Progress in combined modification of rubberized asphalt

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Abstract. The traditional rubberized asphalt has various disadvantages such as low resistance to rutting, difficulty in stabilized during manufacture, poor fluidity during processing and pollution. Combined modification of rubberized asphalt is reviewed in this paper for developing an effective way to resolve the above mentioned problems. According to the chemical properties of rubber asphalt composite modifiers, it is divided into four types: polymer, inorganic substance, plasticizer and other four types. The reasons for the incorporation of each type of composite modifier and the current research and application status at home and abroad are introduced. The advantages and disadvantages and modification mechanisms of various composite modifiers are reviewed. It is believed that the existence of chemical cross-linking network of rubber powder brings challenges to the compounding process and effect evaluation. Nevertheless, compound modification and environmentally friendly processing of multiple modifiers under the premise of green de-crosslinking is the future development direction of rubber asphalt.

1. Introduction
Rubber asphalt is a kind of binding material obtained by grinding waste tires into hot asphalt, heating and stirring for a certain period of time, and the swelling or degrading. Rubber asphalt not only has excellent low temperature performance, anti-aging performance[1], but also can consume waste tires in large quantities, in line with the requirement of ecological economy. However, compared with SBS, PE and other modified asphalt, Rubber asphalt has five disadvantages: poor rutting resistance at high temperature; required higher temperature, high energy consumption, heavy pollution; easy segregation, poor thermal stability, construction difficult to control; application of the high cost[2-4]. The reason is that rubber power has a chemical cross-linking network, and the swelling and degradation of the rubber power are always occurred during the interaction between the rubber power and asphalt, which is difficult to control; in addition, the rubber power has a larger specific gravity than asphalt and is easy to precipitate[4]. The compound modification technology of rubber asphalt has become an effective way to solve the above problems. This article starts with four types of composite modification methods of polymers, inorganics, plasticizers and other categories, summarizes the research status at home and abroad, and explains its development trends, in order to develop high-performance, multi-functional and low-cost new rubber asphalt materials, to meet the new demand of green transportation development in China.
2. Polymer modified rubber asphalt

Because of the existence of chemical crossing network structure in rubber powder, its compatibility with asphalt is low, so it is difficult to form network structure in asphalt, relatively SBS, PE and other modified asphalt, and its high temperature rutting resistance is insufficient. General polymer modifiers include thermoplastic elastomers, thermoplastic resins, thermosetting resins and rubbers. Commonly used thermoplastic elastomers are styrene-butadiene-styrene block copolymer (SBS), polyolefin elastomer (POE) and polytrans cyclooctene (TOR); thermoplastic resins include polypropylene (PP), polyethylene (PE) and polyvinyl chloride (PVC), etc. thermosetting resin is mainly epoxy resin (EP); rubber is mainly styrene butadiene rubber (SBR), etc. Among them, SBS and PE modified rubber asphalt research is the most extensive.

2.1. Thermoplastic elastomer SBS

The composite modification of SBS and rubber powder can make SBS network embedded around the rubber powder to form a double network structure, which effectively improves the rutting resistance of the composite modified rubber asphalt. The research and application of SBS modified rubber asphalt at home and abroad focus on the types and dosage, preparation process, durability impact and economic evaluation of SBS and rubber powder.

In terms of mixing amount and type, on the one hand, the influence of rubber powder mixing amount and mesh number on the performance of modified asphalt when the SBS mixing amount is a fixed value\cite{5, 6}. On the other hand, the rubber powder content is used as a fixed value\cite{7, 8} to study the effect of SBS content and type on the performance of modified asphalt. In some publication\cite{9, 10}, the researchers studied the change of asphalt performance when the content of SBS and rubber powder were both variable. In the above study, the value range of the SBS content is not much different, but the value range of the rubber powder content has a large gap. When the construction conditions permit, the amount of rubber powder and SBS should be increased as much as possible.

In terms of preparation process, SBS main processes of modified asphalt include swelling, shearing and development, in which the shearing process includes two stages: high speed shearing and low speed stirring, while the rubber powder is divided into stirring swelling and degradation dissolution according to its dispersion degree in asphalt. According to the different swelling and degradation states of rubber powder in asphalt, it can be divided into three processes: swelling mainly; partial dissolution; complete dissolution. Among them, the SBS is mainly compounded with the partial dissolution or complete dissolution process, which can jointly improve the mechanical properties of modified asphalt.

