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# *Effect of Nano Modified Rubber on Physical and Rheological Properties of Asphalt*

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***Abstract:*** At present, asphalt pavement is widely used because of its smooth surface, large transportation capacity and convenient construction. However, in the long-term operation process, due to the complex environment, transportation load gradually increased and other reasons, a variety of diseases were caused. Due to its own composition and structural characteristics, it is easy to be softened at high temperature, brittle at low temperature, poor elasticity, anti-aging and other characteristics, and the maintenance cost in the project is high. It is necessary for highway maintenance personnel to improve the high temperature resistance of asphalt by adding various modifiers. The unique surface effect, size effect, quantum scale effect and quantum tunneling effect of nanoparticles make them have unique applications in many aspects. In recent years, people have done a lot of research work on traditional nano reinforced particles such as carbon black, white carbon black and clay, and developed some natural rubber nanocomposites with excellent performance. With the progress of science and technology, the application of new nano materials such as carbon nanotubes has greatly improved their performance in all aspects. It has become a research hotspot in the material field that the surface of rubber can be modified by nano particles and nano composite technology to obtain excellent comprehensive properties. This paper mainly studied the role of nano modified rubber in the physical and rheological properties of asphalt, and through the performance comparison and micro test analysis before and after modification, it was concluded that nano modified rubber can better promote the improvement of physical and rheological properties of rubber asphalt. It was found that the rutting factor of modified asphalt was increased by 10.49%, and the aging resistance of asphalt was also improved. The modification of nano rubber can not only expand its consumption market, but also significantly improve the performance of asphalt roads.

## 1. Introduction

With the development of nanotechnology and nanocomposites, the modification research of rubber has a new direction, the comprehensive properties have been improved, and the application scope has also been expanded. By using different nano materials to modify, new nano composite materials with special functions can be developed, which is an important development direction of rubber. As an excellent elastomer polymer material, rubber can improve the performance of asphalt, effectively improve the comprehensive use performance of asphalt, and improve the self repair ability of mixture.

Now there are relevant researches on nano modification by scholars. Karuppaiah M modified Mn2O3/NiO composite, and the results showed that the composite system had fast curing rate and physical and mechanical properties [1]. Benmansour Djazia Leila studied the laminated silicate/rubber composite system composed of styrene butadiene rubber and natural rubber, and analyzed the scale bending and dynamic characteristics of nano rubber in detail [2]. Rath Punyaslokl grafted the natural rubber damaged by high temperature and strong shear with carbon black by analyzing the cracking mechanism of rubber modified asphalt. The research shows that the in-situ grafting of carbon black can not only accelerate the vulcanization rate of rubber, but also improve its tensile, tensile stress, tear strength and other performance indicators [3]. Arabani Mahyar pretreated maleic anhydride and analyzed its kinetic performance evaluation. The test results show that the rolling resistance of NR can be effectively reduced by the pretreatment of carbon black with maleic anhydride [4]. Hung Albert studied the mechanical properties of natural rubber and found that elemental sulfur can affect the aging track and adhesion characteristics of rubber modified asphalt [5]. In a word, after nano treatment, its tensile strength, strength, hardness and tear strength have been significantly improved, and its comprehensive properties have been significantly improved.

Now there are some scholars' researches on rheological properties. Wang Haopeng has studied the rheological behavior of rubber chip modified asphalt containing warm mix additives, but has not comprehensively evaluated the rheological performance of modified asphalt [6]. Yetgin Salih Hakan pointed out that at present, there are many researches on the optimization of the production process and method of modified asphalt, while the evaluation of its rheological properties focuses on viscosity. He has less research on its rheological properties, so he lacks guidance on road performance [7]. Hu Zhongliang also found that there are few references to asphalt shear modulus, unrecoverable creep softness and nano rheological properties in the current specification, and the accuracy of viscosity test was questioned by some scholars [8]. Kim Hyun Woo has modified different base asphalt, and the study found that hydrocolloids can have different effects on rheological properties and printing properties [9]. Julianti Elisa would carry out tests on composite flour to test its function and rheological properties. Research shows that with the increase of modified composite flour, the expansion force of composite flour decreases and the rheological properties increase [10]. Therefore, the rheology experiment can provide a reasonable reference for the future asphalt research.

