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Study on Road Performance of Graphene Oxide Composite Modified Epoxy Asphalt for Bridge Deck Pavement

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Abstract: *Graphene composite modified epoxy asphalt and its mixture for road and bridge pavement were prepared by adding different proportions of graphene oxide into epoxy asphalt. The effect of graphene oxide on its properties was discussed. The results show that with the increase of GO content, the tensile strength of GO composite modified epoxy asphalt increases and the elongation at break decreases. The high temperature properties, low temperature properties and water stability properties of the mixture were improved to varying degrees.*

Keywords: *Pavement Material, Graphene Oxide Composite Modified Epoxy Asphalt, Mixture, Road Performance*

1. Introduction

With the rapid development of China's highway construction, bridge construction is also a rapid development trend, has built the Xia-Zhangzhou cross-sea bridge, Hong Kong-Zhuhai-Macao bridge, Wuhan Yangsigang bridge, Wufengshan Yangtze River Bridge and so on. With the construction of these long-span steel Bridges, the requirements for steel bridge pavement are also getting higher and higher, China's heavy load, high temperature and other harsh conditions of use on steel bridge pavement materials also put forward higher requirements[1]. Epoxy asphalt pavement system has excellent mechanical properties and high temperature stability, since the successful application in Nanjing Yangtze River Bridge two and achieved good results, epoxy asphalt pavement system in many domestic steel bridge pavement projects have been promoted and applied, become one of the mainstream steel bridge pavement programs in China, by the end of 2022, More than 100 Bridges in China have adopted epoxy asphalt paving, with a cumulative application area of more than 3 million[2] square meters.

Epoxy asphalt paving material has the advantages of high strength, rigidity, excellent fatigue resistance, etc. At present, China's epoxy asphalt paving material mainly relies on imports, represented by the United States ChemCo epoxy asphalt and Japan TAF epoxy asphalt, in China's steel bridge deck pavement has been applied to a certain extent, but in the actual engineering application of epoxy asphalt mixture construction conditions are harsh, And frequent early cracking diseases[3]. This is because epoxy asphalt is a thermosetting material with high cross-linking degree after curing, resulting in the brittleness, poor toughness and impact resistance of epoxy asphalt after curing, and low cracking strain resistance, which is easy to lead to early cracking of epoxy asphalt mixture. After cracking, the integrity of steel bridge deck pavement is significantly affected, the comprehensive performance is attenuated, and the service life of the bridge is directly shortened. Therefore, the toughening modification of ring resin is the fundamental way[4] to solve the poor cracking resistance of epoxy asphalt pavement system and extend its service life. This paper tries to blend graphene oxide into epoxy asphalt in order to obtain better performance of graphene composite modified epoxy asphalt and its mixture for road and bridge pavement.

Since 2004, British physicists Andre Geim and Konstantin Novoselov obtained graphene in the state of two-dimensional nanomaterials by using tape to peel graphite sheets for the first time, graphene has been used in various engineering fields, and graphene oxide (GO) as a derivative of graphene is also the same. [5-7]GO, as the strongest two-dimensional material known to mankind in nature, has excellent mechanical and thermal properties, making the strength and toughness of epoxy resin significantly improved. Fang et al.[8] synthesized amine-functionalized graphene (NH2-GN), which enabled GNs to be covalent bonded with epoxy resin, and at the same time adjusted the crosslinking density of epoxy network by changing the stoichiometric ratio of reaction groups, which could effectively eliminate strain

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energy. Due to the similar molecular structure of the graft chain and epoxy resin, NH2-GN achieved molecular level dispersion in the epoxy resin matrix, and when the addition of NH2-GN in the epoxy resin was 0.6 wt%, the fracture toughness and bending strength of the composite were increased by 93.8% and 91.5%, respectively. Lu [9]research found that the polar oxygen-containing groups in graphene oxide can react with the epoxy resin, and the reactants are superimposed on each other to form a three-dimensional interaction network structure, which can enhance the toughness of the epoxy resin. In this study, GO composite modified epoxy asphalt and its mixture were prepared by mixing GO with epoxy asphalt. The effect of GO content on its properties was compared, and the best content of GO was obtained, which provided a new idea and direction for the modification of epoxy asphalt.

