Research Article

Multiscale Study on the Effect of Nano-Organic Montmorillonite on the Performance of Rubber Asphalt

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Received 27 April 2018; Accepted 22 May 2018; Published 17 September 2018

Academic Editor: Victor M. Castaño

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In order to improve the high-temperature performance, antiaging performance, and storage stability of rubber asphalt, nano-organic montmorillonite (NOMMT) was mixed with rubber asphalt. Macroscopic influences of NOMMT on rubber asphalt were measured through penetration, softening point, ductility, rotational viscosity tests, dynamic shear rheology test, and bending beam rheology test at low temperature and were conducted on rubber asphalt with different contents of NOMMT. Then, the microscopic mechanism of NOMMT on the microscopic performance of rubber asphalt was studied through using scanning electron microscopy (SEM), infrared spectroscopy (IR), and differential scanning calorimetry (DSC). The results showed that the rubber particles were smoother, uniform, and dispersed after NOMMT was introduced, and the compatibility between NOMMT and crumbed rubber powder was good. Some stable structures were formed in the composite modified asphalt. The disappearance of alcohol phenol and the increase in related groups such as alkane, benzene, and hydrocarbon indicated that chemical reaction occurred between NOMMT and rubber asphalt, resulting in the changes of the performance of the composite modified system, so that high-temperature stability, antiaging properties, and storage stability were improved but its low-temperature performance was decreased.

1. Introduction

The wasted tire is a kind of black pollution to the natural environment and ecology [1, 2]. The main components of waste tire powder are natural rubber (NR) and synthetic rubber (SR), containing styrene butadiene rubber, isoprene rubber, polybutadiene rubber, and other polymers [3]. These polymers in the natural state are difficult to be decomposed naturally. But specially treated crumb tire rubber can not only solve the problem of ecological environment but also improve the performance of asphalt. It has significant environmental, social, and economic benefits, so all countries are promoting the application of rubber asphalt [4].

Magdy found that high temperature can make the rubber particles swell to 2 to 3 times, and the viscosity of rubber asphalt system increased to some extent [5]. Cetin considered that rubber particle was only bulged in asphalt. It was physical mixing, and there was no chemical reaction between rubber and asphalt [6]. Mashaan et al. affirmed that the cause of the increased viscosity of rubber asphalt was that light oil

in asphalt was absorbed by rubber particles [7]. Meanwhile, because of poor compatibility between rubber particles and asphalt, rubber particles were dispersed in asphalt in a suspension state, which was not dissolved in asphalt, and did not achieve the real sense of modification. Runde thought that rubber powder was an island structure state in asphalt and that their compatibility was not good [8]. So, it was prone to segregate during storage. To solve the storage problem, Oyekunle found that some kinds of acid will improve the storage of rubber asphalt [9]. Al-Mansob et al. investigated the effects of Al_2O_3 addition to the base asphalt and epoxidised natural rubber modified asphalt (ENRMA), including the physical properties, storage stability, rheological properties, and microstructure of the binders [10]. Han et al. evaluated the physical and rheological properties of crumb rubber (CR) and nanosilica-modified asphalt through a set of physical property tests and fluorescence microscopy, scanning electron microscopy, and Fourier-transform infrared spectroscopy techniques [11]. Jeffry et al. evaluated the microstructure properties of asphalt mixtures using atomic

TABLE 1: Technical index of the 70A matrix asphalt.

Penetration at 35°C (0.1 mm)	Penetration index (PI)	Softening point (°C)	Ductility at 15°C (cm)	Quality change after TFOT (%)	Ratio of residual penetration after TFOT (%)	Residual ductility after TFOT (cm)	Relative density at 25°C
67	−0.37	47.5	>100	0.1	65.2	6.8	1.043

TABLE 2: Technical index of the waste rubber powder.

