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Research Article

Impacts of Graphene Oxide on the Physical Property and Microstructure of Asphalt Material

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In order to determine the effect of graphene oxide (GO) on the physical property and microstructure of the asphalt binder, GO material was selected to prepare GO-modified asphalt for analysis. Firstly, the surface/interface characterization and chemical analysis were performed on GO and GO-modified asphalt. The physical properties of GO-modified asphalt were investigated through softening point, ductility, and penetration tests. The impact of GO material on the rheological behavior of asphalt was characterized by Brookfield viscosity and dynamic shear rheological tests (DSR). Furthermore, the typical microstructure of GO inside asphalt material was also identified and analyzed through a scanning electron microscope (SEM) and transmission electron microscope (TEM). The results showed that the modifying effect of GO materials could promote the fusion between asphalt and GO. A modified GO could improve the high temperature of asphalt, but it would degrade the low temperature property of the asphalt binder. A modified GO could be integrated with asphalt and achieve a modifying effect on the asphalt material. This study would provide references for the application of graphene oxide in asphalt pavement materials.

1. Introduction

Because of its excellent structure and functional properties, graphene oxide (GO) is widely applied in many areas, such as gas sensors, carbon-based electronics, impermeable membranes, and polymeric composite materials Li, Wu, and Amirkhanian, cited in [1–6]. Regarding its perfect property and chemical stability, researchers began focusing on the studies of GO used in asphalt material as a modifier to improve the physical property of asphalt. For instance, Habib et al. used GO material as a bitumen modifier and studied the improving effects on the properties of asphalt. They investigated the interaction between GO and asphalt binder, and its impact on asphalt pavement performance was confirmed by Li, Wu, and Amirkhanian 2018. They evaluated and analyzed the performance and modifying mechanisms of asphalt binders modified by different types of GO materials Liu, Zhang, and Shi [7–10]. Moreno–Navarro et al. evaluated the mechanical and thermal properties of GO-modified asphalt binders. Wu et al. investigated the aging resistance of GO-modified asphalt. Yao et al. studied the rheological properties, low-temperature cracking resistance, and optical performance of exfoliated graphite nanoplatelets in modified asphalt binders. Zeng et al. examined the utilization feasibility of GO in asphalt. All these studies indicated the potential for utilizing GO in asphalt materials. The effect of GO on the physical property was key to achieving its application inside asphalt, and the impact of the asphalt microstructure was the source of improving the effect of GO.

Regarding the research status, the characterization of impacts on GO on-road performance and the microstructure of asphalt material would be conducted in this study, which could provide references for the GO applied pavement materials. Firstly, the GO material was modified through different techniques, and the surface-modifying effect was evaluated on the microscale. The GO-modified asphalt was prepared and the physical and rheological properties were investigated to explore the modifying effect of GO on asphalt. Furthermore, the typical microstructures led by GO were also identified and analyzed through the scanning electron microscope. This study would provide a reference for the application of GO materials to asphalt pavement material.

2. Material and Preparation

2.1. Material

2.1.1. Graphene Oxide. The GO used in this work was TNRGO graphene (Figure 1) provided by the Chengdu Organic Chemicals Co. Ltd. in China. The technical indices of selected GO materials are shown in Table 1.

2.1.2. Asphalt. In order to investigate the impact of GO on the performance of asphalt, Shell AH-90 asphalt was selected to prepare GO-modified asphalt. The technical properties of AH-90 are shown in Table 2.

2.1.3. Graphene Modifier. Octadecylamine and tetramethylthiuram disulfide were selected as modifiers to improve the surface property of GO material and promote the fuse state between asphalt and GO. The technical properties are shown in Table 3.

2.1.4. Additives. N-methyl-2-pyrrolidone was chosen as the accelerant to promote the dispersibility of GO inside asphalt. The technical capacities of N-methyl-2-pyrrolidone are shown in Table 4.

2.2. Preparation. The preparation process for GO-modified asphalt is given as follows:

a. Perform the modification of graphene oxide: Octadecylamine and tetramethylthiuram disulfide were dissolved into distilled water to prepare the modifying solution. GO was mixed with the modifying solution and kept for 16 h. Then, the modified GO was filtered into a clean container through filter paper. b. Dry and mill graphene oxide: the modified GO was dried at a low temperature (at −10°C) by freeze-drying for 12 h. Then GO was processed by high-energy ball milling in ethanol under inert gas protection. c. Premix graphene oxide and dispersant: GO and dispersant were moved into a mixing machine and premixed for 30 min at a mixing speed of 200 rad/min. [11–15] d. Heat asphalt and add graphene oxide and dispersant: [16] AH-90 asphalt was heated at 160°C, and the mixture consisting of GO and dispersant was addedto the heated asphalt. Then, high-speed shearing was performed on the mixture at 3000 rad/min for 8 min and then 1500 rad/min for 5 min. After that, the preparation of GO-modified asphalt was completed.