In terms of durability effects, many studies have shown that rubber powder improves the aging resistance of SBS modified asphalt. Li\cite{11} obtained the composite asphalt with good anti-aging performance through the rotating film oven aging test (RTFOT), analysis of its mechanism for the powder and SBS first degradation, and then with a small amount of components in the asphalt reaction, resulting in reduced aromatic hydrocarbons and resin content in the asphalt, asphalt and toluene insoluble content increased, thereby improving the aging resistance of asphalt. Tian\cite{12} compounded the reactive terpolymer RET and rubber powder, and the results showed that the combination of RET and rubber powder can improve the fatigue resistance of asphalt pavement to a certain extent.

In terms of economic evaluation, Cui\cite{13} and Li\cite{14} believed that although the unit price of SBS modified rubber asphalt increase, it can extend the service life, reduce maintenance and repair costs, save operating costs, and have good application prospect. In contrast, Hu\cite{7} and Yang\cite{15} stated that its increased cost is not conducive to actual engineering, especially the use of low-grade pavements.

2.2. Other thermoplastic elastomers

Other thermoplastic elastomers include polyolefin elastomers (POE), polytrans cyclooctene (TOR) and atactic polypropylene (APP), etc.

Xie\cite{16} studied the influence of the mixing ratio of POE and rubber powder on asphalt, and obtained that POE modified rubber asphalt can be added with compatibilizer to obtain good thermal stability
and aging resistance. To improve the performance of rubber powder modified asphalt, Liu\cite{17} obtained through SEM, DSC and FTIR tests that the addition of TOR increase the interaction of rubber asphalt, and reacted with rubber powder to change the rheology of asphalt and improve storage stability. Wang\cite{18} studied the influence of APP content on the high temperature performance and elastic recovery performance of rubber asphalt, and concluded that the quality content of APP is 15% and the performance is the best.

2.3. Thermoplastic resin
Different from SBS of the elastomer class and rubber powder composite have synergistic elastic superposition effect, thermoplastic resin composite modification of rubber asphalt is easy to reduce the elasticity of rubber asphalt. However, thermoplastic resin is easy to swell in asphalt or asphalt mixture, forming resin reinforced network, which obviously improves the rutting resistance of rubber asphalt.

Thermoplastic resins mainly include polypropylene (PP), polyethylene (PE) and polyvinyl chloride (PVC). Among them, PVC has the defects of high temperature processing degradation and environmental pollution, so it needs to be used with caution. Chen\cite{19} studied PP and PVC modified rubber asphalt mixed with 20% PE and determined the best ratio and process. Zhu and others\cite{20-24} studied PE modified rubber asphalt. The literature\cite{20,21} through the study of the properties of modified asphalt and mixture, got the best quality PE content is 4%. Zhu\cite{22} further studied the performance of PE and rubber powder composite modified asphalt mixture, and obtained that the high temperature asphalt, low temperature crack resistance and water stability of the composite asphalt mixture were significantly improved. Huang\cite{25} obtained through the fatigue performance test, the self-healing performance of PE and rubber powder composite is not as good as SBS and rubber powder composite, but better than rock asphalt and hard asphalt. Chen Peifeng\cite{21} and Lu Libo\cite{24} studied the properties of high density polyethylene (HDPE) and low density polyethylene (LDPE) modified rubber asphalt, respectively.

PE and PP are mostly used as "dry method" in the form of anti-rutting agents, and there are many experimental studies and engineering applications. The mixture of rutting agent and crumb rubber was studied in Ref.\cite{26-29}. It was found that this method had higher high temperature stability and water stability than SBS and crumb rubber compound asphalt. At the same time, Fu\cite{28} proposed the best combination ratio of anti-rutting agent and rubber powder.

2.4. Thermosetting resin
Because of the unique bond and high temperature resistance of epoxy asphalt, it is widely used in steel deck pavement, but the aging resistance and fatigue resistance of epoxy asphalt bring about cracking problems, which leads to the shortening of life and has begun to attract people's attention. It has great potential to improve the durability by compounding it with rubber asphalt.