The nano modified rubber asphalt was studied from a new angle and its basic physical properties were measured. The rheological properties were measured with a dynamic shear rheometer. The preliminary results show that the mechanical properties of asphalt modified by nano materials can be significantly improved, and it is feasible both technically and economically.

## 2. Technical Progress of Rubber Modified by Nano Particles

(1) Nanometer clay

The unique structure and characteristics of layered silicate clay can be used to develop nano silicate/polymer nanocomposites with excellent performance, which is an important research direction of material science at present. Nano clay/natural rubber can be prepared in many ways. Adding nano clay into rubber can significantly improve its mechanical properties, heat resistance, barrier, flame retardancy and other properties. The results showed that the tensile strength, tear strength and hardness of the vulcanizate were significantly improved compared with those of the natural vulcanizate without additives [11].

(2) Nano carbon black

Nano carbon black shows excellent performance in reinforcement, coloring, conductivity, wear resistance, etc., as shown in Figure 1. It has been the most important and cheapest reinforcing agent and filler in rubber industry for many years. Carbon black can be added to rubber products, which can not only reduce the amount of rubber, but also achieve the same mechanical properties, and improve the wear resistance of products [12]. However, in general, when the filling amount of carbon black is more than 50 phr, its physical properties will become very good. Due to the small particle size, large amount and low dispersion of carbon black, it is easy to lead to low reinforcement of rubber compound. Therefore, how to improve its dispersion performance has become an important issue in the rubber industry, and the surface modification and pre-dispersion of carbon black are the current research focus. At present, some scholars have discussed the physical and mechanical properties of carbon black during granulation. It was found that after carbon black granulation, the mixing property of rubber was improved, and the torque and mixing heat generation of rotor were significantly improved. At the same time, it also improved the dispersion of carbon black in rubber, shortened the curing time, and significantly improved the mechanical properties [13]. In addition, the vulcanization characteristics, physical properties and oxidation resistance of carbon black reinforced rubber were also studied. It was found that the tensile strength and constant elongation stress of the curing chamber reached the maximum and the aging resistance was the best when the addition of Si69 was about 1/10 of carbon black.

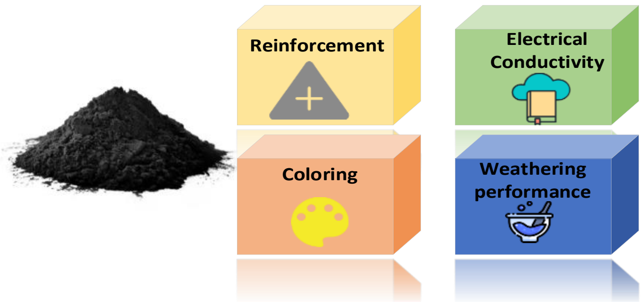


Figure 1: Nano carbon black has relevant characteristics

(3) Nano silica

Sodium silicate is an important lightweight filler in rubber industry, and its toughening effect is only inferior to that of carbon black. Sodium silicate can endow the rubber with excellent properties. However, due to its good surface polarity and strong hydrophilicity, the compatibility of silica gel with rubber molecules is not as good as that of carbon black. In addition, a large amount of silica gel is added to rubber, and the constant elongation stress, wear and mechanical processing properties are worse than that of carbon black [14]. At present, researchers have developed a new modified silica gel with cyclohexylamine solution as modifier. The results showed that the vulcanization property and strength of silica gel can be improved by modifying silica gel with cyclohexylamine, and the hysteresis property of silica gel after vulcanization is remarkable.

(4) Nanometer calcium carbonate

Nanometer calcium carbonate belongs to the category of new type calcium carbonate. Its application in natural rubber filling can improve the comprehensive properties of rubber, improve processing technology, and reduce production costs. It is an excellent white reinforcement material. It was found that the adhesion of modified nano calcium carbonate particles was better than that of natural rubber. When 40 phr of modified nano calcium carbonate was added to natural rubber, the tensile strength reached the maximum value, and the tensile permanent deformation of rubber compound increased with the addition of nano calcium carbonate, but the hardness changed little.

