

Research Article

Experimental Study on Macro-Mesoscopic Components and Indexes of Buton Gilsonite-Modified Asphalt

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Gilsonite-modified asphalt is one of the hotspots of high-performance modified asphalt research. In this paper, the macroscopic composition and index of gilsonite-modified asphalt are analyzed; gilsonite, matrix asphalt, and modified asphalt with different gilsonite contents were analyzed by high- and low-temperature performance tests, antiaging performance test, four-component test, infrared spectral test, and SEM electroscope scanning test. The results showed that the content of aromatic fraction, colloid, and asphaltene in Buton gilsonite was 3.1%, 95.6%, and 32.9% higher than that of matrix asphalt, respectively. The gilsonite-modified asphalt had better high-temperature performance and antiaging performance. The optimum amount of gilsonite in gilsonite-modified asphalt should be 9%~15%. The pore structure of gilsonite is more distributed, the surface is rough, which can produce greater cohesion with matrix asphalt, and the compatibility is better.

1. Background

With the rapid development of the economy, there has been a growing demand for transportation, and the vehicles have become large, heavy, and channelized. In summer, most parts of China are hot and rainy [1]. These traffic load conditions and climatic conditions are one of the many unfavorable factors that have caused severe challenges in China's asphalt pavement. Early damage occurs soon after the road is completed and opened [2]. The damage type is especially serious due to rutting and water damage. Far from reaching the design of the age of use, it is hard for the traditional asphalt pavement to shoulder the heavy responsibility. Starting from the basic raw materials, the use of better materials may solve or alleviate the early road damage. Asphalt is especially important as cement [3]. Therefore, it is necessary to carry out high-performance modified asphalt research to solve the problem of early road surface defects and extend the service life of the pavement [4].

Gilsonite is a bituminous substance obtained by oxidizing and polymerizing oil in a natural environment for millions of years. It is formed in rock crevices with a depth of several hundred meters or more and a width of only several tens of centimeters to several meters. Due to its long-term coexistence with the harsh natural environment, its properties are particularly stable, especially aging resistance [5, 6]

Gilsonite was discovered as early as the 1860s [7], but it has not been widely used until the mid-1880s when gilsonite was used for waterproof coating [8]. At present, natural asphalt is widely used as a new type of modifier because of its excellent road performance, simple construction process, and reasonable price and economy. Since natural gilsonite can effectively reduce the occurrence of rutting and water damage, paving asphalt pavement with natural gilsonite as a modifier is an effective measure to deal with the early damage of roads, thereby extending road life and improving road surface [9]. Natural gilsonite is a kind of asphalt, and it is a petroleum derivative with matrix asphalt. Therefore, the compatibility between the two is good, and there is no

segregation. Most of the natural asphalt is solid, and it is easy to process in production [10]. There is only the need to add natural asphalt modifier according to the blending ratio to obtain the corresponding modified asphalt mixture, so no special mixing equipment is needed. Part of the road performance has improved to some extent. At the same time, the natural asphalt is easy to store and transport, and the construction is simple, which makes it widely used in highways, bridge decks, airports, and tunnels [11].

In the research on gilsonite, North American gilsonite was found to be the most widely used. It has four characteristics: antifracking, antirutting, antiaging and durability, so it has wide application, especially in the roads with high stress and near toll stations. Roads, urban expressways and main roads, heavy-duty lanes, bridge deck pavements, intersections, parking lots, stations, bends, and ramps are widely used [12]. Successful applications have been achieved on heavy-duty roads in Oslo, Norway, and New South Wales, Australia. In order to reduce the deformation problems such as rutting and displacement of asphalt pavement, the Australian Road Society (AAPA) recommends the use of gilsonite-modified asphalt [13]. Because of this, the performance of gilsonite has been recognized in Australia. The traffic volume of the New Jersey toll road is about 500,000 per day, and the heavy-duty trucks account for about one-fifth of the number. After five years of using the gilsonite pavement, the road performance is still good and there is no obvious rutting and congestion problems [8].

