

Research Article

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Micro-structure and rheological properties of graphene oxide rubber asphalt

<https://doi.org/10.1515/ntrev-2019-0021>

Received May 16, 2019; accepted Jun 07, 2019

Abstract: Compared with traditional materials, nanomaterial has rich and excellent performance due to the size advantages. If nanotechnology is combined with pavement materials, it is foreseeable that it will have a profound impact on the development of the road. In the study, graphene oxide nanomaterial was added as modifiers to rubber asphalt and its microstructure was characterized. The rheological properties of graphene oxide rubber asphalt and the road performance of asphalt mixture were studied in accordance with the SHRP test. The results showed that graphene oxide could form a dense and stable microstructure with rubber powder and asphalt polymer. As a modifier, it could improve the anti-rutting performance of rubber modified asphalt and was more suitable for heavy traffic. Moreover, it had low sensitivity to temperature and was more suitable for areas with large temperature difference between day and night. The pavement performance of the graphene oxide rubber asphalt mixture had also been greatly improved.

Keywords: Graphene oxide rubber Asphalt; Micro-structure; Rheological properties; Pavement performance

1 Introduction

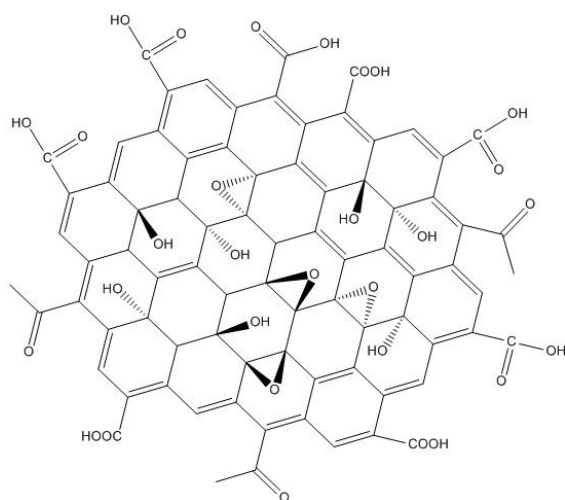
At present, asphalt pavement materials are mainly SBS modified asphalt. Although they have excellent high and

low temperature performance, the pavement using SBS modified asphalt still has some problems. For example, under heavy traffic and severe climatic conditions, their road performance had not been further improved, and due to a single modification method, it was prone to early disease, aging, performance degradation, and could not meet special road sections [1, 4]. Compared with waste rubber, SBS modifier is not only expensive but also be poor in anti-aging properties. However, there are so many problems in traditional rubber asphalt such as unstable quality, sulfur-containing pollution, large odor, high temperature viscosity, and low temperature crack resistance [5, 6]. Its pavement performance has not been able to exceed SBS modified asphalt. Therefore, it is particularly important to modify the traditional rubber asphalt to add polymer technology. Among the many modifiers, nano-modifiers have shown great potential in meeting the needs of the pavement industry. Many studies had analyzed the performance of nanomaterial modified asphalt [7–10]. Asphalt material is composed of many organic molecules, its chemical composition is extremely complex and can react with various types of modifiers. Therefore, it was necessary to study the modification effect of different types of nanomaterials such as nano-silica powder [11], carbon nanotubes [12, 15], carbon nano-fibers [16, 17], polysiloxane-modified montmorillonite [18, 19] as well as non-modified and polymer modified Nano-clay [20, 21] used on asphalt. W.J. elaborated that more than a dozen nanomaterials could contribute to the performance optimization of pavement materials at the SATC meeting [22]. Jamshidi [23] discussed the rate of change that takes place in the rheological properties of asphalt binders modified with numerous types and contents of nano-materials. Through domestic and foreign research, graphene has a certain degree of research and application in the field of composite materials and intelligent transportation [24–27] due to its own mechanical properties [28], electrical properties [29, 31], thermodynamic performance [32] and other advantages [33]. Although graphene exhibits various excellent physical properties and low production costs, it is prone to agglomeration. A rich oxygen-containing group is attached to the base surface and the edge of the graphene