(5) Nanometer zinc oxide

Zinc oxide is an important vulcanization active substance of rubber, which can improve the crosslinking density of rubber, thus ensuring various physical and mechanical properties of rubber [15]. The results show that nano ZnO is a new functional material with excellent comprehensive properties. Relevant scholars have studied the vulcanization reaction of nano zinc oxide in natural rubber, and found that rubber containing zinc oxide has good resistance to vulcanization reversion to a certain extent, but nano zinc oxide has the effect of delaying scorching, and also improves the safety of rubber processing. Relevant scholars studied the properties of nano zinc oxide processing silica gel, and compared it with ordinary zinc oxide and nano zinc oxide. It is found that the addition of nano zinc oxide would reduce the scorching time and improve the mechanical properties. When the amount of nano zinc oxide is reduced to 3 phr, its physical and mechanical properties are the same as those of 5 phr zinc oxide, and the production cost can be reduced by using nano zinc oxide.

(6) Carbon nanotubes

Carbon nanotubes, also known as Bucky tubes, belong to the Fuller carbon system. They are seamless nanotubes wound by one or more layers of graphite sheets. Their ends are sealed with fullerene hemispheres. Because of its large length to width ratio, it is an ideal fiber for reinforced composites. Using carbon nanotubes to modify natural rubber, it can get nanocomposites with high strength, high density and good conductivity. Some scholars have studied the preparation methods and material properties of nano rubber composites. It was found that after the carbon nanotubes were compounded with rubber, the crystallization melting peak on the DSC (Dynamic Stability Control) curve of rubber decreased, the vulcanization reversion rate was reduced, and the scorching time was slightly shortened. The dispersion and interfacial bonding of carbon nanotubes in the dispersion bonding system have been improved. Its comprehensive mechanical properties have been improved. Carbon nanotube composites have good rebound and dynamic compression properties, and have good dynamic elastic and dynamic compression properties.

## 3. Main Factors Affecting Rheological Properties of Rubber Asphalt

(1) Factors of rubber powder

Rubber powder plays a basic role in rubber asphalt, which is closely related to three aspects: mainly rubber powder varieties, fineness of rubber powder, and amount of rubber powder, as shown in Figure 2. The bias ply tire made of natural rubber powder has good service performance, and the viscosity of the obtained rubber powder is relatively high, but there are significant differences in the fineness and dosage. In terms of engineering construction, the diameter of rubber powder particles is usually 0.15-0.60 mm, and the addition amount is 18-20%, which can ensure good comprehensiveness. In addition, the surface of rubber powder should also be treated. The adhesion between rubber powder and asphalt can be improved by microwave irradiation, so that it has better storage stability.

(2) Type of base asphalt

The degradation of rubber powder in asphalt is related to its solubility and the properties of matrix, and the content of polymer has a certain influence on the adhesive property of rubber compound. Through many tests, it is found that the quality of rubber asphalt obtained from different types of base asphalt is also different, indicating the influence of base asphalt on its performance. Through the comprehensive analysis of various technical indicators of base asphalt, it is found that the softening point, ductility, viscosity, etc. have a greater correlation with the base asphalt.

(3) Processing factors

Generally, when the asphalt temperature is high, its viscosity is low. However, with the increase of temperature, the asphalt itself would become more vulnerable. At the same time, at high temperature, the rubber powder would undergo desulfurization reaction, which would reduce the viscosity of rubber powder. It can be seen that in the production process, the relationship between rubber asphalt and temperature is not very big, but a scientific and reasonable temperature must be selected.

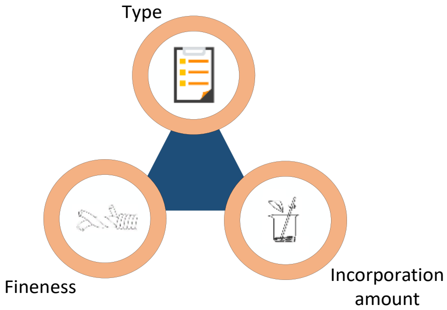


Figure 2: The rheological properties of asphalt are mainly affected in three aspects

There are two production methods of modified asphalt, one is direct injection method and premixing method. The working principle of direct input mode is to use rubber and asphalt mixture for treatment, and modify asphalt through rubber.