Density (g/cm ³)	Water content (%)	Metal content (%)	Fiber content (%)	Acetone extraction (%)	Ash content (%)	Carbon black (%)	Rubber hydrocarbon (%)	Natural rubber (%)
1.23	0.57	0.03	0.13	4.3	3.9	30	57	76

force microscopy (AFM) and field-emission scanning electron microscopy (FESEM). Results showed that 6% NCA has the lowest surface roughness which improved the adhesion of asphalt mixture. Flat and dense asphalt mixture was observed from FESEM which contributed to the enhancement of asphalt mixture engineering performance [12]. Wu et al. scanned asphalt composite modified with layered dihydroxy composite metal hydroxide (LDHS) and rubber powder with atomic force microscope (AFM) and found that the introduction of LDHS can make the waste rubber powder to be grinded more finely, improve the high-temperature performance, reduce the softening point, and enhance the antiaging performance of asphalt rubber [13]. Azarhoosh et al. assessed the effects of using nano-TiO₂ on the adhesion between the asphalt binder and the aggregate using the surface free energy (SFE) method [14]. Iskender investigated the performance of nano-clay-modified asphalt mixtures [15].

It can be seen from the above researches that the compatibility between rubber powder and asphalt is poor and that the rubber is simply oil absorbing and swelling; there is no chemical reaction in asphalt from macroscopic tests, so the modification effect on asphalt is limited. Therefore, in this paper, nano-organic montmorillonite (NOMMT) was used as a coupling agent to enhance the interaction between rubber powder and asphalt and to improve the modified effect of rubber asphalt. The effects of NOMMT on the macroscopical properties, such as high-temperature stability, aging resistance, and storage stability, of rubber asphalt were studied from the macroscopic point of view, including penetration, softening point, ductility, viscosity, dynamic shear rheological test (DSR), and bending beam rheological test (BBR). Meanwhile, the microscopic mechanism of NOMMT on rubber asphalt was also studied from the microscopic perspective by means of scanning electron microscopy (SEM), infrared spectroscopy (FTIR), and differential scanning calorimetry (DSC). This research is significant to understand deeply the mechanism of NOMMT on rubber asphalt and to optimize its performance.

2. Raw Materials

- (1) Asphalt: 70A grade matrix produced by Taihe Asphalt Products Co., Ltd. Its technical index according to the Chinese Technical Specifications

for Construction of Highway Asphalt Pavements [16] is shown in Table 1.

- (2) Waste rubber powder: size 100, mesh rubber powder produced by Yuxin Building Material Co., Ltd., and its technical index is shown in Table 2.
- (3) Organic montmorillonite: produced by Zhejiang Walter New Material Co., Ltd., the largest particle size is 120 nm, its average particle size is 70–80 nm, the specific surface area is 750 m²/g, and the diameter thickness ratio is >200.

3. Preparation of Rubber Asphalt and NOMMT-Rubber Asphalt

In this paper, high-speed shear emulsifier MBE-10 was used in the preparation of rubber asphalt and NOMMT-rubber composite modified asphalt. The preparation processes are as follows:

- (1) The 70A matrix asphalt was heated to 160°C in the oven and was held for 1 h, then 20% of rubber powder and different contents (0%, 1%, 2%, 3%, 4%, and 5%, resp.) of NOMMT were added into the asphalt after mixing; the mixture for 15 min was stirred with a glass rod and then heated for 10 min in the oven at 180°C for the rubber to fully absorb the oil.
- (2) The mixed asphalt was high-speed sheared with MBE-10. Firstly, the mixed asphalt was sheared for 5 min at low speed (500 rpm) and then rotated at 4000 rpm for 45 min. Temperature was controlled to 180°C in the shearing process.

So, NOMMT and rubber powder composite modified asphalt with different NOMMT contents were prepared. The prepared samples were labeled separately and preserved in a thermostat oven at 180°C for subsequent tests.

4. The Effect of NOMMT on the Macroscopical Properties of Rubber Asphalt

4.1. Normal Performance Indexes of Asphalt: Penetration, Softening Point, Ductility, and Viscosity. Conventional performance tests, including penetration at 25°C, softening

point, ductility at 5°C, and viscosity at 135°C, were conducted on rubber asphalt with different contents of NOMMT (0%, 1%, 2%, 3%, 4% and 5%, resp.) [17]. The variations of technical indexes to the contents of NOMMT were obtained as shown in Figures 1–4.