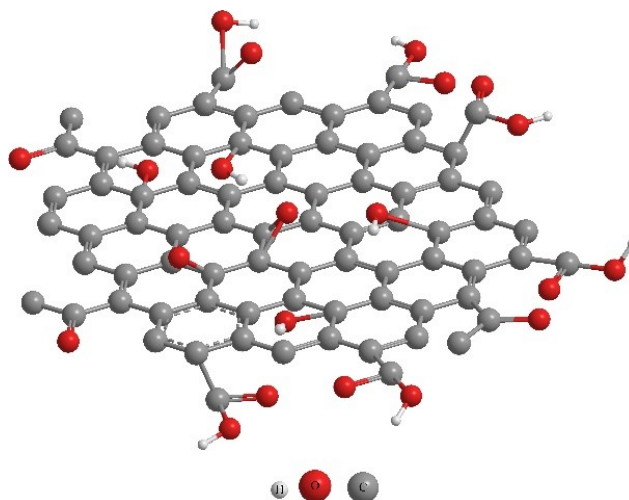
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(a) Graphene oxide (2D)



(b) Graphene oxide (3D)

Figure 1: The structure model of GO**Table 1:** Technical indicators of SK-90 asphalt

Name of the pilot project		Test Results	requirements
Penetration (25°C, 100g, 5s) /0.1mm		87	80-100
Softening Point/°C		46.5	≥ 44
Ductility (15°C) /cm		115	≥ 100
RTFOT aging	Quality change/%	-0.12	-0.4~+0.4
	Residual penetration (25°C) /%	72	≥ 57
	Residual Dilution (10°C) /cm	21	≥ 8

oxide (GO) structural unit, and these oxygen-containing groups mainly include -COOH, -OH, C=O, and epoxy-O-, etc [34]. These oxygen-containing groups provide a large number of active sites for the functionalization of the surface of GO [35, 36], shown as Figure 1. At present, only few scholars had studied the addition of GO to modify asphalt and asphalt mixtures [37–39]. However, the microscopic properties of GO and its macroscopic mechanical properties were not combined. In this study, GO was added as a modifier to rubber-modified asphalt, and the microstructure changes of the samples were scanned and analyzed. Then, the rheological properties and road performance of the samples were systematically tested according to the SHAP plan. The excellent properties of GO adding to rubber asphalt had been fully verified, and this study provided more options for the modification of asphalt materials.

2 Test materials and methods

2.1 Raw materials and preparation of asphalt samples

Raw materials

The style of matrix asphalt was SK-90 (Whose penetration grade is 90#). 90 means the penetration of the asphalt binder at 25°C ranging from 80 to 100 (units in 0.1 mm). The technical indicators were detected accordance with the “Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering” (JTG E20-2011), and the results were shown in Table 1. The indicators of matrix asphalt met the specification shown as in Table 1. The rubber powder was the A grade 40 mesh rubber powder. The technical specifications of grapheme was shown as in Table 2.

Table 2: Technical indicators of graphene oxide

Indicators	Thickness / nm	Specific surface area / (m ² / g)	Slice size / nm
value	1.6~2.2	1500~2500	0.7~12

Preparation method of rubberized asphalt

The three asphalt samples were prepared by melt blending method, mainly using high-speed shear emulsifier to disperse nano-material graphene, rubber powder and SBS modifier in asphalt.

2.2 Test methods

Graphene oxide rubber asphalt (GORA), rubber asphalt (RA) and SBS modified asphalt (SBSA) were selected as the research objects, and Scanning Electron Microscopy (SEM), multi-stress repeated creep (MSCR) and dynamic shear rheology (DSR) tests were performed. Mixing design of GORA, RA and SBSA asphalt mixture by rotary compaction with AC-13 as the mix structure type (called AC-13-GR, AC-13-R, AC-13-SBS). High temperature stability, low temperature performance and water stability of the mixture were tested.

2.2.1 SEM test

The surface of three asphalt were observed by using a scanning electron microscopy (SEM, ZEISS, EVO18, Germany). The maximum magnification of SEM is 100,000 times. The specific methods for SEM test are followed: (1) Samples of three asphalt were melted into trichloroethylene to form the solution with a mass fraction of 10%. (2) The solution were uniformly dropped on a metal-like pile with a conductive cloth and dried under an infrared lamp. (3) The dry samples were sprayed as trichloroethylene was completely volatile. (4) SEM tests were carried out on samples.

2.2.2 DSR test

According to the SHRP plan, the three kinds of asphalt were subjected to DSR rheology test. The instrument used in the DSR test is a high precision TA-HR-1. The DSR test took the following two sweeping methods.

- (1) Temperature sweep (TS)

TS was in a stress-controlled loading mode with a test temperature range of 58°C to 88°C, an interval of 6°C, a stress level of 100 Pa, and a test frequency of 10 rad/s.

- (2) Frequency sweep (FS)

FS was in strain-controlled loading mode with an angular frequency range of 0.1~100 rad/s, which strain level was 1.25%. The three asphalt samples were subjected to FS test at 30°C~70°C.