Epoxy resin and high durability paving (HDP) are the only thermosetting resin materials used to modify rubber asphalt. Epoxy resin can improve the thermal stability of modified asphalt and the high-temperature rutting resistance of mixture in Ref.\cite{30}. Gao\cite{31} studied the effect of HDP additive content on the properties of rubber-modified asphalt and mixtures. Through mechanism analysis, it was found that the main agent and hardener in HDP had an autocatalytic reaction, and the optimal HDP content was determined to be 20%.

3. Inorganic modified rubber asphalt
Inorganic modified asphalt is widely used, including cement, diatomite and nano-scale inorganic materials, because it can significantly improve the bonding and high-temperature performance of asphalt and aggregate.

The earliest research on inorganic modified rubber asphalt was the use of cement and slaked lime to improve its water resistance. Among them, there are relatively many studies on the combination of cement and rubber powder\cite{32,33}. Through preparation of mixture by composite modified asphalt of cement glue powder, it is concluded that rubber powder can improve the bonding performance
between asphalt and aggregate, and cement can enhance the anti-stripping performance, water temperature performance and high temperature of mixture. Other research on inorganic modified rubber asphalt also includes natural mineral materials and nano materials. Zhao\cite{34} studied the compound modification of diatomite rubber powder, the high temperature stability, low temperature crack resistance, water stability and fatigue resistance of asphalt were improved. Wei\cite{35} studied the frost resistance of rubber powder and diatomite modified asphalt by designing a test device by himself. Zhang\cite{36} studied the compound modification of rare earth rubber powder to improve the ultraviolet absorption capacity of asphalt and determine the optimal rare earth content.

Over the past years, various authors studied modified rubber asphalt with inorganic nano materials. Organic montmorillonite (OMMT) improves the high temperature storage stability of asphalt, Na-MMT reduces the storage stability of asphalt, and the addition of OMMT can improve the aging performance and dynamic viscosity of asphalt\cite{37}. Nano ZnO can improve the anti-aging properties of rubber asphalt and thermal storage stability\cite{38, 39}.

Inorganic modification is mostly to study the influence of a certain mineral and its content on the performance of asphalt and mixture, so as to judge whether the mineral is suitable for rubber powder modified asphalt. Except for the composite modification of cement, the other inorganic modification is not a system, the materials studied are scarce and expensive, so the research value needs to be further considered. If the existing mineral waste materials, such as lithium slag, fly ash, silicon powder, steel slag and so on, are used to modify rubber asphalt, it will be more valuable for research.

4. Plasticizer modified rubber asphalt

Plasticizer is an important processing aid for polymeric materials. Plasticizer molecules can be dispersed and embedded between polymer molecular chains to increase the distance between molecular chains, thereby enhancing the flexibility of the material, improving processing fluidity, and reducing processing temperature. Regarding plasticizer-modified rubber asphalt can be divided into petroleum-based modification and bio-oil-based modification.

In terms of petroleum plasticizer modified rubber asphalt, aromatic oil compound rubber asphalt has been studied most. Tan\cite{40} significantly improved the elastic energy storage capacity of asphalt and counteract the effect on composite modulus. In order to make the rubber powder and SBS more compatible, Huang C. et al.\cite{41-45} studied the blending of SBS and aromatic oil to modify rubber asphalt. Among them, Huang\cite{41} added furfural extraction oil during the mixing process to prepare blended asphalt, Cheng\cite{42} mixed grafting rubber powder and furfural extract oil into the asphalt to swell, but Xia\cite{43} first blended furfural extract oil with rubber to pretreat the rubber powder to improve the dispersion ability of the rubber powder. Ding\cite{44} prepared SBS modified asphalt by adding rubber powder into SBS and extracted oil, while Wei\cite{45} prepared modified asphalt by mixing rubber powder with SBS and then adding asphalt and aromatic oil mixture.

Aromatic oil and rubber powder have good compatibility, which can reduce the preparation temperature of rubber asphalt, promote the swelling reaction of rubber powder in asphalt, further increase the amount of rubber powder, reduce energy consumption, and reduce pollution. However, the modification of aromatic oil does not change the chemical structure of rubber powder and asphalt, and cannot fundamentally solve the disadvantages of rubber asphalt, such as high viscosity and easy segregation.