2. Experimental part

2.1 Ingredients

2.1.1 Base asphalt

The compatibility of matrix asphalt and epoxy resin is a key factor to ensure the performance, and the difference between the solubility of the two can be better integrated when the difference is less than 1.5. The solubility of epoxy resin is about 9.6~10.5, while that of ordinary matrix asphalt is about 8.65. Therefore, 70# petroleum asphalt with a high content of asphaltene is selected as the matrix asphalt in this paper. The asphalt is provided by Chongqing Bishan New Building Materials Co., LTD. The specific technical indicators of this asphalt are shown in Table 1.

Table 1 Table of technical indexes of 70# asphalt

Technical specifications	Units	Technical requirement	Test results
Needle penetration (25 ℃,100g,5g)	0.1mm	60 to 80	68
Needle penetration index	PI	-1.5 to 1.0	0.2
Softening point	℃	P 46	47.5
Ductility (15 ° C,5cm/min)	cm	P 100	> 100

2.1.2 Epoxy and curing agent

After the epoxy resin is mixed with a certain proportion of curing agent, it will react to produce curing products, forming an irreversible three-dimensional network structure. The overall performance of the mixture is continuously enhanced. Bisphenol A epoxy resin because of large yield, excellent performance and other characteristics. It is the main raw material for the development of epoxy asphalt. This paper uses HY-128 epoxy resin (HY-128 means A liquid epoxy resin synthesized by bisphenol A and epichlorohydrin), and the curing agent uses the high temperature curing agent developed in the previous research basis (HTC-1), its technical indicators are shown in Table 2 and Table 3.

Table 2 Technical index table of HY-128 epoxy resin

Technical specifications	Units	Technical requirements	Test results
Viscosity (25 ° C)	Pa · s	9000-11000.	9865
Epoxy equivalent	g/mol	170 to 190	189
Density	g/cm3	1.0 to 1.1	1.05
Appearance	-	Transparent	Transparent

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Table 3 HTC-1 High temperature curing agent list

Technical Specifications	Units	Technical requirements	Test results
Viscosity (25 ° C)	Pa · s	1000~ 5000	2450
Amine value	mg KOH/g	145~ 165	156
Density	g/cm^3	0.8 ~ 0.9	0.85
Appearance	-	Tawny	Tawny

2.1.3 Graphene oxide

Graphene oxide is provided by Jiangsu Xianfeng Nanomaterials Co., LTD. It is prepared in the laboratory according to the Hummers method. It has a specific surface area and the appearance is black powder.

2.1.4 Aggregate

The ore materials required for the epoxy asphalt mixture in this test are all produced from Chongqing Tongnan Mixing Station, in which the coarse aggregate is basalt gravel, including the three grades of 0~3mm, 3~5mm and 5~10mm, the fine aggregate is made of limestone machine-made sand, and the ore powder is made of limestone ground fine.

2.2 Preparation of graphene oxide composite modified epoxy asphalt

Graphene oxide was modified with different algebraes and different end-group hyperbranched polyester, and dispersed in bisphenol A epoxy resin by weight ratio of 0.5wt%, 1wt%, 1.5wt%, 2wt% and 3wt%, respectively.5 groups of graphene oxide composite modified epoxy resin were stirred and mixed at 60 ℃ for 30 min by magnetic heating stirrer, respectively. After the liquid level was stable and did not rotate, the graphene oxide composite modified epoxy resin was dispersed by ultrasonic wave in the voice circuit ultrasonic cleaning machine and dispersed by ultrasonic wave in the water bath at 60 ℃ for 45 min to obtain uniform and stable dispersion of graphene oxide composite modified epoxy resin.

The prepared 5 groups of graphene composite modified epoxy resin were poured into the shear machine with the curing agent and the matrix asphalt according to the ratio, and the compatiber was added. The mixture was stirred at 150 ℃ for 30 min at the speed of 1 000 r/min. The stirring speed should not be too fast, and the stirring time should not be too long, because the rapid stirring of epoxy resin and curing agent under high temperature conditions will cause the rapid movement of epoxy resin molecules and curing agent molecules, so that the two are fully in contact, and the collision chance between epoxy resin molecules and curing agent molecules is increased microcosmic, resulting in too fast reaction; Macro high temperature rapid stirring will make the contact area between the epoxy resin and the curing agent increase, so that it quickly form a cross-networking. Too fast reaction speed will lead to the viscosity of the finished product is too large, can not be formed graphene composite modified epoxy asphalt, and even will be completely cured, destroy the instrument.