2.2.3 Multi-stress creep recovery (MSCR)

MSCR was carried out for 10 times (Creep 1s, recovery 9s) at the temperature of 64°C and shear stress of 0.1 kPa and 3.2 kPa using DSR tester.

2.2.4 High temperature stability test of mixture

Rutting test, which can be evaluated performance of rubber asphalt mixture at the high temperature, were carried out on the asphalt mixtures in accordance with “Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering” (JTG E20-2011). The rutting plate samples with the size of 300 mm×300 mm×50 mm were formed according to rolling method at the temperature of 60°C and pressure of 0.7 MPa.

2.2.5 Low temperature performance test of mixture

The bending test of the trabecular, which could evaluate the performance of the rubber asphalt mixture at low temperature, were performed on the asphalt mixtures with two grades and three contents according to JTG E20-2011. The trabecular specimens were formed with rock cutting machine. Firstly, rutting plate samples were cut into beam specimens with the size of 250mm×30mm×35mm. Secondly, the trabecular specimen was in incubator at -10°C for 3 hours before test. Lastly, the bending test of the trabecular was carried out at the temperature of -10°C and a loading rate of 50mm/min.

2.2.6 Water stability test of mixture

The cylindrical marshall specimens with size of 101.6×63.5mm after repeated freeze-thaw cycles was tested by the Marshall Stability meter produced by Changsha Yaxing Company. Specimens of repeated freeze-thaw cy-

Table 3: Technical indicators of asphalt samples

Name of the pilot project	Asphalt type		
	GORA	RA	SBSA
Penetration (25°C, 100g, 5s)/0.1mm	55	52	63
Softening Point/°C	81	71	81
Ductility (5°C) /cm	22	16	38
Kinematic viscosity (177°C) /Pa.s	2.9	1.7	81
Flexible recovery (25°C)%	93.5	87	96
RTFOT aging			
Quality change/%	−0.02	0.05	−0.30
Residual Softening Point/%	101	114	/
Residual penetration (25°C) /°C	87	76	84
Residual Dilutility (5°C) /cm	21	13.9	23
Storage stability, segregation, 48h softening point difference	0.7	0.9	2.0

cles were tested as follows: (1) The specimens were been for 15 min under a vacuum of 98.0 KPa. (2) Normal pressure was restored, and the specimens keep for 0.5h in the sink of constant 25°C. (3) The test pieces were placed in a plastic bag containing 10 ml of water and frozen for 6h in a refrigerator with constant temperature of −20°C. (4) The specimen was placed in a sink with constant temperature of 25°C and melt for 2 hours. The four steps were a freeze-thaw cycle. The split test was carried out according to the conditions of the freeze-thaw cycle for 0, 3, 6, 9 and 12 times.

3 Results and discussion

3.1 Testing of three asphalt samples

The three asphalt samples of GORA, RA, SBSA were tested. The test results were provided in Table 3.

As given in Table 3, general performance indicators of GO had been improved compared to RA and SBSA.

3.2 Microstructure analysis of materials

The microstructures of GO, RA, GORA, SBSA scanned by SEM were shown in Figure 3.

Figure 2(a) showed that rich oxygen-containing groups were attached to the base surface and the edge of the graphene Oxide structural unit, and these oxygen-containing groups mainly included -COOH, -OH, C=O, and epoxy-O-. These oxygen-containing groups provide a large number of active sites for the functionalization of the surface of GO. Figure 2(b) showed that the rubber particles with irregular shape could not be dissolved in the asphalt,

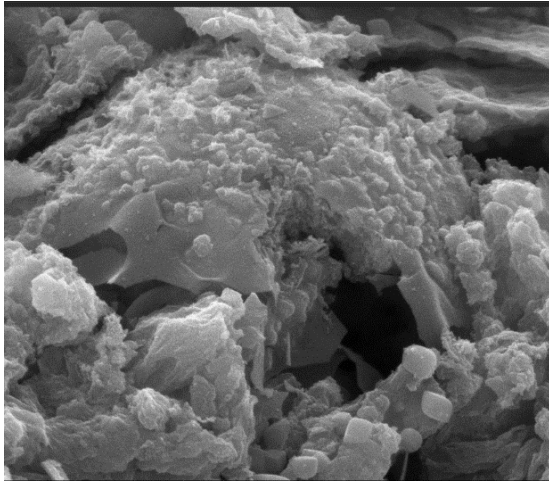
which were wrapped by binder. There was a good adhesion between particles and binder for two-phase interface united closely. Figure 3(c) illustrated the addition of GO to rubber asphalt to form a denser and more stable grid structure. Figure 3(d) showed SBS modified asphalt had agglomerated.