Plasticizer and rubber have good chemical compatibility. Combining the two in asphalt can not only use waste tires in large quantities, but also reduce the production temperature and viscosity of rubber asphalt, and improve the fluidity and low temperature performance of asphalt. However, the research on plasticizer-modified rubber asphalt is still in its infancy. Difficulties such as many types and complexity have limited its application and development to a certain extent, but it still has great development potential.
5. Other modified rubber asphalt

In addition to the above three types of modified rubber asphalt, some scholars have studied some rarer substances and rubber powder modified asphalt, which we classify as other modified rubber asphalt. In order to further promote the degradation of rubber powder, Liu [46] studied the compound modification of garlic powder and rubber powder, obtained garlic powder with good viscosity reduction effect, and determined the optimal dosage. Li [47] studied the compounding of polyphosphoric acid and rubber powder, and obtained that polyphosphoric acid would react with asphalt, thereby greatly improving the high temperature stability and anti-aging performance of modified asphalt, and determining the optimal compound ratio. Ling [48] studied the effect of an anti-ultraviolet material-Mg-Al double metal hydroxide (LDH) on the short-term and long-term aging performance of rubber asphalt. LDH can slow down the formation of carbonyl groups in asphalt and improve its anti-aging performance.

Other modified rubber asphalt have gradually attracted attention. The main reason is that adding a small amount of modified in rubber asphalt can achieve a more significant effect, and solve some of the shortcoming of rubber asphalt. However, there is no fixed type of these modifiers, and the mechanism research is still in the process of development. The experimental data obtained cannot be compared with the research results of others.

6. Summary and outlook

Compound modified rubber asphalt has become the development trend of modified asphalt, and the asphalt which can meet the needs of practical road engineering can be prepared by making use of the advantages of various modifiers. The contents are summarized as follows:

1) polymer modification can improve the disadvantages of rubber asphalt, such as poor rutting resistance, easy segregation, and poor thermal stability. It can provide a basis for the factory supply of modified asphalt and the use of waste plastics can also reduce construction costs. However, higher temperatures are still required for processing when adding polymers, resulting in pollution;

2) Inorganic modified rubber asphalt mostly improves the specific properties of rubber asphalt. For example, the compound of rubber powder and cement can improve the water resistance of asphalt, and the compound with rare earth can improve the aging resistance of asphalt, but basically does not improve the storage stability of asphalt and high temperature anti-rutting performance, high temperature required for production and high energy consumption;

3) Plasticizer modified rubber asphalt improves the fluidity and low temperature performance of asphalt, promotes the use of large amounts of rubber powder, thereby reducing construction costs, lowering production temperature, reducing energy consumption and pollution, but it does not improve the high-temperature rutting resistance of asphalt;

4) There are fewer studies on other types of modification, mostly to improve a certain performance of rubber asphalt, and the expected effect can be achieved when the amount is small. However, there are many types of modifiers, and the research has not formed a system.

The above four types of compound modified rubber asphalt can solve the disadvantages of traditional rubber asphalt. Each has its own characteristics, but cannot completely solve all the shortcomings. In order to obtain the advantages of the above two or more crumb powder modified asphalts, blending modification (the composite of non-single material and rubber powder) is a hot spot for current and future research. Blending modification can synthesize the performance of various modifiers, improve the anti-segregation and thermal stability of rubber asphalt in all aspects, reduce high-temperature viscosity, improve the anti-rutting performance of the mixture, and provide a basis for the factory application of modified asphalt. However, the increase in the types of modifiers has led to the complexity and diversification of the asphalt preparation process. Therefore, the formation of standardized preparation standards and the improvement of the cost performance of modified asphalt are problems that need to be solved urgently for blending modification.

In order to meet actual engineering applications, it is often not enough to study modified asphalt alone. Some scholars study modified asphalt mixtures, that is, mixing asphalt, stone and modifiers together, such as fiber-modified rubber asphalt mixtures. Compound modified rubber asphalt mixture
is also a hot spot for future research.

In addition, because rubber asphalt consumes a large amount of waste tires, it is bound to become the trend of future development. However, the cost of adding modifiers should be fully considered. The compounding of rubber powder and existing industrial waste slag has a certain research value, not only waste It is also possible to produce green building materials that are more suitable for actual projects.

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