## 4. Preparation of Rubber Modified Asphalt

(1) Production of asphalt raw materials

A petroleum asphalt is used for the preparation of asphalt, and its performance test results are shown in Table 1. The PG (Performance Grade) is PG52, which shows that it has good high/low temperature resistance, that is, the average maximum temperature within 7 days is not more than 65 ℃, and the minimum temperature is not less than - 20 ℃.

Table 1: Technical indicators for petroleum asphalt testing

| Inspection items | Technical requirement |
|---|---|
| Softening point | >45(℃) |
| Penetration | 70-80(0.1MM) |
| Ductility | 15(CM) |
| Viscosity | 4(Pas) |
| Solubility | >97.5(%) |

(2) Preparation of nano modified rubber modified asphalt

First, the asphalt substrate can be heated to 150 ℃ to keep it flowing. On this basis, the required nano modified rubber powder is added to the asphalt to make its temperature reach 200 ℃, and then it is stirred by a high-speed shear machine to shear it within 2500 r/min. Then it adjusted the rotation speed to 4000 r/min, stirred and sheared for 1 hour. On this basis, the stirring shall be carried out for 10 minutes at the speed of 80 r/min to remove the residual gas.

## 5. Evaluation Method of Nano Modified Rubber for Asphalt Performance

(1) Evaluation method of physical properties of asphalt by nano modified rubber

Physical performance evaluation indexes of nano modified rubber for asphalt include softening point, penetration, ductility and viscosity, as shown in Figure 3.

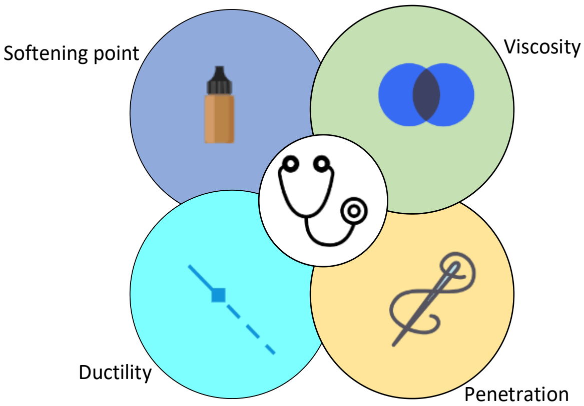


Figure 3: Four evaluation indexes of nano modified rubber for asphalt physical properties

1) Softening point

The so-called "softening point" refers to pouring molten asphalt into a ring of specified size to form, and scraping off the rest of the area beyond the ring. Then it fills a container with steel balls of preset weight, and then heats the water at 5 ℃/min to make it soft. The softening point of asphalt depends largely on the viscosity and temperature sensitivity of asphalt. If the softening point of asphalt is very high, the viscosity is large, the temperature stability is better, and the heat resistance is stronger.

2) Penetration

Asphalt penetration test is a widely used method to measure asphalt viscosity in the world. Penetration refers to the standard penetration depth in 0.1mm under specific load, temperature and action time. Penetration can reflect the viscosity of asphalt. Generally, the lower the penetration, the higher the viscosity and the harder the asphalt. On the contrary, with the decrease of viscosity, the penetration of asphalt would increase and the asphalt would become soft. With the decrease of penetration index, the sensitivity of asphalt to temperature increases accordingly. The penetration index also reflects the colloidal properties and structure of asphalt. When the penetration index is below - 2, its mechanical properties are similar to that of Newtonian fluid, while at low temperatures, asphalt shows greater brittleness. When the penetration is more than 2, its mechanical properties are greatly different from Newtonian fluid, and it has significant gelling characteristics. Therefore, the penetration index should be within a reasonable range. Too high or too low would affect the use of asphalt.