Cha et al. [14, 15] selected Esso 70# asphalt as the base asphalt and prepared the modified asphalt at 5% intervals in the range of 5% to 50% of the BRA asphalt mixture by the external blending method, and the properties were determined. The research shows that the deformation resistance, high-temperature performance, low-temperature performance, and temperature-sensing performance of Buton rock-modified asphalt are obviously improved with the increase of Buton gilsonite content. Moreover, the gilsonite belongs to natural asphalt and does not contain wax, and the antiaging ability of the modified asphalt is obviously improved. When used in engineering, some of the matrix asphalt and mineral powder can be replaced by pure asphalt and mineral particles contained in the gilsonite. According to this characteristic, the recommended range of the optimal blending capacity of the Butonite asphalt is 10%~20%. Based on the Beijing-based construction overhaul project, Dong et al. [16–18] comprehensively summarized the experience and effect of Buton gilsonite in practical engineering applications. The study shows that the high-temperature stability performance, low-temperature crack resistance, and water stability performance of the asphalt mixture are significantly improved. The Buton rock-modified asphalt mixture has the characteristics of simple construction process and easy operation and only needs to be crushed. When the temperature of the mixture is high, it is not necessary to increase the number of rolling passes, and good results are obtained.

Wang and Ge et al. [19, 20] evaluated the high-temperature dynamic rheological properties of Buton rock-modified asphalt cement with different dosages, including the main evaluation indexes such as phase angle, rutting

factor, dynamic viscosity, and storage modulus. The results show that the high-temperature performance of Buton rock-modified asphalt cement is obviously improved compared with the matrix asphalt. The resistance of the Buton rock-modified asphalt cement to the SBS-modified asphalt mortar is equivalent. The blending amount of Buton rock has a significant effect on the performance of the cement. The high-temperature performance of the asphalt cement is significantly improved when Buton gilsonite and matrix asphalt have the same quality and generally meets the requirements for pavement performance.

Liu et al. [21] used electron probe technology to detect that BRA asphalt mainly contains mineral elements such as C, O, S, Si, Mg, Al, Ca, K, and Fe. From the micromorphological structure analysis, the minerals with pore structure in the BRA asphalt are obtained, which are mutually inclusive and interpenetrated.

Zhang et al. [22, 23] have done a lot of research in order to study the interaction between asphalt and filler. The research shows that the finer the particle size of the filler, the larger the surface area and the stronger the interaction ability between asphalt and filler. The stronger the alkalinity of the filler, the stronger the interaction with the asphalt. The oxides such as SiO_2 and CaO contained in the gilsonite can increase the complex shear modulus of the asphalt compound but reduce the phase angle, explaining why gilsonite-modified asphalt and filler have strong adhesion.

In summary, the experimental study on the macroscopic composition and index of gilsonite-modified asphalt has important guiding significance for the application of gilsonite-modified asphalt in engineering.

2. Principle of Gilsonite Modification

Gilsonite is a kind of blend formed by high-viscosity pure asphalt and high-activity minerals under the combined action of long-term heating, oxidation, and geological changes. It is a bituminous material. Compared with ordinary asphalt, its content of asphaltene is more and has a very good affinity with aromatic hydrocarbons and saturated hydrocarbons in asphalt. Although the molecular weight of gilsonite is relatively large, in the high-temperature environment, the micelles will rupture, have certain activity, and be surrounded by the molecules of the matrix asphalt, so the gilsonite has good modification and comelting effect on the matrix asphalt.

3. Design of Test Scheme

The asphalt material used in this study is Buton gilsonite, and the matrix asphalt is Korean SK90# asphalt. According to the variation of the performance components of traditional polymer materials, through the orthogonal test, combined with the application results of performance indexes in asphalt engineering, the components and performance changes of two kinds of asphalt and modified asphalt are explored. At the same time, the advanced asphalt test method is used to study and analyze the gilsonite-modified asphalt. The design of the test scheme is shown in Tables 1 and 2.

TABLE 1: Experimental scheme of gilsonite and SK90[#] asphalt.

Project	Technical indicators	Unit	The National Standard for SK90 [#] Asphalt	
Performance testing	High-temperature stabilization performance	Softening point	(°C)	Not less than 42
	Low-temperature stabilization performance	Ductility (10°C, cm)	cm	Not less than 15
	Antiaging properties	Penetration (25°C, 5 s, 100 g)	0.1 mm	80~100
Microscopic mechanism test	Project	Component analysis	Test method	Four-component analysis
		Mesoscopic analysis		Thermogravimetric test
				Infrared spectroscopy
				SEM electroscope scanning test

TABLE 2: Experimental design of modified asphalt with different gilsonite contents.

