

A Review on the Anti-Aging Properties of Modified Asphalt

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Abstract

Asphalt pavement is widely used in road engineering due to its advantages such as short construction cycles and excellent road performance. However, asphalt is prone to brittleness and hardening during production and service under the combined effects of heat, oxygen, and ultraviolet (UV) radiation, leading to degraded pavement performance. To address this, modifiers are commonly incorporated to enhance the anti-aging properties of asphalt.

Keywords

Asphalt; Road Performance; Anti-Aging Performance; Modifier

1. Introduction

Asphalt pavement is widely used in the field of road engineering due to its short construction period and excellent road performance, accounting for 85% of the road pavement mileage in China [1]. However, asphalt is susceptible to aging caused by heat, oxygen, and ultraviolet (UV) light leading to brittleness and hardening during production, storage, transport, paving, and use [2]. These aging processes lead to a decrease in asphalt's resistance to rutting at high temperatures and cracking at low temperatures, which significantly reduces the serviceability and life of the pavement [3]. Asphalt aging can be divided into short-term aging and long-term aging. Short-term aging occurs in the asphalt mixture production and construction stage, mainly triggered by the role of thermal oxygen volatilisation of light components; long-term aging mainly occurs in the service period of the pavement, mainly triggered by the light-oxygen coupling effect of the light components of the continuous loss of the asphalt binding material gradual oxidation. Thermo-oxidative and photo-oxidative aging are accompanied by a reduction in asphalt performance, the

macro-level embodiment of the needle penetration drop, softening point rise, etc., the micro-level embodiment of the component changes, the increase in the number of oxygen-containing functional groups, etc. [4]. Aging will cause many adverse effects on asphalt, therefore, in-depth study of asphalt anti-aging properties is very necessary. In engineering practice, modifiers are often used in asphalt to improve its performance, the addition of modifiers can improve the mechanical properties of asphalt, rheological properties and aging resistance and other characteristics. Commonly used asphalt modifiers include rubber, resin, various types of polymers and nanomaterials. Among the many types of modifiers, nano-modifiers achieve the improvement of asphalt performance with small dosage. At present, the use of carbon nano-modifiers to improve the performance of asphalt mixtures is gradually being applied in practice as a cutting-edge hot technology [5].

2. Study on the performance of asphalt modified by nanomaterials

Thermo-oxidative and photo-oxidative aging are accompanied by a reduction in asphalt performance, which is reflected in the macro level in the decline of the degree of penetration, rheological performance deterioration. The addition of modifiers can improve the mechanical properties, rheological properties and anti-aging properties of asphalt, etc. Among the many types of modifiers, nano-modifiers achieve the improvement of asphalt performance with small dosage. Scholars at home and abroad have revealed the effect of nanomaterial modifiers on the aging resistance of asphalt through systematic research.

Jahromi et al [9] found that the incorporation of nanoclay improved the aging resistance of asphalt along with altering the rheological properties of asphalt. Zare et al [10] organically modified bentonite clay (BT) and found that organically modified bentonite clay (OBT) significantly improved the rutting resistance, low temperature rheological properties and cracking resistance of asphalt. Expanded vermiculite (EVM) with organically modified expanded vermiculite (OEVM) effectively mitigated the thermo-oxidative aging of asphalt, but did not substantially optimise its photo-oxidative aging [11,12]. Montmorillonite (MMT) and organic montmorillonite (OMMT) mixed with asphalt can form an interpolated structure and an anti-stripping structure, so that the modified asphalt has better viscoelasticity, low-temperature cracking resistance, and resistance to thermo-oxidative aging, whereas OMMT shows better results in improving the softening point and rutting resistance of asphalt [13-16]. Nano ZnO as an additive can improve the softening point, ductility (5°C), rheological properties and fatigue properties of matrix asphalt, and this modifier can significantly enhance the resistance of asphalt to UV aging while imparting viscoelastic recovery properties [17,18]. Zhu [19] investigated the effect of nano ZnO by differential scanning calorimetry (DSC), dynamic shear rheometry (DSR), and Brinell's viscometer on the on the road performance of SK-70