3) Ductility

Ductility reflects the tensile deformation of asphalt under external force, but there is no damage and fracture, that is, the length of time after asphalt fracture at a certain temperature and speed. The ductility of asphalt mainly depends on its own composition, that is, the volume ratio of each atom in the asphalt molecule. When the asphalt is uncracked, it can produce greater tensile strength. Most of the reasons are that the asphalt has a ring like and chain like chemical structure, and the position between the ring like and chain like chemical components would change significantly, so that the asphalt has a certain ductility. At the same time, the larger the molecular weight of asphalt, the stronger the interaction between its molecules, so that its tensile strength would also increase. Ductility also directly reflects the elasticity of asphalt. Generally, the ductility is relatively high. The greater the elasticity of asphalt, the better its crack resistance at low temperature.

4) Viscosity

Viscosity is the basic characteristic of asphalt, which reflects the friction between internal molecules of asphalt in the rheological process. The viscosity of asphalt should be controlled within a reasonable range. If the viscosity of asphalt is too high, its performance would be adversely affected. The low viscosity asphalt pavement is prone to rutting.

(2) Evaluation of rheological properties of asphalt by nano modified rubber

1) Viscoelastic generalized Maxwell model

The viscoelastic generalized Maxwell model is composed of multiple units. The modulus $E_i$ and viscosity $\mu_i$ in each Maxwell model are different. The relaxation time $\mathrm{T}_i$ of nano modified rubber can be expressed as

$$\mathrm{T}_i = \frac{\mu_i}{E_i} \tag{1}$$

The relaxation time of each Maxwell I model is different, and the relaxation modulus function of the generalized Maxwell model is:

$$E(t) = E_\infty + \sum_{i=1} E_i \mathrm{e}^{-t/T_i} \tag{2}$$

In the formula, $E_\infty$ is the static modulus of nano modified rubber, and the relaxation modulus $E(t)$ obtained in the Formula (2) can be respectively the storage modulus $E_s(\omega)$ and loss modulus $E_l(\omega)$ in the frequency domain:

$$E_s(\omega) = E_\infty + \sum_{i=1} E_i \frac{\omega^2 \mathrm{T}_i^2}{1+\omega^2 \mathrm{T}_i^2} \tag{3}$$

$$E_l(\omega) = \sum_{i=1} E_i \frac{\omega^2 \mathrm{T}_i^2}{1+\omega^2 \mathrm{T}_i^2} \tag{4}$$

The formulas of viscoelastic generalized Maxwell model in frequency domain are:

$$E_s(\omega) = E_0\left[1-\sum_{i=1} g_i\right] + E_0 \sum_{i=1} \frac{g_i \omega^2 \mathrm{T}_i^2}{1+\omega^2 \mathrm{T}_i^2} \tag{5}$$

$$E_l(\omega) = E_0 \sum_{i=1} \frac{g_i \omega \mathrm{T}_i^2}{1+\omega^2 \mathrm{T}_i^2} \tag{6}$$

$\omega$ is the angular frequency of nano modified rubber and $g_i$ is the dimensionless modulus expression of nano modified rubber

$$g_i = \frac{E_i}{E_0} \tag{7}$$

$E_0$ is the instantaneous modulus of nano modified rubber, which is consistent with the quasi-static modulus $E_\infty$ of nano modified rubber as follows:

$$E_0 = \frac{E_\infty}{1-\sum_{i=1} g_i} \tag{8}$$

The above formula can reflect the stress relaxation, creep and mechanical properties of rubber. Under each cycle load, the energy loss of viscoelastic material per unit volume due to delay can be expressed by the following formula:

$$\Delta W = \int_0^{2\pi/\omega} \sigma(t)\frac{d\varepsilon(t)}{dt}dt \tag{9}$$

$\sigma(t)$ is the dynamic stress amplitude of modified rubber asphalt, and $\varepsilon(t)$ is the dynamic strain amplitude of modified rubber asphalt.

