS MA) were first heated to 155 °C and 170 °C, respectively, then placed in an aluminum foil (4.0 cm × 4.0 cm × 2.0 mm), ensured that the thickness of the asphalt film was about 1.5 mm, and set the dish in the oven at designed (155 °C or 170 °C) temperature for 30 min to make sure the asphalt membrane was flat. The interlayer distance of the GO was calculated according to the Eq. (1).

$$d = \frac{\lambda}{2\sin\theta} \quad (1)$$

where, d is the interlayer distance of the GO in nm; λ is the wavelength of the CuKα ray in nm; 2θ is the diffraction angle in radian.

2.2.3. Thermo gravimetric (TG) test

Thermo gravimetric (TG) analysis of the GO and GO MA were performed on a ZCT-B thermal analyzer at a heating rate of 10 °C/min and under air atmosphere, the test temperature was from room temperature to 700 °C and the mass of every sample was 15 mg.

2.3. Preparation of GO MA

First, 90A was heated to 155 °C (or SBS MA was heated to 170 °C); then the design contents of the GO (1.0 wt% or 3.0 wt%) were added to the asphalt binder by using a high speed shear mixing machine (4000 r/min) for 30 min. Because the heating of the preparation process inevitably causes the aging of the asphalt binder, and it is rational to evaluate the aging resistance performance of GO modified asphalt binders at the same condition. The virgin

Table 1
Technical information of 90A and SBS MA.

Asphalt	Technology parameters	Unit	Test results	Method
90A	25 °C Penetration	0.1 mm	85	ASTM D5 [17]
	Softening point	°C	44.1	ASTM D36 [18]
	10 °C ductility	cm	>100	ASTM D113 [19]
	60 °C viscosity	Pa.s	206	ASTM D4402 [20]
SBS MA	25 °C Penetration	0.1 mm	69	ASTM D5
	Softening point	°C	56.0	ASTM D36
	5 °C ductility	cm	56	ASTM D113
	135 °C viscosity	Pa.s	1.337	ASTM D4402

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