asphalt, and the results showed that nano ZnO can significantly improve the low temperature ductility, high temperature rutting resistance and aging resistance of asphalt. Li et al [20] found that nano ZnO reduced the mass change rate and viscosity aging index of asphalt, and the nano ZnO surface modification was more effective in enhancing the anti-aging performance of asphalt. Liu et al [21] found that nano TiO₂ can significantly inhibit the growth of viscosity aging index and carbonyl generation of asphalt, and improve the anti-UV aging performance of asphalt, while nano TiO₂ has better effect after surface modification. The addition of nano-SiO₂ significantly improved the rheological properties of matrix asphalt and enhanced its long-term service durability [22,23]. Zhang et al [24] experimentally confirmed that three inorganic nano-modifiers, nano-SiO₂, nano-TiO₂, and nano-ZnO, significantly enhanced the resistance of asphalt to UV aging, and the surface modification of the three modifiers revealed that the addition of inorganic nanoparticles reduced the softening point increment and viscous aging index of asphalt. The surface modification of inorganic nanoparticles further improved the UV aging resistance of the modified bitumen, among which the nano-ZnO-modified bitumen had the best UV aging resistance. Gong et al [25] found that carbon nanotubes (CNTs) could significantly improve the rheological properties of asphalt. Wang et al [26] conducted aging tests on asphalt by rotating film oven test (RTFOT) and UV and physical property tests, Fourier transform infrared spectroscopy (FTIR) analysis and DSR were used to study the modification effect of GO and CNTs on asphalt, and it was found that both GO and CNTs could inhibit the degradation of asphalt's physical properties as well as the attenuation of the complex modulus and the phase angle, which indicated that both GO and CNTs improved the aging resistance of asphalt. The study of Wang [27] concluded that GO could increase the asphalt's surface free energy and improve the self-healing ability of asphalt after aging. Wu et al [28] investigated the effect of GO on the anti-aging properties of asphalt, and found that GO can improve the resistance of asphalt to photo-oxidative and thermo-oxidative aging. Zeng et al [29] showed that GO improves the high and low temperature properties of asphalt, and effectively inhibits asphalt aging. Liu et al [30] revealed the effect of GO on the thermal stability of SBS. Liu et al [30] revealed the effect of GO on the thermal stability of SBS modified asphalt through aging test, and found that GO could induce the formation of a denser three-dimensional crosslinked network structure and reduce the thermal sensitivity of asphalt.

Jiang Haitao et al [31] investigated the resistance of OMMT-modified asphalt to thermo-oxidative aging by thin film oven aging test (TFOT) and pressure vessel aging test (PAV), and found that the incorporation of OMMT effectively mitigated the thermo-oxidative aging of asphalt. With the increase of OMMT dosage, the effect of OMMT modification was gradually enhanced, but the modification effect tended to level off after the dosage exceeded 3%. Fu Yu et al [32] showed that OMMT can improve the UV aging resistance of asphalt. Yang Qun et al [33] studied the effect of

aging on the performance of nano-TiO₂ modified asphalt, the results show that when the dosage of nano-TiO₂ is 4%, the effect on the physical properties of asphalt is small, but can effectively improve the aging resistance of asphalt. Fan Liang et al [34] used nano TiO₂ on two grades of petroleum asphalt modification research, found that the modification effect of nano TiO₂ is much better than ordinary mineral powder filler, in significantly improve the asphalt resistance to thermal and oxygen aging performance at the same time to improve the asphalt resistance to light and oxygen aging performance. Hou Linjie et al [35] found that graphene has substantially improved the structural strength, rheological properties, and anti-aging properties of asphalt. Li Ke [36] found that the addition of graphene improved the thermal and oxygen aging resistance and high temperature performance of asphalt, but had a negative effect on the low temperature performance of asphalt. Liu Kefei et al [37] analysed the effect of GO on the anti-aging properties of matrix asphalt through indoor tests, and found that the addition of GO can slow down the effect of thermo-oxidative aging on the needle penetration, softening point and ductility of asphalt, and the aging asphalt resistance to water damage is significantly improved, while its low-temperature resistance to cracking performance is also effectively enhanced.