Project	Technical indicators	Unit	Gilsonite content of modified asphalt (%)				
High-temperature stabilization performance	Softening point	(°C)	0	5	10	15	20
Low-temperature stabilization performance	Ductility (10°C, cm)	cm	0	5	10	15	20
Antiaging properties	Penetration (25°C, 5 s, 100 g)	0.1 mm	0	5	10	15	20
Component analysis	Thermogravimetric test		0	5	10	15	20
	Infrared spectroscopy						
Mesoscopic analysis	SEM electroscope scanning test		0	5	10	15	20

4. Experimental Data Processing and Analysis

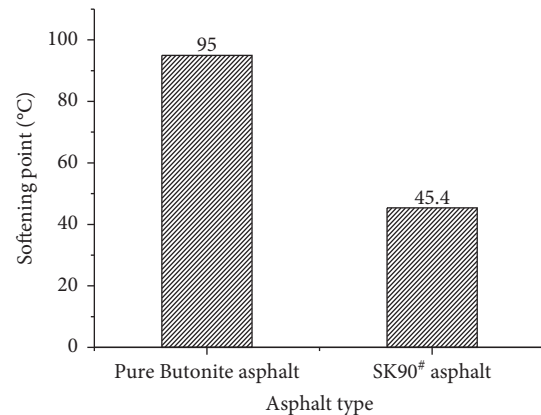
The modified asphalt composition of matrix asphalt, gilsonite, and different gilsonite contents was analyzed by thermogravimetric analysis, scanning electron microscopy, and infrared spectroscopy. And their performances were explained by combining the high- and low-temperature stabilization performance and antiaging performance test data.

4.1. Performance Comparison and Analysis of Pure Buton Gilsonite and SK90[#] Asphalt

4.1.1. High-Temperature Stability Test. Buton gilsonite contains a lot of minerals and hence the Buton gilsonite is purified to obtain pure Buton gilsonite. The high-temperature stability of the two asphalts is tested by softening point and the DSR dynamic rheological shear test at 64°C. In contrast, the test results are shown in Figures 1 and 2.

As can be seen from Figures 1 and 2, pure Buton gilsonite has excellent high-temperature resistance and its softening point is about twice that of SK90[#] asphalt, indicating that the asphaltene content is much more than the matrix asphalt content, and the pure Buton gilsonite anti-rutting factor is 22.7 times of SK90[#] asphalt; the phase angle is smaller than SK90[#] asphalt, indicating that the addition of pure Buton asphalt can effectively improve the high-temperature performance of asphalt.

4.1.2. Low-Temperature Stability Test. To evaluate the low-temperature extension performance of pure Buton gilsonite and SK90[#] asphalt, the ductility index test was used. The results are shown in Table 3.

FIGURE 1: Pure Buton gilsonite and SK90[#] asphalt softening point experiment.

As can be seen from Table 3, the pure Buton gilsonite is hard to be stretched at 10°C due to its hardness and brittleness. The Buton rock pure asphalt still cannot be stretched at 15°C testing temperature, indicating that the low-temperature performance of SK90[#] asphalt is better than that of pure Buton gilsonite, and the addition of the Buton gilsonite to the matrix asphalt can significantly reduce the low-temperature ductility of the asphalt.

4.1.3. Antiaging Performance Test. The short-term aging was simulated by a rotating film heating test (RTFOT), and the properties of pure Buton gilsonite and SK90[#] asphalt before and after aging were tested. The results are shown in Figure 3.

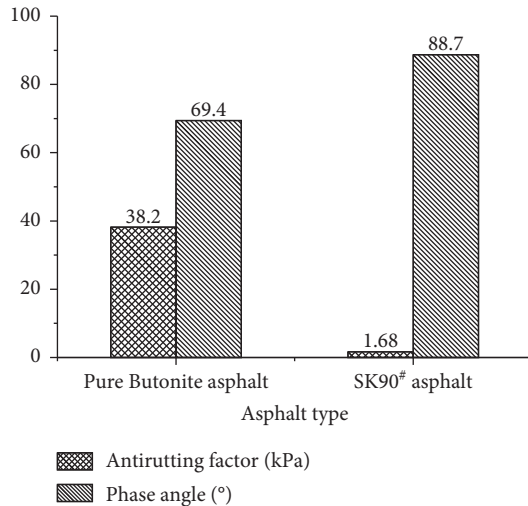


FIGURE 2: Pure Buton gilsonite and SK90[#] DSR experiment.

TABLE 3: Comparison of low-temperature performance between pure Buton gilsonite and SK90[#] asphalt.