2) Dynamic shear rheology test

According to the research of highway research project, the application of asphalt must be combined with relevant technical specifications, and the stability and medium temperature of asphalt must be improved under high temperature. In this process, a dynamic shear rheometer must be used for testing. (G) and (δ) can be used to express the composite shear modulus and phase angle of asphalt binder, showing the viscoelastic properties of asphalt binder. G is the measured value reflecting the overall resistance of materials in the process of repeated shear deformation, which is composed of two aspects. The real number part stores the first part of the elastic modulus G '. In the second section, G ″ is used to express the memory modulus of real part and the elastic modulus of imaginary part loss. δ Generally, it represents the cost of adhesion of materials. When the material is under the input of metaphysical stress, it cannot occur simultaneously with the reaction of normal metaphysical stress.

The relationship among G ′,G ″and δ is as follows

$$G^{'} = G\cos\delta \tag{10}$$

$$G^{''} = G\cos\delta \tag{11}$$

$$\tan\delta = \frac{G^{''}}{G^{'}} \tag{12}$$

The SHRP specification defines G/sinδ as the rutting factor. The higher the value, the better the permanent deformation resistance of asphalt.

## 6. Influence of Nano Modified Rubber on Asphalt Performance

(1) Influence of nano modified rubber on physical properties of asphalt

1) Ductility test

The influence of nano modified rubber content on asphalt ductility is shown in Figure 4. The experimental results show that when the amount of nano modified rubber powder is increased, its ductility generally shows a decreasing trend.

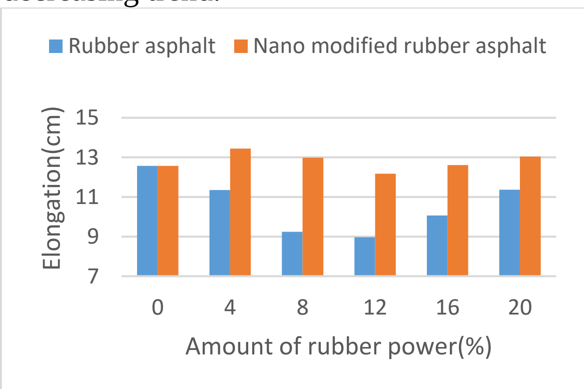


Figure 4: Influence of modified rubber content on asphalt ductility

Because the light oil in the rubber powder system is swelled and absorbed after adding asphalt, its toughness, plasticity and brittleness are obviously reduced. At the same time, rapid tension makes the tensile area in the middle of the sample continuously narrow, while at the interface, stress concentration is easy to occur, and the stress is difficult to dissipate, resulting in low-temperature embrittlement. In addition, the ductility of modified rubber asphalt is low, which is mainly because the modified rubber and its surface polar groups have good adsorption capacity. In the same case, asphalt has higher oil content and lower plasticity, while in the experiment, it has higher adhesive strength with asphalt. Therefore, there would be greater stress concentration in the experiment, so that the ductility of modified rubber asphalt is lower than that of rubber asphalt.

2) Penetration test

The influence of the content of nano modified rubber on the penetration of asphalt is shown in Figure 5. When the content of rubber powder increases, the penetration decreases significantly. The results show that the viscosity of asphalt increases and asphalt tends to harden after adding rubber powder. The addition of nano modified rubber reduces the penetration of asphalt. This is because the interface structure layer composed of nano modified rubber and asphalt has high shear strength, which can resist a wide range of shear stress and deformation. The results show that the adhesive strength of modified rubber and asphalt is higher, and the chemical bond with asphalt is closer. At the same time, the content of oil and resin decreases, which increases the viscosity and hardness of modified asphalt, thus reducing the penetration under the same conditions.