3. Study on the aging mechanism of asphalt binding material

Asphalt is an extremely complex mixture, and the essence of asphalt aging lies in the evolution of molecular structure and component migration. Therefore, researchers investigate the aging mechanism by analysing the changes in components and properties during the aging process.

Qu et al [38] used Corbett's separation method to separate asphalt to study the composition of asphalt under different aging degrees, and found that with aging, the aromatic content decreased, asphaltene content increased, and saturated and colloidal content were more stable. Li et al [39] determined the molecular weights of the components and the changes of the characteristic functional groups of the asphalt in the aging process by using GPC and FTIR, and found that the light component in asphalt gradually shifted to the recombinant component with the increase in age, and the carbonyl and sulfoxide groups gradually increased. Xing et al [40] used atomic force microscopy (AFM) to determine the molecular weights of the components in asphalt. and FTIR respectively, and found that the light component of asphalt gradually shifted to the heavy component with the increase of age, and the carbonyl and sulfite groups gradually increased. Xing et al [40] analysed the chemical composition and aging behaviour of matrix asphalt by AFM and FTIR, and found that the aging increased the content of carbonyl and sulfite groups. Zou et al [41] carried out PAV, TFOT and UV aging of five asphalts with different H/C ratios, and found that low H/C ratio asphalts presented lower carbonyl index aging susceptibility, and PAV aging mainly promoted the generation of aromatic ring and sulfoxide

groups, while UV aging significantly accelerated the cleavage of aliphatic structures. Saha et al [42] investigated the effect of aging on crumb rubber, polymer, and Sasobit-modified asphalts by employing RTFOT and PAV, and found that aging resulted in a significant increase in the content of ketone and sulfoxide and led to a decrease in the light component content and an increase in the heavy component content of the asphalt.

Li Jing et al [43] studied the effect of aging of RTFOT and PAV on asphalt based on FTIR, GPC, and elemental analysis tests, and found that aging led to an increase in the oxygen-containing functional groups of asphalt, and more macromolecular substances, the molecular weight of the asphalt increased, and aging caused the elemental content of asphalt changes in the following ways: the content of sulfur (S) and oxygen (O) increased, and the content of carbon (C), hydrogen (H) and nitrogen (N) decrease. Yang Zhen et al [44] investigated the characteristics of elemental distribution and relative molecular weight changes in asphalt during aging by elemental analysis and GPC, and found that aging leads to an increase in the O content of asphalt and a decrease in the C, H, and N contents, and that aging drives the molecular weight distribution to migrate towards macromolecules, resulting in an increase in the molecular weight of asphalt. This is consistent with the findings of Li Jing et al. Chang Rong et al [45] pointed out in the study of asphalt aging behaviour and aging mechanism, asphalt aging process, the light component is heated and oxygen (O₂) reaction to generate new components and new functional groups; asphalt intermolecular forces are enhanced, resulting in the deterioration of asphalt rheological properties, viscosity decreases, and brittleness increases. Cui Yannan et al [46] studied the aging process of SBS-modified asphalt based on FTIR, and found that the short-chain alkanes underwent addition polymerisation and reconfiguration into long-chain structures. As aging progresses, the carbonyl and sulfoxide content gradually increases, the asphaltene content increases, and the aromatic and gum content decreases.

4. Molecular dynamics modelling in the field of asphalt materials

MD simulation can break through the experimental limitations, from the microscopic scale to study the laws of molecular motion, and the development of computers and computing power to promote the use of this technology in a variety of disciplines. MD simulation can accurately track the asphalt molecular motion and energy changes, but also simulate the outside world is difficult to reach the temperature, pressure, and other conditions, in order to improve the accuracy of the study at the same time to reduce the time and cost.