3. Methodology

Different testing methods were used in this study to perform an impact analysis of GO on the asphalt binder. The detailed information and instruments of the main tests in this work are shown in Figure 2 and Table 5, respectively.

4. Results and Discussions

4.1. Characterization of Graphene Oxide

4.1.1. Morphology Characterization. The modification was vital for GO material applied inside asphalt. Excellent modification of GO materials would enhance the improving effect of GO on the properties of asphalt. Therefore, the SEM analysis was performed on the unmodified and modified specimens of GO to evaluate the impact of the modification on the microstructure of GO. The SEM images of the optimized specimen and the control specimen are shown in Figure 3. It could be observed that the unoptimized GO was gathered and accumulated, which indicated that the dispersion of GO powder was poor and the GO powder was clustered. After specific modification and freeze-drying, the micrograph showed excellent microstructure and the perfect dispersion of GO.

In order to characterize the lamellar structure of a modified GO material, the TEM analysis was conducted on a modified GO, and the result is detailed in Figure 4. From Figure 4, the lamellar structure of GO is clear and obvious. Furthermore, there were no significant defects on the surface of GO, suggesting the modified GO had an excellent microstructure that could guarantee the applied effect inside asphalt.

4.1.2. Chemical Composition Analysis. Besides microstructure characterization, the chemical composition was investigated to guarantee the purity of GO. The results of the chemical composition analysis are demonstrated in Figure 5 and Table 6.

It could be analyzed from Figure 5 and Table 6 that C and O were the main chemical elements of GO and that no impurities existed inside the chemical composition of GO. The XPS results also confirmed the high purity of GO. Therefore, the GO materials of high purity could provide excellent potential for the modification of asphalt materials.

4.2. Impact of Graphene Oxide on Performance of Asphalt

4.2.1. Effects on Physical Property. Aiming to investigate the effect of GO on asphalt, the penetration, softening point, and ductility of modified asphalts with different contents of GO were tested in this section. The test results are shown in Figure 6.

Based on Figure 6, the addition of GO could promote the increase of the softening point of asphalt, which meant that GO could lead to a positive effect on the high-temperature performance of asphalt. However, the low-temperature property of asphalt was degraded following the increase of

Figure 1: Selected graphene oxide.

Table 1: Technical indices of selected GO.

Technical indices	Purity wt (%)	Layers	Thickness (nm)	Diameter (μm)	Specific surface area (m^2/g)
Graphene oxide	99	<10	0.55–3.74	0.5–3	500–1000

Table 2: Technical properties of AH-90 asphalt.

Performance indices	AH-90	Requirements
25°C penetration (0.1 mm)	89.2	80–100 (AH-90), 60–80 (SBS)
Softening point (°C)	49.3	≥45 (AH-90), ≥55 (SBS)
10°C ductility (cm)	37	≥30 (AH-90)
Solubility (%)	99.7	≥99.5 (AH-90), ≥99 (SBS)

Table 3: Technical properties of graphene oxide.

Technical indices	Density (g/cm^3)	Molecular weight	Boiling point (°C)	Molecular structure
Octadecylamine	0.82	269.51	232/32 mmHg	
Tetramethyl thiuram disulfide	1.34	240.43	307/760 mmHg	

Table 4: Technical capacities of N-methyl-2-pyrrolidone.

Technical indices	Molecular weight	Relative density	Viscosity (mPa s)	Refractive rate (%)
Graphene oxide	99.13	1.028	1.65	1.468

GO content. When the GO content increased to 15%, the growth rate of softening point and decrease rate of ductility reached 9.7% and 16.2%, respectively.

4.2.2. Impacts on Rheological Behavior

A. Brookfield Viscosity. Brookfield viscosity was selected to analyze the viscosity variation of asphalt following the increasing content of GO. The results of Brookfield viscosity are shown in Figure 7.