From Figures 1–4, it can be seen that the penetration and ductility at 5°C of rubber asphalt decrease with the increase of NOMMT contents and the softening point and viscosity at 135°C increase with the increase of NOMMT contents.

4.2. High-Temperature Performance. DSR tests were conducted at different temperatures (52°C, 58°C, 64°C, and 70°C) on the NOMMT-rubber compound modified asphalt with different NOMMT contents [16, 17]. The influence of NOMMT and its contents on the dynamic shear modulus G^* , phase angle, and rutting factor $G^*/\sin \delta$ of different rubber asphalts was obtained as shown in Figures 5–7.

From Figures 5–7, it can be seen that

- (1) Complex modulus G^* increases with the increasing of NOMMT content at the same temperature. G^* decreases as temperature rises at the same NOMMT content.
- (2) At the same temperature, the phase angle, δ , decreases first and then increases with the increase of NOMMT content, and it reaches the minimum when NOMMT content is 3%. In the same NOMMT content, δ increases with the increase of temperature.
- (3) At the same temperature, the rutting factor, $G^*/\sin \delta$, becomes larger with the increase of NOMMT content. This indicates that the introduction of NOMMT will improve the high-temperature performance of rubber asphalt. With the increase of NOMMT content, the high-temperature performance of asphalt is getting better and better, but when the content is more than 3%, the improvement is less.

4.3. Low-Temperature Performance. Bending beam rheological test (BBR) was carried out on NOMMT-rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%) [17]. The influence of NOMMT and its contents on the low-temperature performance index of rubber asphalt was obtained as shown in Table 3 and Figures 8 and 9.

From Figures 8 and 9, it can be seen that the addition of NOMMT makes the creep stiffness of rubber asphalt increase but the m value decreases. Therefore, the low-temperature performance of rubber asphalt is reduced to a certain extent by the addition of NOMMT, and the influence is different at different temperatures.

4.4. Storage Stability. Storage stability tests were carried out on NOMMT-rubber asphalt with different NOMMT contents according to the Chinese test specification [16]. Tubes containing NOMMT-rubber composite modified asphalt were put into the oven at 163°C for 48 h. Then, the tubes were cut into three segments with the same length after removing from the oven. The softening points of the asphalt in the upper and lower sections were measured. The storage

FIGURE 1: Variation of penetration to NOMMT contents.

FIGURE 2: Variation of softening point to NOMMT contents.

FIGURE 3: Variation of ductility at 5°C to NOMMT contents.

stability of NOMMT-rubber asphalt was evaluated according to the difference in the softening points of asphalt in the upper and lower tube sections. The results are shown in Figure 10.

FIGURE 4: Variation of viscosity at 135°C to NOMMT contents.

FIGURE 5: Variation of G^* to NOMMT contents. FIGURE 6: Variation of δ to NOMMT contents.

It can be seen from Figure 10 that the softening point difference between the rubber asphalt in the top and the lower sections is about 10°C, which indicates that there is an obvious segregation in rubber asphalt (NOMMT content is 0%). After adding NOMMT, the softening point of asphalt in the lower section changed a little, but that in the upper section

FIGURE 7: Variation of $G^*/\sin \delta$ to NOMMT contents.

TABLE 3: Effects of NOMMT content on the low-temperature performance index of rubber asphalt.

NOMMT content (%)	Creep stiffness (MPa)			m value		
	-12°C	-18°C	-24°C	-12°C	-18°C	-24°C
0	35.6	107.2	182.8	0.463	0.352	0.322
1	41.4	120.5	252.0	0.455	0.356	0.248
2	46.0	123.5	339.0	0.460	0.336	0.238
3	47.8	113.0	275.5	0.475	0.354	0.238
4	50.7	127.5	346.5	0.454	0.344	0.258
5	52.2	103.0	223.5	0.460	0.412	0.230

FIGURE 8: Variation of creep stiffness with NOMMT content.

gradually increased with the increase of NOMMT content. The softening point curves are gradually approached, and the softening point difference between the upper and lower sections gradually decreased. It shows that NOMMT can improve the storage stability of rubber asphalt and, with the increase of NOMMT content, the segregation degree of rubber asphalt is smaller and the storage stability is gradually improved.