4.1. Current status of research on bitumen modelling based on molecular dynamics simulation

Molecular dynamics can study the micro-mechanism of asphalt through all-atom modelling, which can reveal the changing law from the molecular level. Currently,

the average molecular model method [49] and the assembly method are mainly used to construct molecular models of asphalt. The average molecular model method for asphalt is based on nuclear magnetic resonance spectroscopy (NMR) and FTIR tests to construct statistically representative average molecular models. Jennings et al [49] used NMR to analyse the average structure of eight asphalt molecules for different configurations, which are AAA-1, AAB-1, AAC-1, AAD-1, AAF-1, AAG-1, AAK-1 and AAM-1. Yi-Jing Lin [50] constructed an average molecular model for asphalt based on elemental analysis (C/H/N/S) with the improved Brown-Ladner method, combining GPC and FTIR tests to obtain asphalt structural parameters. Although the average molecular model method simplifies the modelling process, it does not take into account the interactions between molecules and the effects of different chemical environments, while the assembly method is to select the important components of asphalt molecules according to the actual mixing ratio of the molecular model, which can not only analyse the interactions between the components of the asphalt, but also analyse the relationship between the properties of the asphalt and the components. The assembly method is divided into three-component method: oil, resin, asphaltene, and four-component method: saturated fraction, aromatic fraction, gum, asphaltene, according to the accuracy of the components. A more accurate asphalt model can be established by the four-component method, but the corresponding calculation is also larger.

Zhang et al [51] pioneered a three-component model for asphalt in 2007, containing oil (n-C22 alkanes), resin (1,7-dimethylnaphthalene) and asphaltene (C₆₄H₅₂S₂ and C₇₂H₉₈S). However, the model ignored the presence of polar aromatic components, resulting in the aromaticity and polarity distributions of the components not matching the actual asphaltenes. In order to compensate for the shortcomings of the three-component model, this research team [52] proposed a modified four-component model, i.e., saturated fraction, aromatic fraction, colloid and asphaltene, for AAA-1 bitumen. Based on the three-component model framework, the polar aromatic fraction was constructed by integrating three heterocyclic compounds, namely, benzoquinoline, ethylbenzothiophene and pentylthiophene. In order to more accurately model bitumen, a new 12-molecule structure [53] was further proposed with different molecules representing the four components of bitumen. The asphalt model consisting of 12 molecules is the most widely used model, which simulates the structure and properties of asphalt more accurately, and the density of the asphalt model matches the experimental values.

4.2. Current status of research on asphalt ageing behaviour based on molecular dynamics simulations

Aging significantly reduces the serviceability of asphalt, and MD simulations can help to study the aging behaviour of asphalt by investigating the effects of aging on asphalt at the molecular scale.

Fallah et al [54] studied the effect of aging on the mechanical properties of asphalt

by adding oxygen atoms to the matrix asphalt model to construct asphalt models with different degrees of aging for MD simulation. Pan et al [55] investigated the effect of aging on the internal properties of asphalt based on MD simulation, and found that an increase in the number of oxygen-containing functional groups enhanced the bonding and merging of asphalt molecules and resulted in an increase in bulk modulus, which triggered asphalt's deformation under compressive and tensile stresses due to higher hardness. Qu et al [38] investigated the effect of aging on the mechanical properties and adhesion properties of asphalt based on MD simulation, and found that the parameters obtained by MD simulation could better reflect the macroscopic properties of the asphalt. Luo et al [56] investigated the interactions between the interface of asphalt and the mineral material, and found that the intermolecular interactions between asphalt and mineral material would be enhanced in the aging process, which would lead to the increase in the bulk modulus of the asphalt, and thus the increase in the bulk modulus of the asphalt. The enhancement of intermolecular interaction in the aging process will significantly enhance the asphalt; the diffusion performance of asphalt is closely related to the free volume; in the aging process of asphalt, hydroxyl, carboxyl and other reactive functional groups and acidic mineral interfaces undergo hydrogen bonding, and this reorganisation of bonding at the molecular level enhances the interfacial interactions between asphalt and aggregates. Pan et al [55] investigated the coupled effects of moisture and aging on asphalt based on MD and found that the effect of moisture on asphalt energy is more significant than that of aging on asphalt energy, and the aging asphalt is less resistant to water damage, which is the same as the findings of Xu [57]. Xu et al [57] also explored the diffusion law of water in asphalt based on MD simulation. Xu et al [58] established an epoxy asphalt aging model by introducing oxygen atoms into the asphalt, and then investigated the effect of aging on asphalt properties, and found that the asphalt molecules were more tightly bonded after aging, and the internal voids were reduced, so the diffusion of oxygenated asphalt molecules was weakened. Qiu Yanjun et al [59] based on the four-component method to establish a molecular model of asphalt to simulate the effect of time and temperature on the physical aging of asphalt, and found that in the isothermal and isobaric conditions, with the extension of the simulation time, asphalt appeared to increase the density of the physical aging phenomenon, the free volume decreases, as well as the activity of the physical aging phenomena such as a reduction in the activity of the physical aging occurs in the asphalt of the light component related to the asphalt.