Asphalt type	SK90 [#] asphalt	Pure Buton gilsonite
Ductility (10°C, cm)	18.29	Unmeasured

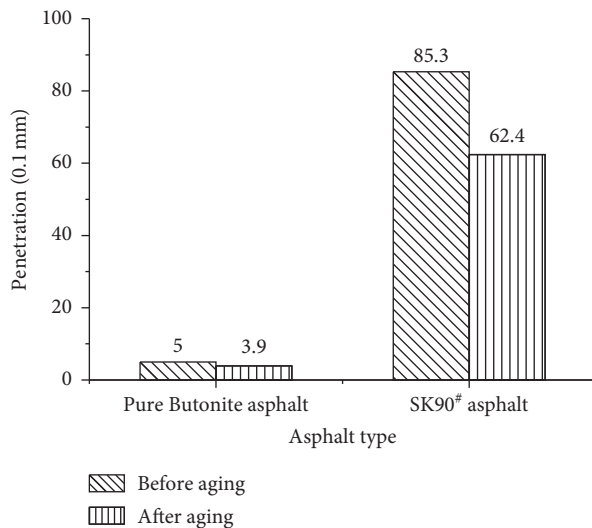


FIGURE 3: Comparison of antiaging properties between Buton rock pure asphalt and SK90[#] asphalt.

Figure 3 shows that the penetration index of pure Buton gilsonite before aging is 94.1% smaller than that of matrix asphalt. It can be seen that the pure asphalt of Buton rock has small penetration, high hardness, and strong resistance to deformation. After aging, the penetration degree of pure Buton asphalt and SK90[#] asphalt was reduced by 22% and 26.8%, respectively, indicating that the antiaging performance of pure Buton gilsonite is better than that of SK90[#] matrix asphalt.

4.1.4. Four-Component Analysis. A comparative test was conducted on four components of Butonite asphalt and matrix asphalt, and Table 4 was obtained.

It can be seen from Table 4 that the content of aromatic fraction, colloid, and asphaltene in Buton asphalt is higher than that of matrix asphalt, indicating that Buton asphalt has good high-temperature characteristics and excellent stability. At the same time, the content of saturated fraction is small, which indicates that the Buton gilsonite has high hardness, and adding the Buton gilsonite to the matrix asphalt can effectively improve the temperature-sensing property of the asphalt.

4.1.5. SEM Electroscope Scanning Test. The SK90[#] asphalt and Buton asphalt were observed by using a Prox scanning electron microscope to study the morphology and phase structure. The scanning electron microscope image is shown in Figures 4 and 5.

It can be seen from Figures 4 and 5 that SK90[#] matrix asphalt has good continuity and rheology. Pure Buton gilsonite is tightly wrapped on mineral particles, and many pore structures are formed between particles, which are beneficial to compatibility and adhesion of the matrix asphalt.

4.2. Comparative Analysis of Performance of Modified Asphalt with Different Gilsonite Contents

4.2.1. High-Temperature Stability Test. The high-temperature stability of modified asphalt with different gilsonite contents was tested by the softening point test and 64°C dynamic shear rheological test. The results are shown in Figure 6.

It can be seen from Figure 6 that the addition of gilsonite can significantly improve the softening point of the modified asphalt, and its high-temperature resistance is significantly enhanced; when the dosage is increased to 20%, the rutting performance is increased by 52.4%; the phase angle decreases with the increase in the amount of asphalt doping; that is, the viscosity is lowered, the elastic portion which can recover the deformation when the load is applied is increased, and the rut recovery ability is stronger.

4.2.2. Low-Temperature Stability Test. The 10°C ductility cannot be used to evaluate the relevant properties of pure gilsonite, but the gilsonite-modified asphalt has low pure gilsonite components, so it can still be used as an index to evaluate the low-temperature tensile properties of modified asphalt. The 10°C ductility of the modified asphalt with different volumes of gilsonite is compared and tested. The results are shown in Figure 7.

It can be seen from Figure 7 that the addition of Buton gilsonite greatly reduces the ductility of the modified asphalt compared with the matrix asphalt. As the blending amount of Buton rock increases, the decrease of the elongation tends to be flat, which indicates that addition of Buton gilsonite to the matrix asphalt makes the modification very sensitive to the effect of making it thicker and harder. The reason may be that the ash particles brought by the addition of Buton gilsonite can affect the uniformity of the asphalt; the

TABLE 4: Pure Buton gilsonite and SK90[#] asphalt four components.