FIGURE 9: Variation of m value with NOMMT content.

FIGURE 10: Variation of softening points of NOMMT-rubber asphalt with NOMMT content after a storage of 48 h.

5. Micromechanism Studies on the Effect of NOMMT on the Performance of Rubber Asphalt

The macroperformance test results of rubber asphalt with different NOMMT contents showed that the addition of NOMMT will improve the high-temperature stability of rubber asphalt, and its storage stability, but will reduce its crack resistance at low temperature. So, scanning electron microscopy (SEM), infrared spectroscopy (IR), differential scanning calorimetry (DSC) were used to study the mechanism of NOMMT on the performance of rubber asphalt from a micro scale.

5.1. Effect of NOMMT and Its Contents on the Distribution of Rubber Particle. SEM can be used to directly observe the microstructure characteristics of the material. KYKY-EM3200 was used to scan the microstructure of rubber asphalt with different NOMMT contents (1%, 2%, 3%, 4%, and 5%) to study the effects of NOMMT and its content on the distribution, shape, and swelling of rubber particles

FIGURE 11: Scanning electron microscope, KYKY-EM3200.

(Figure 11). During the scanning, magnification of SEM was 3000 times, and the resolution was $10\mu\text{m}$ [18]. The particle morphology and distribution of rubber powder were observed.

5.1.1. Processing of the Samples. Firstly, asphalt was poured into blocks, then put into an ethanol solution at -24°C for 1 h. Then, the specimen, a cuboid sheet, $8\text{ mm} \times 3\text{ mm} \times 2\text{ mm}$, was cut from the center of the blocks with a clean knife after taking out.

The specimen was attached to the copper table with conductive adhesive, then it was put into the vacuum coating machine to spray gold on it. Lastly, it was placed into the cabin of SEM for observation [19].

SEM photos of rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%) are shown in Figure 12.

As can be seen from Figure 12, rubber particles have been clustered together, irregular in shape and rough in surface (Figure 12(a)). With the addition of NOMMT and the increase of its content, the size of the crowded rubber particles gradually decreases, and the distribution of rubber particles is gradually uniformed. When the content of NOMMT is up to 3%, the distribution of the rubber particles in the asphalt is more uniform, the size and shape of the particles are more consistent, and the particles' surface is smooth.

Comparing Figure 12(a) with Figure 12(b), it can be found that in Figure 12(a), rubber particles have obvious edges and corners and the expansion extent is low, while the expansion extent on the particle surface is higher in Figure 12(b). Also in Figure 12(b), the color of particle surface has been turn into white and some hairy flocs have been generated on the particle surface, which indicates that when the NOMMT content is only 1%; it has been able to enhance the ability of the rubber particles to absorb the oil content in asphalt, to increase the swelling degree of the particle surface and to deepen the degree of fusion of rubber particles and asphalt. This indicates that NOMMT will affect the dispersion of the rubber particles in the asphalt, the surface smoothness of the rubber powder, and the shape and size of particle clusters.

5.2. Effect of NOMMT and Its Contents on Organic Groups in Rubber Asphalt. The TENSOR 27 Fourier-transform infrared

FIGURE 12: SEM photos of rubber asphalt with different NOMMT contents (3000x).

spectrometer (FTIR) was used, produced by Bruker German, with a resolution of 4 cm^{-1} , scanning time was 32, and test range was $4000\sim 400\text{ cm}^{-1}$. The effect of NOMMT and its contents on organic, functional, and molecular groups in rubber asphalt was analyzed through the comparative analysis of band frequency, infrared spectrum waveform, and intensity in the infrared spectrograms of rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%) [20].