2.3 Preparation of graphene oxide composite modified epoxy asphalt mixture

The ratio of oil to stone is 6.5%, the modified epoxy resin and curing agent are placed in the oven at 60℃ for heat preservation, the asphalt is heated to 160℃ for reserve, the aggregate is dried at 160 ~ 165℃, and the aggregate grading is screened according to the specification in Table 4. Stirring pot heating up to 175℃, take out the aggregate from the oven into the stirring pot dry mix 30s, first epoxy resin A: curing agent B= 3:2 ratio of A added to B stirring until the emergence of uniform small bubbles into the stirring pot, and then according to the ratio of 1:1 add 160℃ matrix asphalt and uniform stirring for 3min. The prepared epoxy asphalt mixture was placed in the oven at 175℃ and kept warm for 30 minutes. The graphene composite modified epoxy asphalt mixture was struck 50 times on both sides of the Marshall electric compacting instrument to form the Marshall specimen; High and low temperature specimens were formed by rut testing machine and combined rut testing machine, and all specimens were cured under 60 ℃ dry condition for 4 days and then their properties were tested.

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Table 4 Aggregate grading

Screen size /mm	Percentage of mass through the sieve /%	
	Design scope	Design Values
13.200	100	100
9.500	95～100	96.2
4.750	65～85	74.5
2.360	50～75	57.7
1.180	35～60	45.8
0.600	25～45	36.5
0.300	17～33	24.8
0.150	12～23	15.7
0.075	7～14	9.9

3. Properties of graphene oxide composite modified asphalt

According to the technical standard of epoxy bond in JTG/T 3364-2019 "Technical Code for Design and Construction of Highway Steel Bridge Deck Pavement". The tensile strength, elongation at break and water absorption of graphene oxide composite modified epoxy asphalt at 23 ℃ and 25 ℃ were tested, and the results were shown in Table 6.

Table 5 Technical requirements of epoxy asphalt binder

Project	Technical requirements	Test method
Tensile strength (23 ° C)/MPa	= 2.0	GB/T 30598-2014 "General technical conditions for epoxy asphalt materials for road and bridge paving"
Elongation at break (23℃)%	= 100	
Thermosetting (300 ° C)	non-melting	

Table 6 Main properties of graphene oxide composite modified epoxy asphalt

Project	Graphene oxide dosage /%				
	0.5	1.0	1.5	2.0	3.0
Tensile strength (23 ° C)/MPa	5.0	5.3	5.5	6.1	5.8
Elongation at break (23 ° C)/%	189	211	229	247	226
Thermosetting (300 ° C)	non-melting				

It can be seen from Table 5 and Table 6 that the tensile strength and elongation at break of the graphene composite modified epoxy asphalt prepared when the graphene oxide content is 0.5%wt, 1.0%wt, 1.5%wt, 2.0%wt and 3.0%wt can meet the requirements of relevant standards; With the increase of graphene oxide content, the tensile strength and elongation at break of graphene oxide composite modified epoxy asphalt gradually increased; The maximum value is reached when the content of GO is 2.0%wt, so the content of GO should be 2.0%wt.

4. Properties of graphene oxide composite modified asphalt mixture

Refer to the test method of JTG E20-2011 "Highway Engineering Asphalt and Asphalt Mixture test Regulations" to test the high temperature stability, low temperature cracking resistance, water stability and Marshall stability of graphene oxide composite modified epoxy asphalt mixture. The results are shown in Table 7.

4.1 High temperature performance

Early disease survey of asphalt concrete bridge pavement in China found that rutting disease is still more common. The external reason is mainly affected by the two environmental factors of high

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temperature and heavy load in our country, the internal reason is that the asphalt mixture itself decreases in the deformation resistance at high temperature, and it is easy to appear permanent rutting damage under the repeated action of driving load. In addition to the treatment of overloaded vehicles, mainly by improving the performance of paving materials themselves to resist rutting damage. Indoor rutting test method was used to evaluate the high temperature performance of graphene oxide composite modified epoxy asphalt mixture.

Based on the thermosetting characteristics of epoxy asphalt, GO composite modified epoxy asphalt mixture has excellent high temperature performance after forming the final strength. As can be seen from Table 7, with the increase of GO content, the dynamic stability of the mixture at 60 ℃ increases, and when the graphene content is 2.0%, the dynamic stability of the mixture at 60 ℃ increases by 22%. Due to the stability of graphene oxide structure and excellent mechanical properties, when it is evenly dispersed in the epoxy asphalt mixture, it can effectively improve the high temperature performance of the mixture, that is, the dynamic stability of the mixture is greater and the rutting deformation at high temperature is smaller.