3.3 Rheological parameter analysis

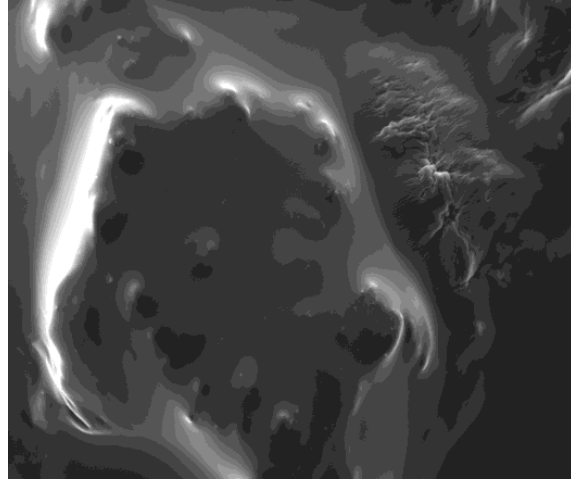
According to SHRP, the shear frequency can be correlated with the amount of road traffic. When the shear frequency is between 0.01 and 100 Hz, it can simulate the normal traffic on the road. Higher frequencies simulate heavier traffic, whereas lower frequencies simulate lighter traffic [40, 41]. Therefore, when the shear frequency is 10^{-3} to 10^3 Hz, it can represent the entire traffic condition. According to the time-temperature equivalent principle [42, 45], the main curve in the wide frequency range of 40°C was synthesized by horizontal displacement superposition, and the relationship between the frequency and the composite modulus G^* was obtained, as shown in Figure 3.

It could be seen from Figure 3 that the composite modulus G^* of the three kinds of asphalt decreased with the decrease of temperature. The modulus value of GORA was the highest under the same temperature, which indicated that the viscoelasticity of GORA increased with the increase of traffic volume, and the sensitivity of change was lower than that of the other two modified asphalts. Therefore, GORA was more suitable for heavy traffic and large temperature difference between day and night.

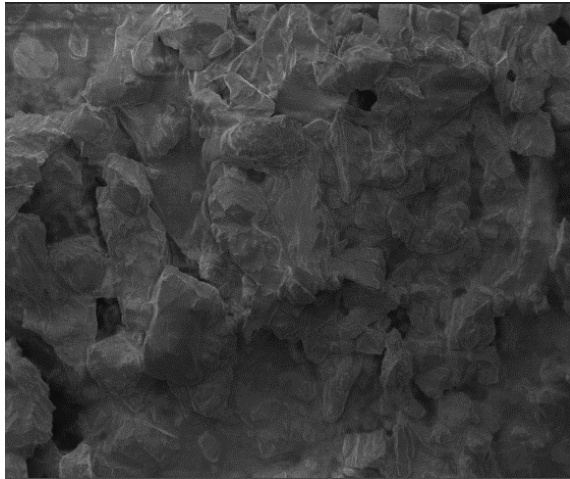
The rutting factor is used to evaluate the ability to resist permanent deformation at high temperatures. The curve of rutting factor with temperature was shown as Figure 4.



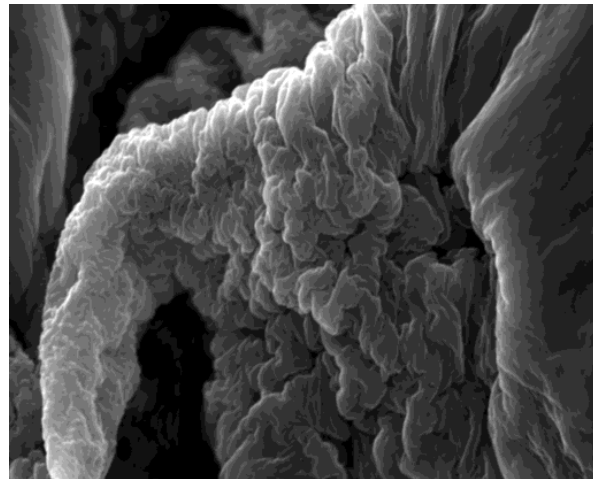
(a) micro - structure of GO



(b) micro - structure of RA



(c) micro -structure of GORA



(d) micro - structure of SBSA

Figure 2: Microstructure of asphalt samples

It could be concluded from Figure 4 that GORA had the best rutting resistance, followed by RA and SBSA. This is due to the fact that GORA's denser and more stable microscopic internal structure increases its ability to resist external loads.