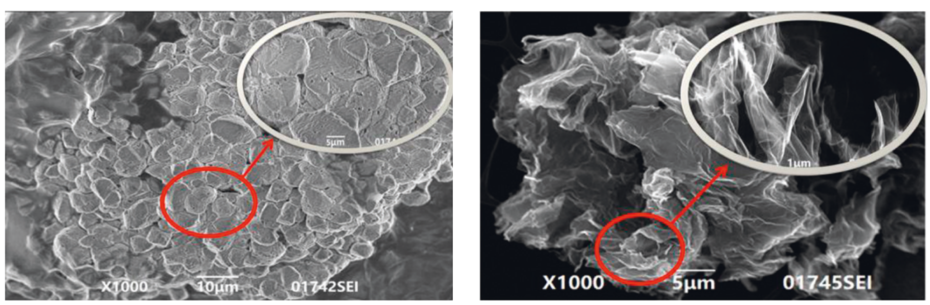
The Brookfield viscosities of different GO-modified asphalts decreased when the temperature increased. 75°C and 90°C were the turning points of the variation curve. When GO was mixed into asphalt, the Brookfield viscosity of asphalt increased obviously, which indicated that GO could lead to the rise of asphalt viscosity. When the content of GO increased to 15%, the growth rates of Brookfield viscosity

(a) (b) (c) (d)

Figure 2: Main test instruments for this work. (a) SEM, (b) TEM, (c) DSR, and (d) XPS.

Table 5: Detail information of methodology.

Test	Method	Condition
Microstructure	Scanning electron microscope (SEM) Transmission electron microscope (TEM)	20.0 kV, 1 μm, 3000 and 5000 times
Chemical composition	X-ray photoelectron spectroscopy (XPS)	Binding energy range 1100–0.00 eV 180 ms/0.60 eV, down scan for 2 times
Basic performance	Penetration, softening point and ductility	According to ASTM D36, ASTM D5 and ASTM D113.
Rheological behavior	Brookfield viscosity (BV) Dynamic shear rheology (DSR)	Load frequency $\omega = 10$ rad/s temperature range 80~160 (BV) 60°C-90°C (DSR) heating rate 2°C/min



(a) (b)

Figure 3: SEM images of GO before and after optimization. (a) Optimized (b) Unoptimized.

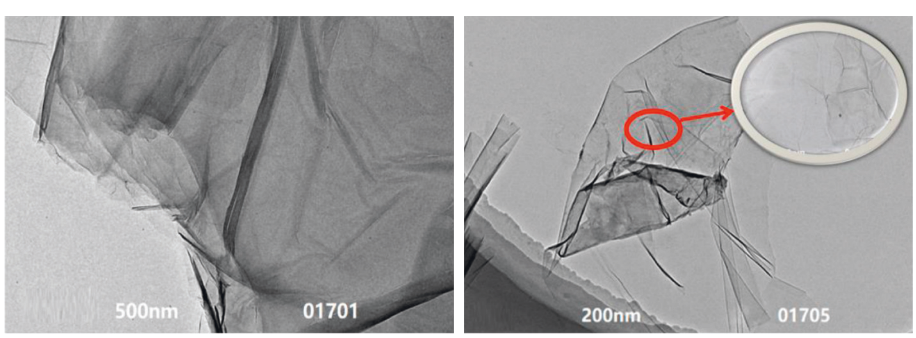


Figure 4: TEM images of modified GO.

could reach 3.3%, 7.9%, 56.4%, 77.8%, and 79.3% at 60°C, 75°C, 90°C, 105°C, and 120°C, respectively. It could be concluded that GO could enhance the viscosity of asphalt at high temperatures, which provided the reference for the improving effect of GO on softening points of the asphalt binder.

B. DynamicSshear Rheology. The complex modulus, phase angle, and G/sin δ were obtained through DSR tests. The DSR results are shown in Figure 8. It could be indicated that GO could cause changes in the rheological behavior of asphalt. These changes are mainly reflected in the evaluating indices of complex modulus, phase angle, and rutting factor

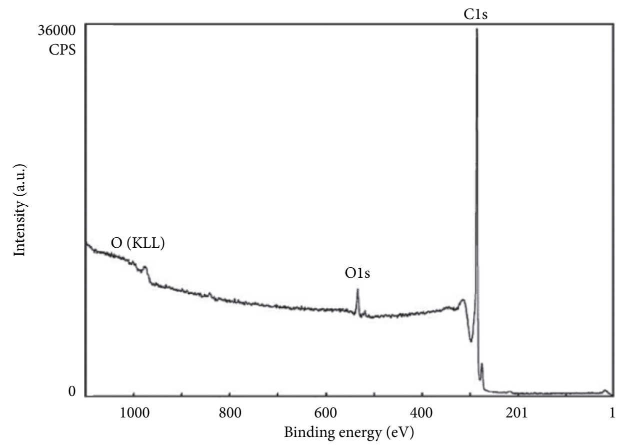


Figure 5: XPS results of GO.