5.2.1. Preparation of Samples. Solution method was adopted in the preparation of the FTIR samples. (1) Qualified KBr windows were washed and then dried with the drying lamp. (2) Rubber asphalt was dissolved using solvent trichloroethylene into a solution with a concentration of 5% (mass

fraction). (3) An appropriate amount of the solution was dropped on the KBr window with a suction pipe, then drying it under the lamp. (4) The KBr window was fixed onto the specimen shelf, and then it was put into the sample chamber of FTIR and was scanned in FTIR.

The infrared spectrograms of rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%) were shown in Figure 13.

Compared with the spectrums of rubber asphalt, Figure 13(a), and NOMMT-rubber asphalt, Figures 13(b)–13(f), it can be found that

- (1) After the introduction of NOMMT, the disorderly absorption peaks were caused by disappearance of alcohol phenol in the range of $3600\sim 3700\text{ cm}^{-1}$.

FIGURE 13: Infrared spectrograms of rubber asphalt with different NOMMT contents.

(2) When the content of NOMMT is 1%, the vibration of C-C skeleton at 812 cm^{-1} , the stretch vibration of $-\text{CH}_2-$ at 2923 cm^{-1} and 2852 cm^{-1} , flexural vibration of $-\text{CH}_2-$ at 1458 cm^{-1} , and the shear scissors vibration of $-\text{CH}_3-$ at 1375 cm^{-1} were enhanced. At this point, the absorption peaks of all groups reached a strong position, which fully indicated that after the introduction of organic montmorillonite, the alcohol

phenol group reacted with other substances in the asphalt, causing the alcohol phenol group to disappear and other new groups to be produced.

The results indicated that the improvement of the performance of NOMMT-rubber asphalt was due to not only the simple physical reaction but also the effect of chemical reaction. It is because of the comprehensive effect of physical and

chemical reactions that the number of groups and the relationship between groups in NOMMT-rubber asphalt were changed; thus, a more stable state was formed and storage stability and high-temperature performance were improved.

Combining the analysis of infrared spectrograms and SEM diagram, both the exfoliation of the edges and the appearance of hairy floc on the surface of rubber particle were probably due to the chemical reaction on the surface of rubber particle. With the increase in NOMMT content, no other peaks appear in the infrared spectrum, and there is no obvious change in the images, which indicated that NOMMT can be used to enhance the stability and compatibility of rubber asphalt.

5.3. Compatibility of NOMMT and Rubber Asphalt. In this paper, the power-compensated differential scanning calorimeter (DSC) produced by PerkinElmer company was used to obtain the DSC curves of the melting section of rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%). The heating rate is 10°C/min.

The values and their variation of the three parameters in the DSC curve (T_m is the peak value of the endothermic peak, H_f is the acreage of the endothermic peak, and T_e is the complete melting temperature) can be used to analyze the microstructures of rubber asphalt and NOMMT-rubber composite modified asphalt, to study the compatibility of NOMMT and rubber asphalt and to determine whether NOMMT has the coupling agent effect in rubber asphalt [21].

5.3.1. Sample Preparation Methods. Firstly, 1–3 g of asphalt sample was dipped with a clean pin randomly after the rubber asphalt with different NOMMT contents was melted, and then were adhered to a 40-microliter flat-bottomed crucible. Secondly, the crucibles were put into the oven at 160°C for 15 min to make the asphalt evenly cover the bottom of the crucible. Thirdly, the crucibles were removed from the oven and put into a refrigerator to cool for 10 min. Finally, the crucibles were taken out after the asphalt hardened and then were put into sealed sample cabin for the DSC test.

The DSC curves of rubber asphalt with different NOMMT contents (0%, 1%, 2%, 3%, 4%, and 5%) are shown in Figure 14 and Table 4.

It can be seen from Figure 14 and Table 4 that

- (1) DSC endothermic curve of rubber asphalt (0% NOMMT) is a smooth flat curve, and there is no absorption peak, indicating that the rubber asphalt has been transited into a fluid state after absorbing heat. DSC curve is relatively flat but the gradient becomes larger when the temperature is higher than 49°C. This indicates that when the heat absorption of rubber asphalt is not uniform when the temperature is below 49°C, and only when the temperature is higher than 49°C, rubber asphalt can be completely transited into a fluid state.
- (2) The heat flow rate of NOMMT-rubber asphalt is smaller than that of rubber asphalt, indicating that NOMMT-rubber composite modified asphalt has

FIGURE 14: DSC curves of rubber asphalt with different NOMMT contents.