Component	Saturated fraction (%)	Aromatic fraction (%)	Colloid (%)	Asphaltene (%)
Pure Buton asphalt	13.03	33.09	34.21	19.67
Matrix asphalt	35.6	32.1	17.5	14.8

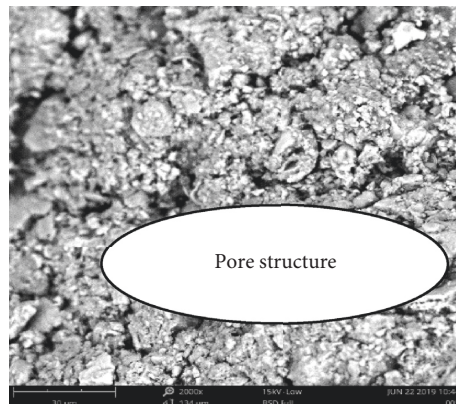
FIGURE 4: SK90[#] matrix asphalt.

FIGURE 5: Buton gilsonite.

phenomenon of stress concentration in the test ingress experiment resulted in a significant reduction in ductility. The results show that the incorporation of Buton gilsonite will reduce the low-temperature stability of the modified asphalt, and the optimum blending amount of Buton gilsonite should be in 9% to 15%.

4.2.3. Antiaging Performance Test. The short-term aging was simulated by a rotating film heating test (RTFOT), and the penetration of the modified asphalt with different amounts of Buton gilsonite before and after aging was tested. The results are shown in Figure 8.

It can be seen from Figure 8 that the addition of Buton gilsonite to the matrix asphalt can significantly improve the antiaging performance of asphalt, and the penetration of Buton rock-modified asphalt with 0%, 5%, 10%, 15% and 20% is reduced. The rates were 26.8%, 19.9%, 16.3%, 10.2%, and 4.7%, respectively. It can be seen that with the increase of Buton gilsonite content, the

antiaging performance of modified asphalt gradually increased.

4.3. Microtesting

4.3.1. SEM Electroscop Scanning Test. Scanning observation of modified asphalt samples with different contents of Buton gilsonite by using a Prox scanning electron microscope is shown in Figures 9~12 [12].

It can be seen from Figures 9~12 that when the Buton gilsonite is incorporated into the matrix asphalt, the overall morphology is uniform and smooth, indicating that the two asphalts have a highly stable fusion structure. The reason is analyzed. The Buton gilsonite contains some fine mineral particles, which are still in the form of particulate matter after being mixed with the matrix asphalt, and is evenly distributed in the modified asphalt. The particle size distribution of the unmelted gilsonite particles is in the range of 0~5 μm, and very few particles are between 5 and 10 μm.