Table 6: Chemical composition of selected GO.

Elements	Area (CPS)	Contents (%)
C	37995.3	96.41
O	3736.8	3.59

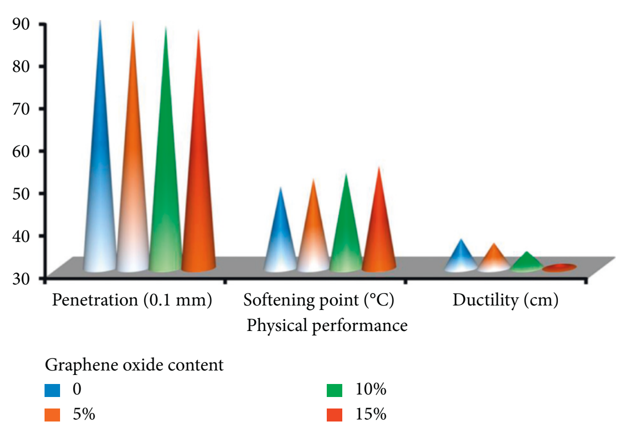


Figure 6: Physical performances of GO-modified asphalt.

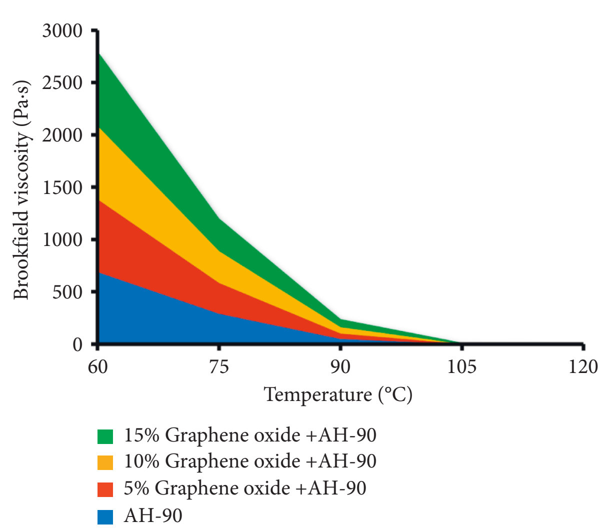


Figure 7: Brookfield viscosity of GO-modified asphalt.