TABLE 4: Parameters of the DSC curves of rubber asphalt with different NOMMT contents.

NOMMT content (%)	T_m	H_f	T_e
0	—	—	—
1	48.671	150.649	52.090
2	48.697	145.682	52.065
3	48.888	147.289	52.072
4	48.658	149.763	52.046
5	48.649	146.187	52.030

better thermal stability than rubber asphalt. The heat capacity of NOMMT is lower than that of asphalt and rubber particles, and the heat absorbed by NOMMT is lower than that of asphalt and rubber powder.

- (3) The DSC curves of NOMMT-rubber asphalt have a heat absorption peak in the melting section, and the corresponding temperature is about 48.6°C and the melting temperature is about 52°C, which is generally smaller than their softening points.
- (4) The three parameters of the heat absorption peak of NOMMT-rubber asphalt do not change with the increase of NOMMT content, which indicates that NOMMT has good compatibility with rubber powder and they can form a stable system.
- (5) The endothermic peak appears in the DSC curve of NOMMT-rubber asphalt, while it does not exist in the DSC curve of the rubber asphalt, indicating that a new structure is generated in the NOMMT-rubber asphalt composite system. This may be due to the compatibility of NOMMT with rubber powder and the organic groups on NOMMT can be linked to asphaltene; thus, the structure of rubber-NOMMT-matrix asphalt is formed. Therefore, NOMMT has played the role of a coupling agent, which is the “bridge” of rubber particle and matrix asphalt. The chemical reaction between rubber particle and

NOMMT has formed a new molecular structure, resulting in the expansion rubber particles and the appearance of hairy white floc.

6. Conclusions

In this paper, rubber asphalt and NOMMT rubber asphalt with different NOMMT contents were prepared. Their macromechanical performance was tested through the penetration, softening point, ductility and viscosity, and DSR and BBR tests. And their microstructures were studied through SEM, IR, and DSC tests. So, the effect of NOMMT and its content on the macroscopic properties of rubber asphalt were studied, and the micromechanism was analyzed according to the test results.

- (1) The incorporation of NOMMT into rubber asphalt can reduce the penetration and ductility at 5°C and increase the softening point and the viscosity at 135°C. The more the amount of NOMMT is added, the greater the change of performance index in the range of NOMMT contents used in the paper.
- (2) The incorporation of NOMMT into rubber asphalt will enhance the high temperature performance and decrease their low temperature performance.
- (3) The storage stability of rubber asphalt can be improved through the addition of NOMMT.
- (4) It was found from SEM scanning that the surface of rubber particles became smooth, the edges and corners fell off, and the distribution was more uniform when NOMMT was added into rubber asphalt.
- (5) FTIR spectrum analysis showed that after adding NOMMT, the phenolic group in rubber asphalt disappeared, while benzene ring, -CH₂- group, -CH₃ group, and olefin group increased, which fully showed that chemical reaction happened between NOMMT and related substances in rubber asphalt, and a new group was formed.
- (6) DSC test results showed that the thermal stability of NOMMT-rubber asphalt is better than that of rubber asphalt. There is an endothermic peak in the DSC curve of NOMMT-rubber asphalt; the three major parameters of the endothermic peak do not change with the increase of NOMMT content. This indicated that there is good compatibility between NOMMT and rubber particle, and they can form a stable system.
- (7) Comprehensive analysis of SEM, FTIR, and DSC test results indicated that chemical reaction occurred between NOMMT and rubber asphalt and some new groups were generated. This may be because NOMMT acts as a coupling agent in rubber asphalt, and a new stable structure of "rubber-NOMMT-asphalt" was formed between asphalt and rubber particles.

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 there is no conflict of interest regarding the publication of this paper.

Acknowledgments

The authors appreciate the support of the National Natural Science Foundation of China (50878032).

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