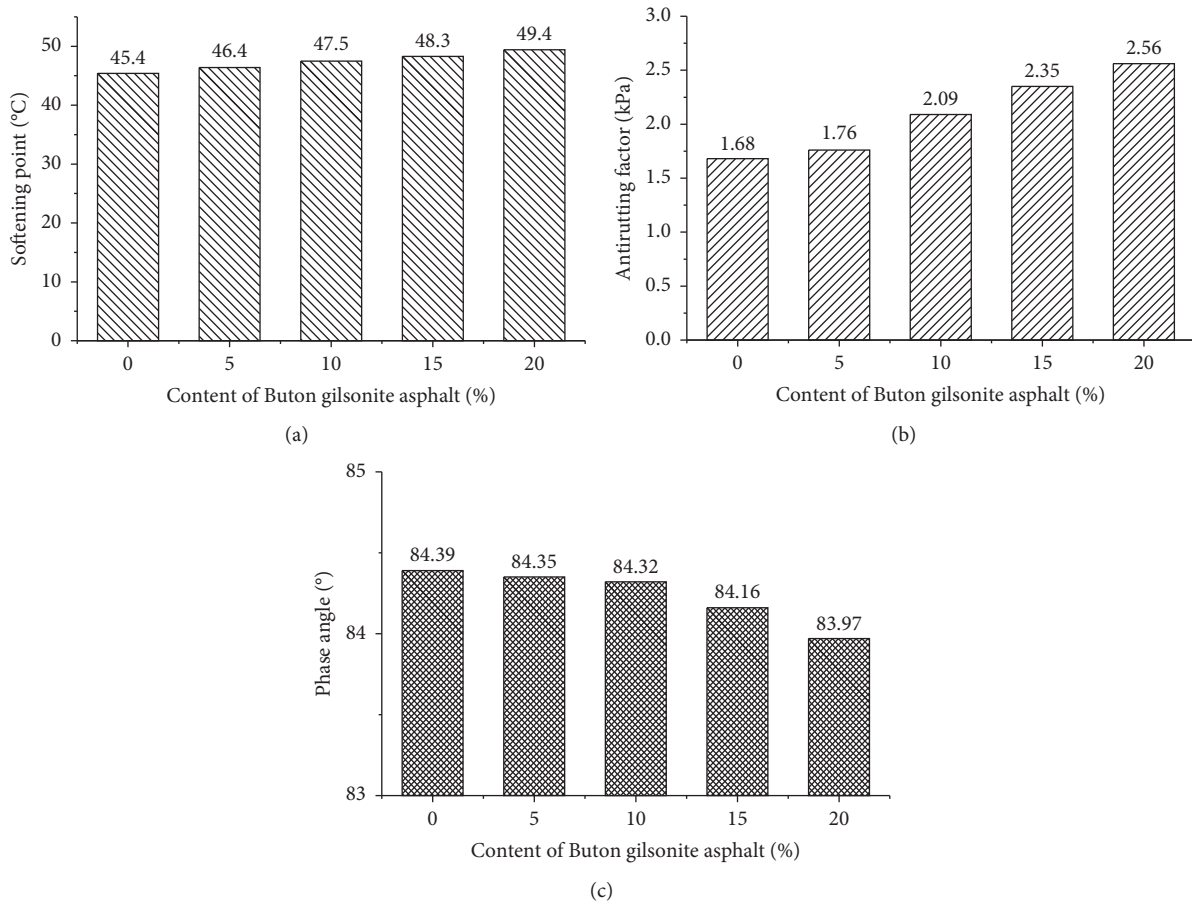


FIGURE 6: Comparison of high-temperature stability of modified asphalt with different gilsonite contents. (a) Softening point. (b) Antirutting factor. (c) Phase angle.

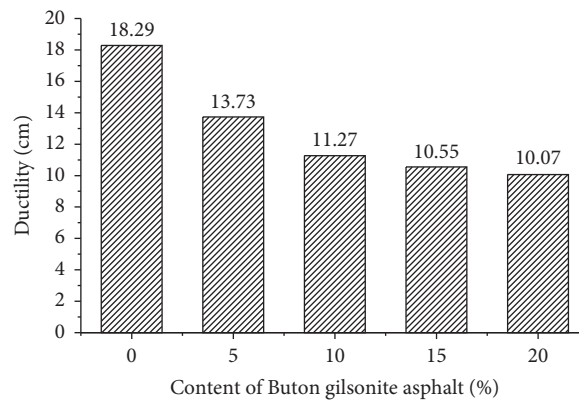


FIGURE 7: Comparison of low-temperature stability of modified asphalt with different gilsonite contents.

4.3.2. *Infrared Spectroscopy.* The chemical composition of Buton gilsonite, SK90[#] matrix asphalt, and modified asphalt with different contents of Buton gilsonite was analyzed by means of infrared spectroscopy. The results are shown in Figures 13 and 14.

Comparing Figures 13 and 14, it can be seen that both Buton gilsonite and Buton gilsonite-modified asphalt are mainly composed of carbonate, silica, or silicate, unsaturated carbon chain, carbonyl group, and amine

group. Compared with Buton gilsonite, there is no new absorption peak in the Buton rock-modified asphalt, and no large displacement in the peak position. There is no new absorption peak and large contrast between different dosages of Buton rock-modified asphalt. It can be seen that the addition of Buton gilsonite to the matrix asphalt does not produce new functional groups, and the addition of Buton gilsonite is a process of physical miscibility.

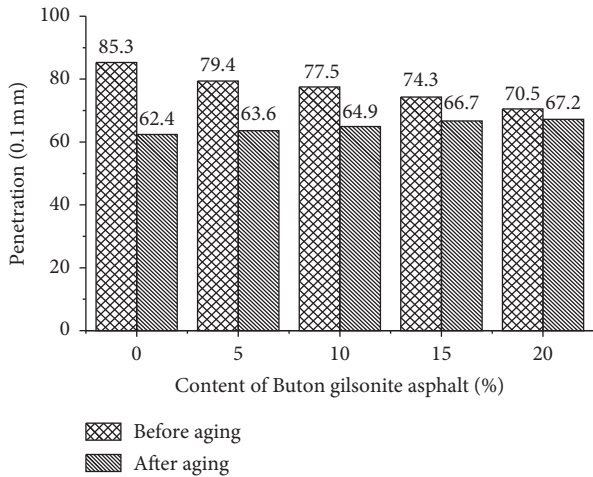


FIGURE 8: Comparison of antiaging properties of modified asphalt with different gilsonite contents.