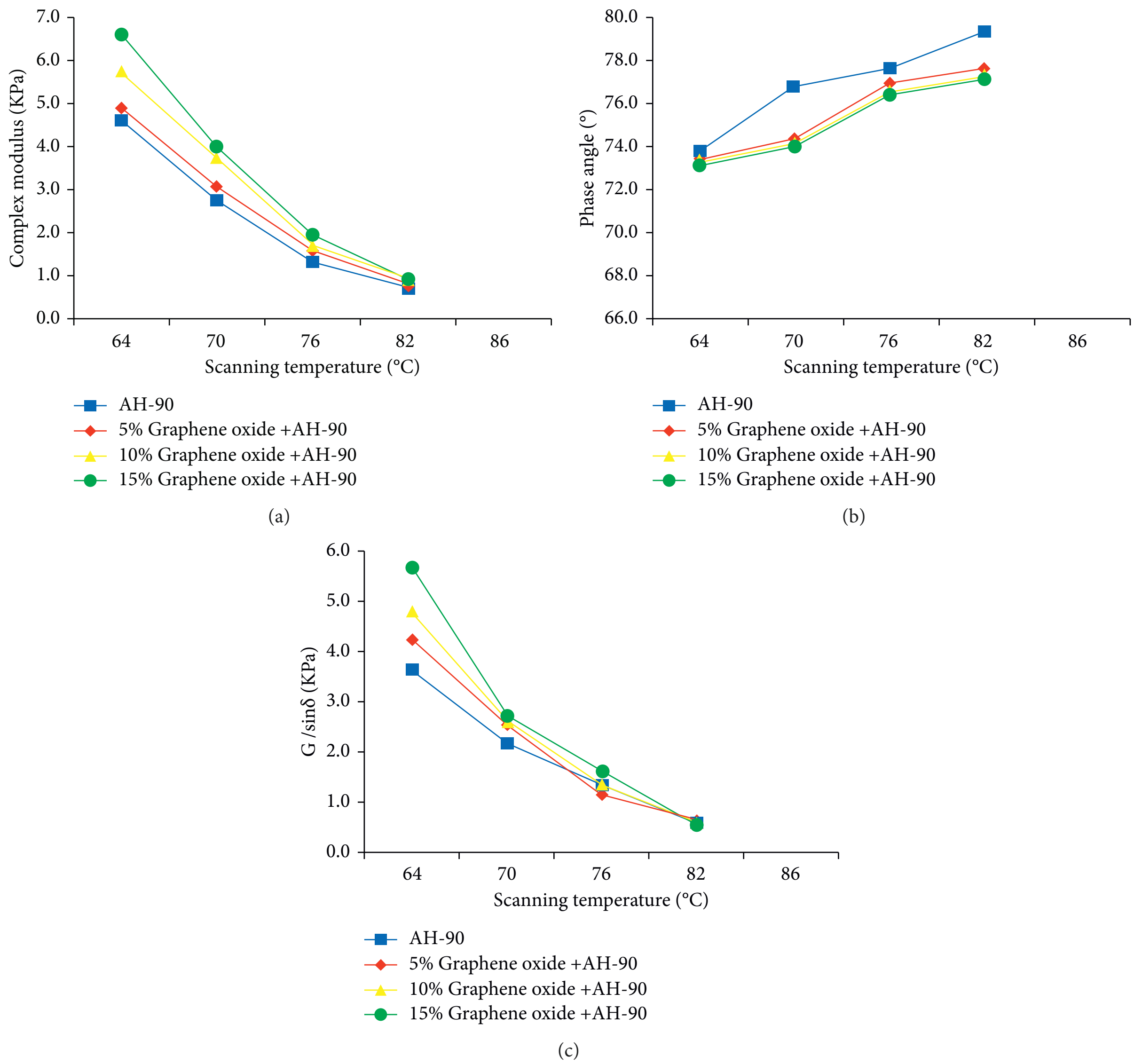
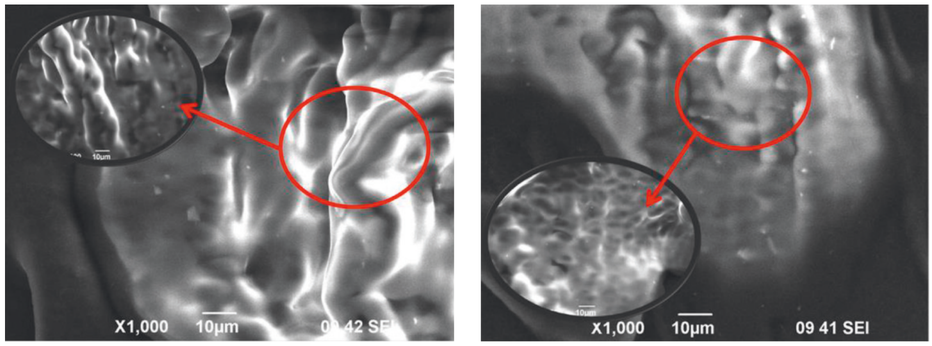


Figure 8: DSR results of GO-modified asphalt. (a) Complex modulus, (b) Phase angle, and (c) G/sinδ.

(G/sinδ). The complex modulus of GO-modified asphalt was increased by 31.9% and 24.5% when temperatures rose to 76°C and 82°C, respectively. The rutting factor of GO-modified asphalt followed the same varying law approximately, whereas the phase angle was decreased remarkably compared to AH-90# asphalt. These varying rules revealed that GO could enhance the thermal stability of asphalt when subjected to a high-temperature environment.

4.3. Modifying the Effect of GO on Microstructure. The morphology of GO-modified asphalt was characterized in this section. The magnified times were 1000 and 5000, and the typical microstructures of asphalt modified by GO are identified and demonstrated in Figure 9. In the SEM image of AH-90 asphalt (Figure 10), there was no apparent microstructure on the surface of the asphalt and the sample surface was flat. According to SEM images, the two types of typical structures of GO-modified asphalt were identified. Figure 9(a) showed pleated asphalt monomer in microscopic appearance. In the magnified image, the pleated asphalt monomers were connected mutually and remarkably stable. Clustered state of the asphalt monomer as shown in Figure 9(b) and obvious asphalt particles gathered closely. These two connected and gathered microstructures might be promoted by GO, and these types of stable microstructures might lay a solid foundation for improving the effect of GO on high-temperature and rheological performances.