The phase angle reflects the ratio of the elastic component to the viscous component in the test samples. The curve of phase angle with temperature was shown as Figure 5.

From Figure 5, the phase angle of GORA did not change significantly with temperature, and was basically maintained at about 50, which indicated that the viscoelasticity of GORA has little effect on temperature. SBSA showed more flexibility. The phase angle of RA showed an increasing trend with temperature, which indicated that

RA was more sensitive to temperature than the other two types of asphalt. This result also showed that GORA was more suitable for the climate with large temperature difference between day and night.

3.4 Results of MSCR

According to the strain collected by the test, J_{nr} and $J_{nr-diff}$ (J_{nr} -dependent change rate of stress) under different stresses were calculated. The calculation method was as Table 4:

$$J_{nr} = \frac{\varepsilon_r - \varepsilon_0}{\delta} \quad (1)$$

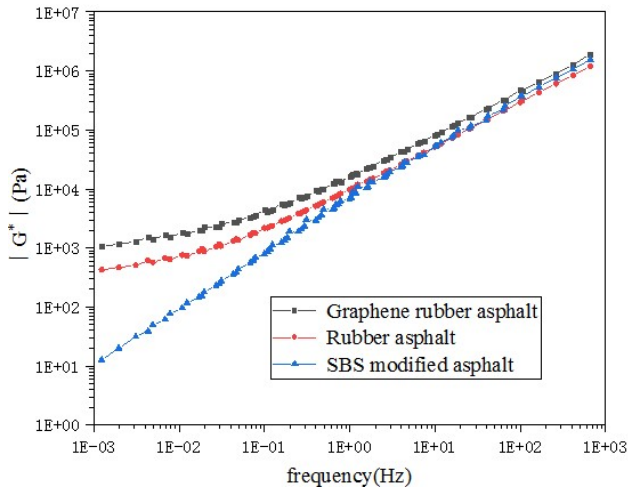
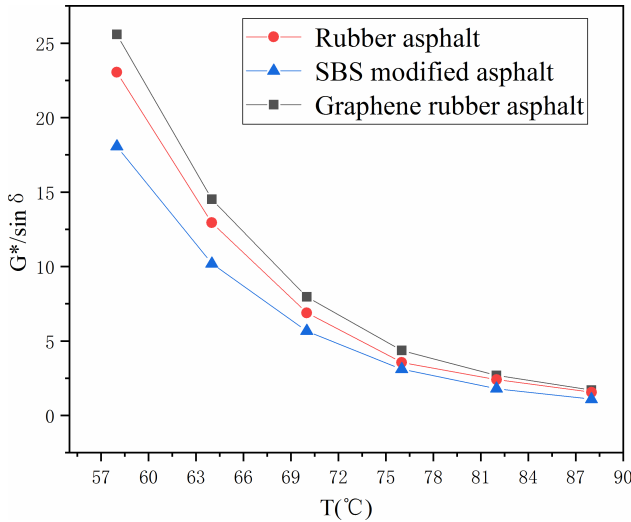


Figure 3: The complex modulus master curve of 40°C

Figure 4: Relationship between $G^*/\sin \delta$ and T

$$J_{nr-diff} = \frac{J_{nr(3.2kPa)} - J_{nr(0.1kPa)}}{J_{nr(3.2kPa)}} \quad (2)$$

ε_0 – Initial deformation; ε_r – Residual deformation after 9s recovery; δ – Shear stress

It could be seen from Table 4 that the $J_{nr-3.2}$ of GORA and RA at 64°C was 0.15 times and 0.26 times of SBSA, and both were less than 0.5, which is a heavy load standard. The $J_{nr-diff}$ of GORA was 0.17 and 0.10 smaller than RA and SBSA, respectively, which indicated that GORA was less sensitive to the increase of stress level than RA/SBSA. So, it indicated that GORA was more suitable for heavy traffic.