Van et al [60] developed the Reaction Force Field (ReaxFF) based on first principles. ReaxFF is based on the concept of bond order and provides a smooth transition from non-bonding to bonding interactions. For mesoscopic reaction systems, the computational accuracy of ReaxFF is close to that of quantum chemical methods (QM) based on density-functional theory (DFT), while its computational resource

cost is comparable to that of other MD methods. Since its emergence, ReaxFF has been parametrized and applied to a wide variety of materials and processes, including hydrocarbon reactions, energetic materials, and transition-metal-catalyzed nanotube formation. ReaxFF is used for the formation of nanotubes through the evaluating the change in the number of chemical bond levels during the kinetic process, the formation and breakage of chemical bonds can be accurately identified as a means of predicting potential chemical reactions. Therefore, some scholars have investigated the ageing process of bitumen by ReaxFF.

Pan et al [61] investigated the ageing of bitumen by ReaxFF with photoelectron spectroscopy (XPS), which initially showed a high tendency to break chains and high reactivity with oxygen, leading to the rapid formation of low molecular weight alkanes and sulfenyls. Subsequently, oxidation and hardening slow down. And different oxidation mechanisms show different mechanisms of action during these two periods. In the initial period, sulphur oxides are the main oxidation products controlling the viscosity increase. Subsequently, ketones become the main products. Therefore, the ratio of ketones to sulphur oxides depends on the sulphur content in the bitumen. Hu et al [62] modelled the ageing of bitumen by ReaxFF and found that the main oxidation products of bitumen are C-O, H-O and S-O bonds. Oxygen concentration had the greatest effect on the C-O bond yield, and temperature had the greatest effect on the H-O bond yield. Yang et al [63] quantified the change rule of ketones, sulfenones and intermediates in the aging process of asphalt by ReaxFF. It was shown that the formation of the sulfoxide preceded the ketone, and the intermediates (hydrogen peroxide and hydrogen radicals) promoted the oxidation reaction. In addition, cyclic phenyl carbons are more likely to form ketones than branched phenyl carbons. The production of both types of radicals is the main cause of asphalt polymerisation. Xie et al [64] simulated the oxidation process of coal tar pitch (CTP) under medium-temperature conditions based on ReaxFF and classified CTP into three fractions, asphalt acetone soluble (PAS), asphalt pyridinium soluble-acetone insoluble (PPS) and asphalt pyridinium insoluble (PPI). The results show that the oxidation process is initiated by C-O and O-O bonds generated after intramolecular hydrogen transfer. Phenoxy radical is an important intermediate product initially formed by hydroxyl cleavage and benzene ring oxidation, which plays a key role in the synthesis of H₂O, CO₂, and glyoxal and promotes the combination of CO₂ and glyoxal. When the temperature is greater than 650 K, the C-H bond is more likely to break. Compared with the PAS component and PPS component, the PPI component is more stable. Yang Fenghua et al [65], in studying the anti-aging properties of lignin-modified asphalt, found that in the case of insufficient supply of O₂, lignin molecules are able to use their phenolic hydroxyl groups to capture free radicals, which in turn significantly slows down the rate of asphalt aging; if the supply of O₂ is sufficient, the lignin molecules increase the chance of contact between asphalt and O₂ by enhancing the diffusion performance of asphalt

molecules, which in turn leads to the enhancement of the rate of aging of the asphalt.

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