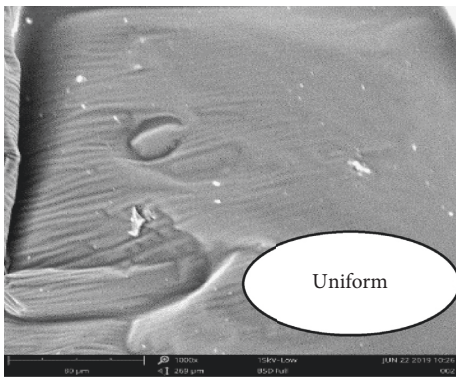


FIGURE 9: 5% of Buton-modified asphalt.

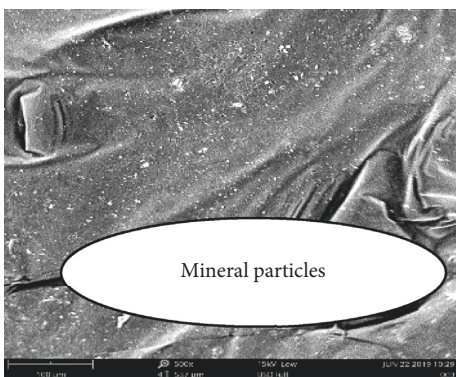


FIGURE 10: 10% of Buton-modified asphalt.

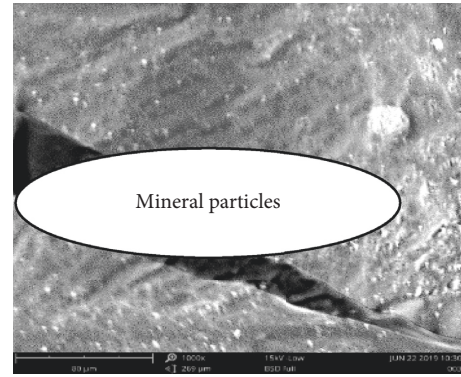


FIGURE 11: 15% of Buton-modified asphalt.

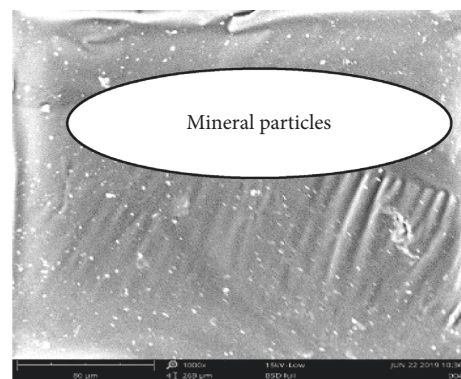


FIGURE 12: 20% of Buton-modified asphalt.

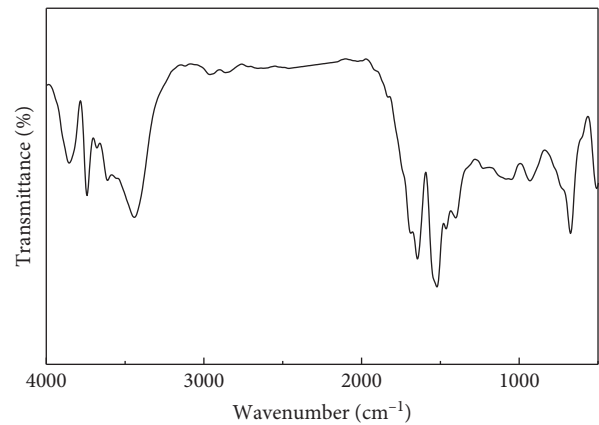


FIGURE 13: Infrared spectrum analysis of Buton gilsonite.

4.3.3. *Thermogravimetric Test.* The thermogravimetric analysis test was carried out on Buton gilsonite, matrix asphalt, and different amounts of Buton asphalt-modified asphalt. The test results are shown in Figure 15. It can be seen from Figure 15 that the Buton gilsonite has excellent high-temperature resistance, and the Buton gilsonite continues to lose weight slowly during the heat treatment. The weight loss

path of the matrix asphalt with different dosages of Buton rock-modified asphalt is consistent. The maximum weight loss rate occurs at 450°C during pyrolysis, and there is only one weight loss interval: 300°C~475°C. At this time, a large amount of pyrolysis occurs. The quality almost does not change after 550°C, and the remaining residue quality is 5% Buton-modified asphalt > 10% Buton-modified asphalt > 20% Buton-modified asphalt > SK90[#] matrix asphalt.