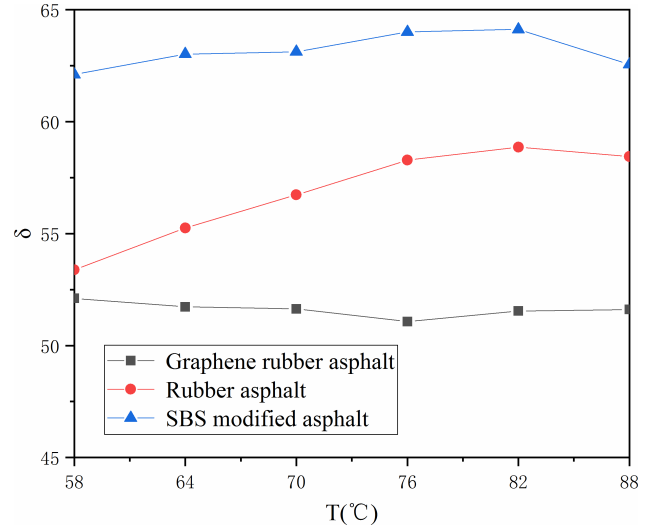
Figure 5: Relationship between δ and T

Table 4: The MSCR results of GORA

Asphalt type	Test value		
	$J_{nr-0.1kPa}$	$J_{nr-3.2kPa}$	$J_{nr-diff}$
Graphene Oxiderubber asphalt	0.04	0.09	0.52
Rubber asphalt	0.05	0.16	0.69
SBS modified asphalt	0.23	0.61	0.62

3.5 High temperature rutting test of mixture

The rutting test of AC-13 were carried out in the study. In order to reduce the test error, three parallel tests were carried out for each sample, and the average values were obtained. The Dynamic stability of AC-13 were shown in Table 5.

The dynamic stability (DS) of the GORA mixture was significantly more improved than that of the other two mixture. The test result investigated that anti-rutting performance was GORA > SBSA > RA.

3.6 Low temperature trabecular test of mixture

The test on beam bending at low temperature was performed for AC-13, and the results were shown in Table 6.

Table 5: Dynamic stability of GORA mixture

asphalt mixture	dynamic stability /time mm ⁻¹			average value /time mm ⁻¹	average value /time mm ⁻¹
	1	2	3		
GORA	6825	7003	7070	6966	≥3000
RA	3946	4221	4020	4062	≥2400
SBSA	5305	6014	5862	5727	≥3000

Table 6: Low temperature cracking test of GORA mixture

Low temperature bending test	Anti-bending strength/MPa	Bending strain /μϵ	Stiffness modulus /MPa	Strain failure requirements/μϵ
AC-13-GR	12.1	7490.8	1682.0	≥ 2800
AC-13-RA	9.6	3886.5	2470.9	
AC-13-SBS	8.7	4679.5	1880.2	

Table 7: Water stability test of GORA mixture

asphalt mixture	Void ratio (%)	Splitting strength	TSR(%)	requirements
AC-13-GR	7.2	0.82	87.1	/
AC-13-RA	7.1	0.69	81.1	≥ 80
AC-13-SBS	7.0	0.47	82.2	≥ 80

3.7 Water stability test of mixture

The Marshall test was carried out to evaluate water stability. The results of the Marshall test on AC-13-GR, AC-13-RA and AC-13-SBS were presented in Table 7.

The experimental results indicate that the addition of GO could significantly improve the durability of asphalt mixture.

4 Conclusion

A rich oxygen-containing group is attached to the base surface and the edge of the GO structural unit. These oxygen-containing groups provide a large number of active sites for the functionalization of the surface of GO. Therefore, mixing GO with rubber powder and asphalt polymer can form a more dense and stable structure.

Through rheological tests, it is found that GORA has better high-temperature rutting resistance and lower temperature sensitivity than RA and SBSA, and is more suitable for heavy traffic and large temperature difference between day and night.

Through the rutting test, low temperature bending test and freeze-thaw splitting test of the asphalt mixture, it was

found that the GORA mixture achieved good road performance.

Acknowledgement: This work was supported by the Research and Development Center of Transport Industry of Technologies, Materials and Equipments of Highway Construction and Maintenance (Gansu Road & Bridge Construction Group) Open Fund (No. GLKF201814), the National Natural Science Foundation of China (Nos. 51868047, 51668041), the Fundamental Research Funds for the Central Universities (31920180077).