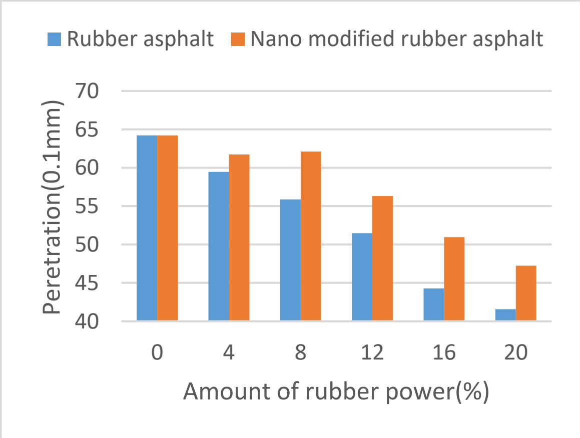


Figure 5: Influence of nano modified rubber content on penetration of asphalt

3) Softening point test

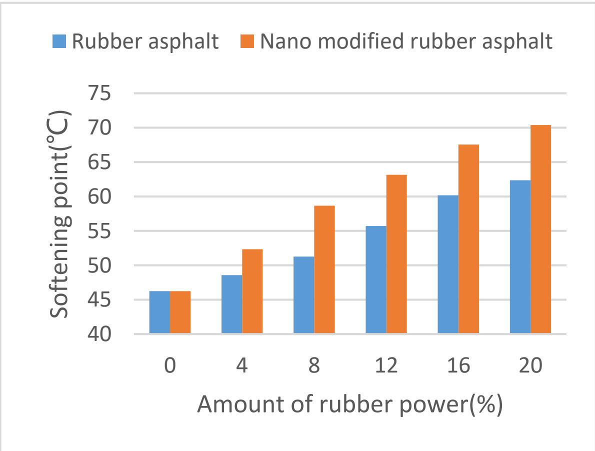


Figure 6: Influence of nano modified rubber content on softening degree of asphalt

The influence of the content of nano modified rubber on the softening point of asphalt is shown in Figure 6. The experimental results show that the softening point of asphalt increases after the addition of modified rubber. With the increase of the amount of rubber powder, the growth rate of its softening point decreases gradually, which indicates that the more rubber powder is added, the lower the sensitivity of softening point to rubber powder.

Compared with the unmodified rubber, the nano modified rubber asphalt has a greater role in improving the softening point, and has a stronger ability to improve the asphalt's resistance to high temperature permanent deformation. Through chemical bonding with asphalt, the relative content of asphaltene in asphalt increases, making it hardened. The increase of softening point is an important reason for the increase of sample density. After the asphalt is softened, the sphere falls. Because there is a continuous interface structure between the sphere and the asphalt, the deformation of the sphere on the asphalt can be further prevented, so as to maintain the high-temperature stability of the asphalt and extend its falling time.

4) Effect of viscosity

The influence of nano modified rubber content on asphalt viscosity is shown in Figure 7: the results show that the mixing and compression temperature of asphalt mixture can be effectively reduced by adding nano modified rubber within a certain range. However, due to different types of asphalt, their own characteristics change, and the degree of viscosity decline is also different. It shows that nano modified rubber has a certain softening effect on aging asphalt, but a certain amount must be controlled, otherwise the rutting resistance of aging asphalt would be reduced. In order to ensure the adhesion between asphalt and aggregate, its viscosity must be reduced to the minimum, so the amount of nano modified rubber should not be too large.