It can be seen from Figure 11 that the C element content of GO-modified asphalt was increased significantly, which reflected the modifying effect of GO on the chemical composition of AH-90 asphalt. Furthermore, the addition of GO would also increase the relevant content of other elements, such as Na, Ca, and Si. The modifying effect of GO on the chemical composition of asphalt might be related to the improving effect on the physical performance of asphalt.



(a) (b)

Figure 9: SEM images of GO-modified asphalt. (a) Typical microstructure 1. (b) Typical microstructure 2.

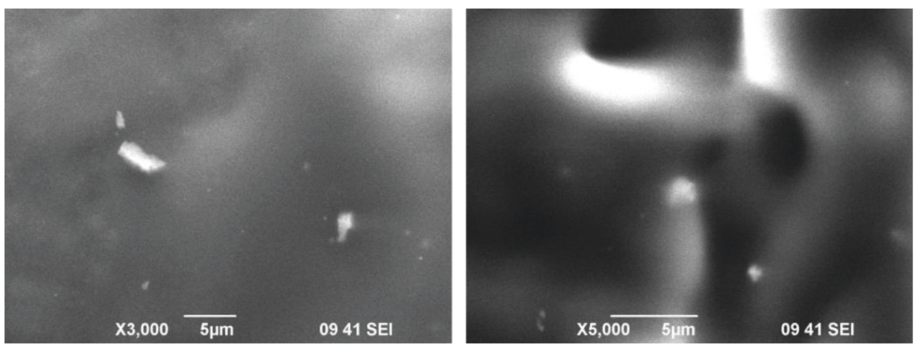
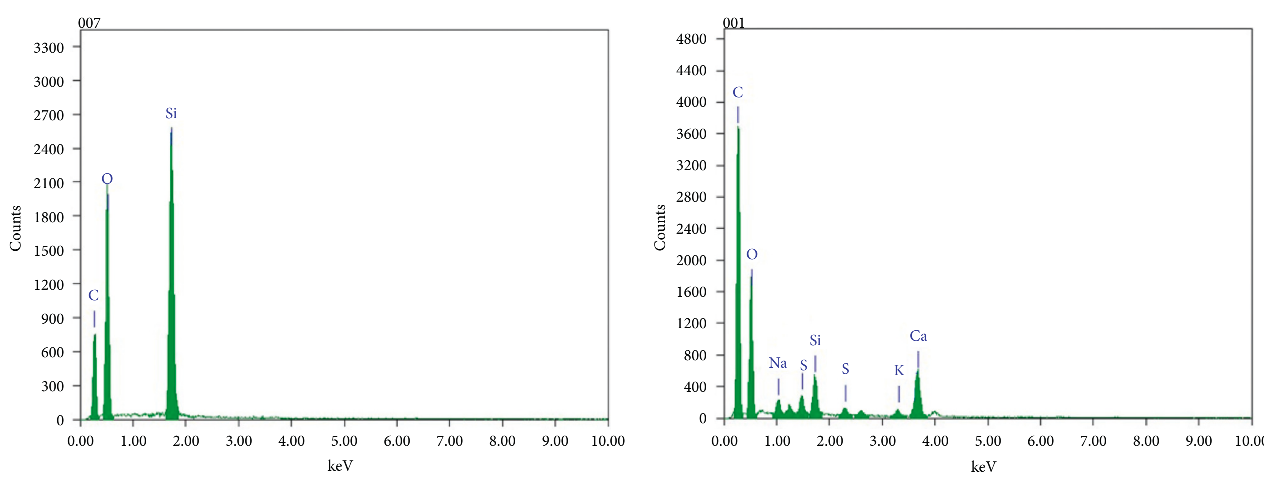


Figure 10: Asphalt without GO.



(a) (b)

Figure 11: EDS analysis results. (a) AH-90; (b) GO-modified asphalt.

5. Conclusions

The conclusions are given as follows:

(i) GO could lead to a positive effect on the high-temperature performance of asphalt. However, the low-temperature property of asphalt was degraded following the increase in GO content.

(ii) GO could enhance the viscosity of asphalt at high-temperature, which provided the reference for the improving effect on softening point.

(iii) GO could enhance the fusion between asphalt and GO, which might provide evidence for a thermal stability improvement of GO-modified asphalt.

(iv) The connected and gathered microstructures of asphalt molecules might be promoted by GO, which lays a solid foundation for improving the effect of GO on high-temperature and rheological performances.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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