References

- [1] Dong F., Zhao W., Zhang Y. et al., Influence of SBS and asphalt on SBS dispersion and the performance of modified asphalt, *Construct. Build. Mat.*, 2014, 62, 1-7.
- [2] Liang M., Liang P., Fan W. et al., Thermo-rheological behavior and compatibility of modified asphalt with various styrene-butadiene structures in SBS copolymers, *Mater. Design*, 2015, 88, 177-185.
- [3] Liang M., Xin X., Fan W. et al., Investigation of the rheological properties and storage stability of CR/SBS modified asphalt, *Construct. Build. Mat.*, 2015, 74, 235-240.
- [4] Xiang L., Wang Z.G., Du Y. et al., Preparation technology and performance analysis of crumb rubber and sbs composite modified asphalt binder, *Adv. Mat. Res.*, *Trans Tech Publ.*, 2011, 160,

- 1320-1324.
- [5] Xiang L., Study on Mechanism and Properties of Waste Rubber Powder/SBS Composite Modified Asphalt, China Univ. Petrol., East China, 2011.
 - [6] Liang M., Xin X., Fan W. et al., Investigation of the rheological properties and storage stability of CR/SBS modified asphalt, *Construct. Build. Mat.*, 2015, 74, 235-240.
 - [7] Fang C., Yu R., Liu S. et al., Nanomaterials applied in asphalt modification: a review, *J. Mat. Sci. Technol.*, 2013, 29(7), 589-594.
 - [8] Yao H., You Z., Li L. et al., Rheological properties and chemical analysis of nanoclay and carbon microfiber modified asphalt with Fourier transform infrared spectroscopy, *Construct. Build. Mat.*, 2013, 38, 327-337.
 - [9] Zhang H., Su M., Zhao S. et al., High and low temperature properties of nano-particles/polymer modified asphalt, *Construct. Build. Mat.*, 2016, 114, 323-332.
 - [10] Yao H., Dai Q., You Z., Fourier Transform Infrared Spectroscopy characterization of aging-related properties of original and nano-modified asphalt binders, *Construct. Build. Mat.*, 2015, 101, 1078-1087.
 - [11] Lazzara G., Milioto S., Dispersions of nanosilica in biocompatible copolymers, *Polymer Degradation and Stability*, 2010, 95(4), 610-617.
 - [12] Motlagh A.A., Kiasat A., Mirzaei E. et al., Bitumen modification using carbon nanotubes, *World Appl. Sci. J.*, 2012, 18(4), 594-599.
 - [13] Shiman L., Shiman A., Spitsyna N. et al., Effects of nanocomposites on the high temperature rheological properties of a PG58 asphalt-binder, *Proc. GeoHunan 2011 Int. Conf.*, 2011.
 - [14] Santagata E., Baglieri O., Tsantilis L. et al., Rheological characterization of bituminous binders modified with carbon nanotubes, *Proc.-Soc. Behav. Sci.*, 2012, 53, 546-555.
 - [15] Shirakawa T., Tada A., Okazaki N., Development of functional carbon nanotubes-asphalt composites, *Int. J. GEOMATE*, 2012, 2(1), 161-165.
 - [16] Khattak M.J., Khattab A., Rizvi H.R. et al., The impact of carbon nano-fiber modification on asphalt binder rheology, *Construct. Build. Mat.*, 2012, 30, 257-264.
 - [17] Khattak M.J., Khattab A., Rizvi H.R., Mechanistic characteristics of asphalt binder and asphalt matrix modified with nano-fibers, *Geo-Frontiers Congress 2011, Amer. Soc. Civil Eng. Industr. Fabrics Assoc. Int. North Amer. Geosynth. Soc. Geosynth. Mat. Assoc.*, 2011.
 - [18] Yao H., You Z., Li L. et al., Rheological properties and chemical analysis of nanoclay and carbon microfiber modified asphalt with Fourier transform infrared spectroscopy, *Construct. Build. Mat.*, 2013, 38, 327-337.
 - [19] Shi X., Goh S.W., Akin M. et al., Exploring the interactions of chloride deicer solutions with nanomodified and micromodified asphalt mixtures using artificial neural networks, *Journal of materials in civil Eng.*, 2011, 24(7), 805-815.
 - [20] Jahromi S.G., Khodaii A., Effects of nanoclay on rheological properties of bitumen binder, *Construct. Build. Mat.*, 2009, 23(8), 2894-2904.
 - [21] Yao H., You Z., Li L. et al., Performance of asphalt binder blended with non-modified and polymer-modified nanoclay, *Construct. Build. Mat.*, 2012, 35, 159-170.
 - [22] Steyn W.J.V.M., Research and application of nanotechnology in transportation, SATC 2008, 2008.