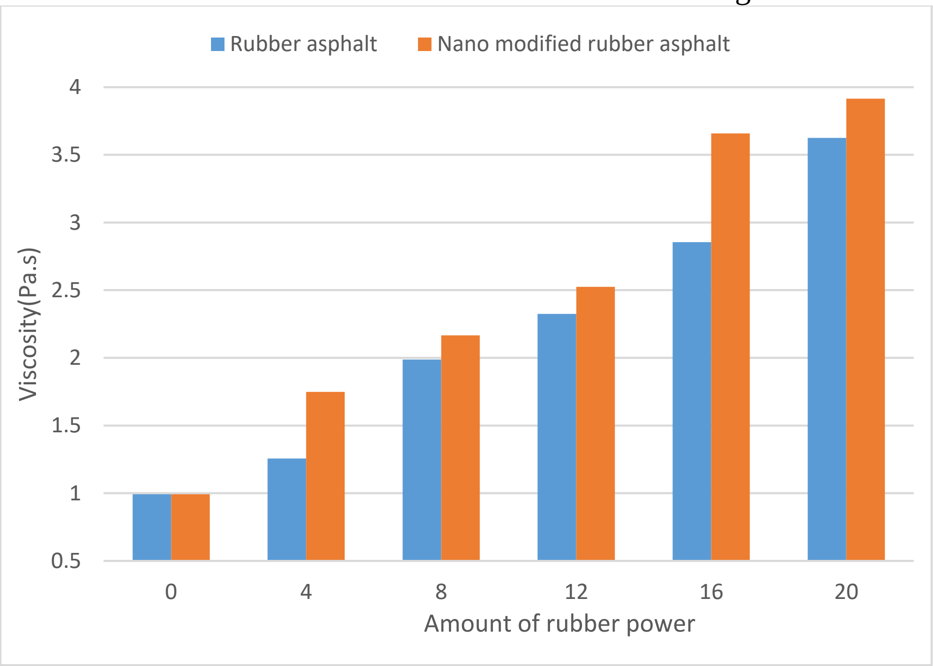


Figure 7: Influence of nano modified rubber content on asphalt viscosity

(2) Effect of nano modified rubber on rheological properties of asphalt

Asphalt is a viscoelastic material, and its viscoelastic properties vary with time and temperature. The effects of composite shear modulus and phase angle on asphalt were measured by dynamic shear rheometer. The phase angle refers to the ratio between the loss elastic modulus and the storage elastic modulus in the composite shear modulus, which can be used as an indicator of stress hysteresis. The phase angle is the viscoelastic property of asphalt. With the increase of viscosity, the elasticity decreases and the viscosity increases. The phase angle changes of rubber asphalt and modified rubber asphalt at different temperatures are shown in Figure 8. The results show that the phase angle of rubber asphalt increases monotonously with the increase of temperature under different temperature conditions. Compared with ordinary rubber asphalt, the phase angle of nano modified rubber asphalt is obviously increased at the same temperature.

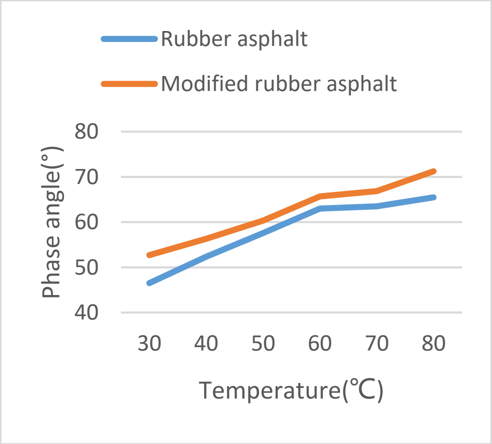


Figure 8: Phase angle change of rubber asphalt and modified rubber asphalt at different temperatures

The influence of nano modified rubber powder on asphalt rutting factor is shown in Figure 9. Compared with the unmodified asphalt rutting factor, the modified asphalt rutting factor has increased by 10.49%, indicating that nano modified rubber can significantly improve the rutting resistance of asphalt.

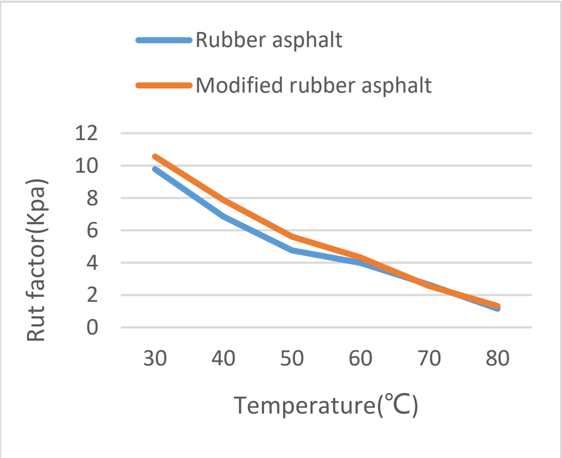


Figure 9: Influence of nano modified rubber powder on asphalt rutting factor

## 7. Conclusions

In this paper, after a brief introduction of the development of nano modified rubber and its influence on its properties, the physical properties such as softening point, penetration, ductility and viscosity are compared and analyzed, and the rheological properties of nano modified rubber on asphalt are studied by using dynamic shear rheology method. The results show that the softening point, stitch length, ductility and rutting coefficient can be significantly increased by adopting the improvement measures. Although there have been many experiments and theoretical analysis, due to the limitations of its own technology, time, environment and other factors, it still needs further discussion.

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