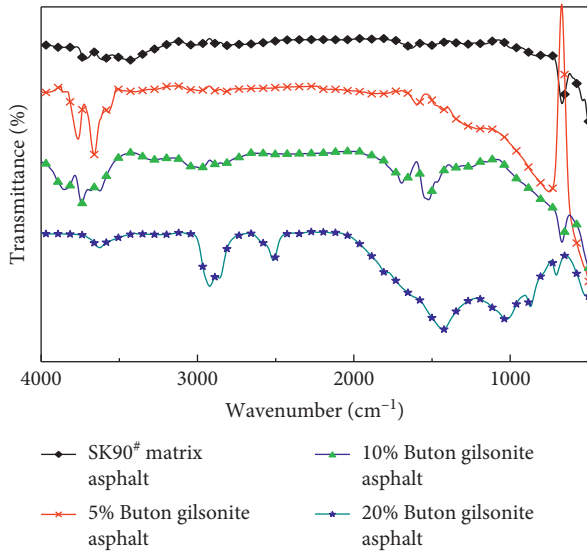


FIGURE 14: Infrared spectrum analysis of different contents of Buton gilsonite-modified asphalt.

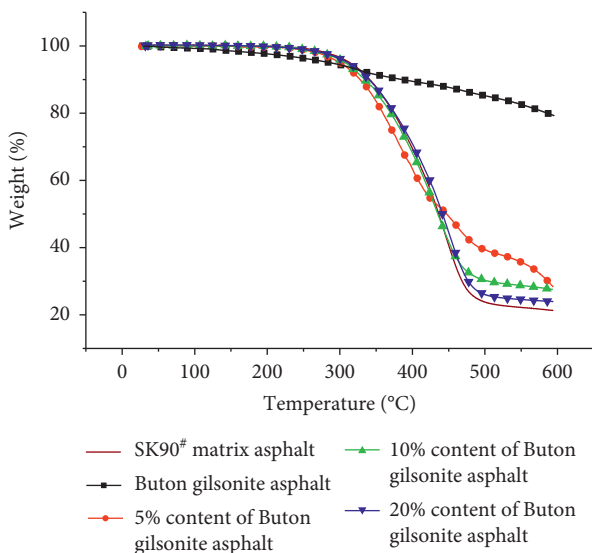


FIGURE 15: Thermogravimetric test analysis.

5. Conclusion

In this paper, the macroscopic and microscopic components and indexes of gilsonite-modified asphalt are studied. High- and low-temperature performance tests, antiaging performance test, four-component test, infrared spectroscopy test, and scanning electron microscopy test are carried out on gilsonite, matrix asphalt, and modified asphalt with different gilsonite contents. The research conclusions are as follows:

- (1) According to the four-component test, the content of aromatic fraction, colloid, and asphaltene in Buton rock pure asphalt is 3.1%, 95.6%, and 32.9% higher than that of the matrix asphalt, respectively, indicating that the gilsonite-modified asphalt has better high-temperature stability, antiaging properties, and better temperature-sensing properties.

- (2) The low-temperature stability test of modified asphalt with different gilsonite contents shows that the doping of Buton gilsonite will reduce the low-temperature stability of the modified asphalt. The optimum doping amount of Buton gilsonite should be 9%~15%.
- (3) Electron microscopy scan shows that the distribution of Buton asphalt is even. The scanning electron micrograph of Buton rock-modified asphalt shows that the particle size distribution of the unmelted gilsonite particles ranges from 0 to 5 μm and very few particles are between 5 and 10 μm .
- (4) Infrared spectroscopy and thermogravimetric analysis show that there is no new absorption peak and large displacement peak position in the comparison of different dosages of Buton rock-modified asphalt, and no new functional groups are produced. The addition of Buton gilsonite is a process of physical miscibility. The Buton gilsonite-modified asphalt has excellent high-temperature resistance, and the Buton gilsonite continues to lose weight slowly during the heat treatment.

Data Availability

All data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

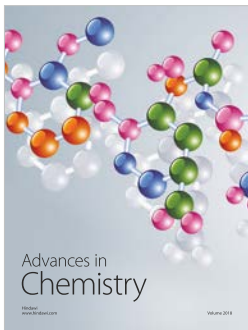
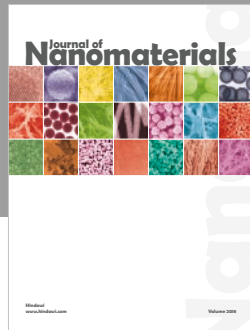
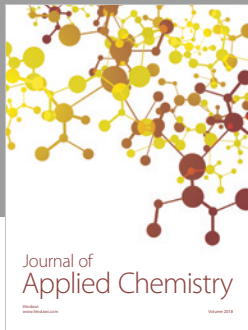
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