 - [23] Jamshidi A., Hasan M.R.M., Yao H. et al., Characterization of the rate of change of rheological properties of nano-modified asphalt, *Construct. Build. Mat.*, 2015, 98, 437-446.
 - [24] Chen W., Lv G., Hu W., Li D., Chen S., Dai Z., Synthesis and applications of Graphene Oxide quantum dots: a review, *Nanotechnology Reviews*, 2018, 7(2), 157-185.
 - [25] Park J., Yan M., Three-dimensional graphene-TiO₂ hybrid nanomaterial for high efficient photocatalysis, *Nanotechnology Reviews*, 2016, 5(4), 417-423.
 - [26] Dreyer D.R., Park S., Bielawski C.W. et al., The chemistry of graphene oxide, *Chem. Society Rev.*, 2010, 39(1), 228-240.
 - [27] Kim K., Regan W., Geng B. et al., High-temperature stability of suspended single-layer graphene, *physica status solidi, Rapid Res. Lett.*, 2010, 4(11), 302-304.
 - [28] Weitz R.T., Yacoby A., Nanomaterials: Graphene rests easy, *Nature Nanotechnol.*, 2010, 5(10), 699.
 - [29] Pumera M., Graphene-based nanomaterials and their electrochemistry, *Chem. Soc. Rev.*, 2010, 39(11), 4146-4157.
 - [30] König K., Riemann I., Fritzsche W., Nanodissection of human chromosomes with near-infrared femtosecond laser pulses, *Optics Lett.*, 2001, 26(11), 819-821.
 - [31] Kim K., Regan W., Geng B. et al., High-temperature stability of suspended single-layer graphene, *physica status solidi, Rapid Res. Lett.*, 2010, 4(11), 302-304.
 - [32] Zhou B., Wei Z., Zhang Y. et al., Generation of 210 fs laser pulses at 1093 nm by a self-starting mode-locked Yb: GYSO laser, *Optics Lett.*, 2009, 34(1), 31-33.
 - [33] Rao C.N.R., Sood A.K., Subrahmanyam K.S. et al., Graphene: the new two-dimensional nanomaterial, *Angewandte Chem. Int. Ed.*, 2009, 48(42), 7752-7777.
 - [34] Dreyer D.R., Park S., Bielawski C.W. et al., The chemistry of graphene oxide, *Chem. Soc. Rev.*, 2010, 39(1), 228-240.
 - [35] Chen D., Zhang H., Yang K. et al., Functionalization of 4-aminothiophenol and 3-aminopropyltriethoxysilane with graphene oxide for potential dye and copper removal, *J. Hazard. Mat.*, 2016, 310, 179-187.
 - [36] Zhang H., Chang Q., Jiang Y. et al., Synthesis of KMnO₄-treated magnetic graphene oxide nanocomposite (Fe₃O₄@ GO/MnO_x) and its application for removing of Cu²⁺ ions from aqueous solution, *Nanotechnology*, 2018, 29(13), 135706.
 - [37] Wang C.C., Preparation, Mechanism and Properties of Carbon Nanomaterials Modified Asphalt, 2017, Chang'an University, Xi'an, China
 - [38] Hou L.J., Study on Rheological Properties of Nano Graphene Modified Asphalt, *J. East China Jiaotong Univ.*, 2018, 35(4), 133-137.
 - [39] Yu R.E., Zhu X.J., Zhang M.R. et al., Properties and Mechanism of Graphene Oxide/Polyurethane Composite Modified Asphalt Mixture, *Science Technology and Engineering*, 2018, 18(33), 209-213.
 - [40] Pellinen T.K., Witczak M.W., Stress dependent master curve construction for dynamic (complex) modulus (with discussion), *J. Assoc. Asphalt Paving Technol.*, 2002, 71.
 - [41] Li X., Zhang X., Wang S., Study of High Temperature Performance of Asphalt Mastic Based on Dynamic Viscoelastic Mechanics, *J. Highway Transport. Res. Develop. (English Ed.)*, 2007, 2(2), 16-20.
 - [42] Hu L.Y., Zheng Q.H., Yu F. et al., Drawing the transverse dynamic modulus master curve of the axis based on the same temperature equivalence factor, *J. Wuhan Univ. Technol. (Transport. Sci. Eng. Ed.)*, 2019, 43(01), 146-152.

- [43] Nguyen Q.T., Di Benedetto H., Sauzéat C. et al., Time temperature superposition principle validation for bituminous mixes in the linear and nonlinear domains, *J. Mater. Civil Eng*, 2012, 25(9), 1181-1188.
- [44] Nguyen H.M., Pouget S., Di Benedetto H. et al., Time-temperature superposition principle for bituminous mixtures, *Europ. J. Envir. Civil Eng.*, 2009, 13(9), 1095-1107.
- [45] Chehab G.R., Kim Y.R., Schapery R.A. et al., Time-temperature superposition principle for asphalt concrete with growing damage in tension state, *J. Assoc. Asphalt Paving Technolog.*, 2002, 71.