4.2 Low temperature performance

In the case of low temperature or large temperature difference, the deformation ability of asphalt mixture is reduced, and when the internal temperature stress cannot be resolved, the asphalt mixture will be pulled apart, resulting in different degrees of cracks on the road surface. The rainwater on the road surface penetrates into the lower subgrade or bridge panel through the cracks, which will cause erosion damage to the subgrade or the main body of the bridge, and seriously affect the safety and life of the road and bridge. The mechanical index of trabecular bending failure at low temperature was used to evaluate the low temperature performance of graphene oxide composite modified epoxy asphalt mixture.

As can be seen from Table 7, the -10 ℃ low temperature bending stiffness modulus of the mixture gradually increases with the increase of graphene oxide content, and graphene oxide can improve the low temperature performance of the mixture. When the graphene content is 3.0%, the -10 ℃ low temperature bending stiffness modulus of the mixture begins to decrease, so the appropriate graphene oxide content can improve the low temperature performance of the mixture, and it is recommended that the graphene oxide is 2.0%wt.

4.3 Water stability

When the asphalt pavement is in the rain-soaked or freeze-thaw cycle environment for a long time, coupled with the repeated action of driving load, the road surface water will penetrate through the mixture gap into the asphalt film and aggregate contact interface, weakening the adhesion of asphalt and aggregate. In the long-term damage, the asphalt film and aggregate are separated, and the road surface appears the diseases such as grain drop and pit. In order to prevent and control such diseases, it is necessary to improve the cracking resistance of asphalt mixture and the adhesion ability between asphalt and aggregate in addition to the anti-drainage measures of pavement. In general, two commonly used test methods are used to evaluate the water stability of epoxy asphalt mixture, including water immersion Marshall test and freeze-thaw splitting test. This test uses water immersion Marshall test to evaluate the water stability of graphene oxide composite modified epoxy asphalt mixture.

Table 7 Properties of graphene composite modified epoxy asphalt mixture

Project	Graphene dosage /%									
	0.5		1.0		1.5		2.0		3.0	
60°C-dynamic stability/(times /mm)	37863	39505	41510	41356	42555	43840	45440	46500	45870	44793
-10℃ low temperature bending stiffness modulus /MPa	6179	6101	6215	6532	6834	6845	7682	7566	7325	7524
Submerged Marshall residual stability MS_0/%	86.5	87.6	88.3	87.4	89.2	90.4	90.2	91.4	92.5	93.8
Marshall stability /kN	81	80	79	82	85	87	91	90	89	87

The experiment shows that basically the GO epoxy asphalt mixture has a high Marshall stability and the stability increases obviously with the increase of the GO content. At the same time, it can also be seen from Table 7 that the water stability of the mixture can be improved to a certain extent with the increase

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of the content of GO. This means that graphene oxide is uniformly dispersed in graphene oxide composite modified epoxy asphalt mixture, acting as a stable and high-strength support system in the pores of the aggregate of the mixture, which further improves the performance of the mixture.

5. Conclusion

Graphene oxide composite modified epoxy asphalt and its mixture for road and bridge pavement were prepared by adding graphene oxide. The effects of graphene oxide content on its properties were compared, and the following conclusions were drawn:

1) The properties of the graphene oxide composite modified epoxy asphalt obtained by adding graphene oxide into the epoxy asphalt system meet the requirements of relevant standards; With the increase of GO content, the tensile strength and elongation at break of GO composite modified epoxy asphalt showed a trend of first increase and then decrease. It is recommended that the content of GO should be 2.0%wt.

2) The effect of GO content on the road performance of GO composite modified epoxy asphalt mixture was studied. The results showed that the addition of graphene can effectively improve the high temperature rutting resistance, low temperature cracking resistance, water stability and Marshall stability of the mixture.

3) Considering the cost of GO and the effect on the properties of GO composite modified epoxy asphalt and GO composite modified epoxy asphalt mixture, it is suggested that the best dosage of GO is 2.00% wt. Graphene of 2.00% wt can significantly improve the road performance of graphene composite modified epoxy asphalt mixture, has a good application prospect and market, and also provides a new idea and direction for the modification of epoxy asphalt.

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