

Article

Study on Interaction Behavior Between Iron Tailings and Asphalt Interface Based on Molecular Dynamics Simulation and Microscopic Test

Yaning Cui ^{1,2,*}, Chundi Si ^{3,*}, Changyu Pu ^{1,2}, Ke Zhao ³ and Zhanlin Zhao ¹

¹ Hebei Provincial Communications Planning, Design and Research Institute Co., Ltd., Shijiazhuang 050091, China

² Yanzhao Modern Transportation Laboratory, Hebei Transportation Investment Group Company Limited, Shijiazhuang 050011, China

³ School of Traffic and Transportation, Shijiazhuang Tiedao University, Shijiazhuang 050043, China

* Correspondence: cuiyaning@stdu.edu.cn (Y.C.); sichundi@stdu.edu.cn (C.S.); Tel.: +86-15903312516 (Y.C.)

Abstract

With the shortage of natural aggregates and the massive accumulation of iron tailings (ITs) solid waste restricting the sustainable development of asphalt pavement engineering, replacing natural aggregates with ITs has become a promising low-carbon solution with prominent economic and social benefits. However, the poor interfacial adhesion between ITs and asphalt severely restricts the engineering application of tailings, and the micro-interaction mechanism at their interface still lacks systematic clarification, which is the key research gap addressed in this work. Different from conventional macro road performance tests, this study innovatively combined molecular dynamics (MD) simulation with microscopic characterization, including Fourier transform infrared spectroscopy (FT-IR) and atomic force microscopy (AFM), to comprehensively reveal the interfacial interaction mechanism between ITs and asphalt at the molecular and microscales. The results indicate that asphalt molecules exhibit higher aggregation concentration and diffusivity on Al_2O_3 and Fe_2O_3 surfaces than on SiO_2 surfaces, proving stronger interfacial interaction between asphalt and iron-rich oxide minerals. Moderate temperature optimizes the adhesion performance of asphalt with Al_2O_3 and Fe_2O_3 , while the interfacial bonding of asphalt on CaCO_3 and SiO_2 weakens as temperature rises. The silane coupling agent KH-550 can effectively react with acidic minerals, SiO_2 minerals in ITs, which significantly increases the concentration, diffusion coefficient, and distribution uniformity of asphalt molecules at the interface. FT-IR results verify that the combination of ITs and asphalt mainly relies on physical adsorption without generating new chemical bonds. AFM tests further confirm that alkaline minerals improve the surface roughness of asphalt mastic, and KH-550 greatly enhances the micro-adhesion force of the interface. The novelty of this work lies in clarifying the mechanism of typical mineral components in ITs and revealing the modification enhancement law of silane coupling agent and alkali minerals at the micro level. This study provides a scientific theoretical support for the high-value engineering utilization of ITs in asphalt pavement, and offers a reference for optimizing the interfacial modification design of solid waste aggregate.

Keywords: iron tailings; asphalt; molecular dynamics; microscopic test; interface interaction mechanism



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1. Introduction

Iron tailings (ITs) are solid wastes generated after extracting valuable elements such as iron during the mining and processing of iron ore. As the world's largest steel producer and consumer, China processes approximately 1 billion tons of iron ore annually, resulting in the output of about 700 million tons of ITs [1]. A large amount of accumulated ITs not only occupies valuable land resources (each million tons of ITs occupies about 100 mu of land) but also causes serious environmental pollution, including soil heavy metal contamination, surface water and groundwater pollution, and dust pollution [2]. In addition, the long-term accumulation of ITs also poses potential safety hazards, such as dam breakage, which threatens the life and property safety of surrounding residents [3]. Therefore, realizing the large-scale, high-value comprehensive utilization of ITs and turning waste into treasure has become an urgent task to promote the green and sustainable development of the iron and steel industry and road engineering, and it has also become a research hotspot of concern to academia and the industry [4].

In the past decade, the application of ITs in asphalt mixtures has been widely studied [5], and a large number of related research results have been reported. However, most of the existing studies focus on the macro road performance evaluation and optimal dosage exploration of IT–asphalt mixtures [6,7], while the in-depth research on the interfacial adhesion mechanism between ITs and asphalt is relatively scarce, which has become a key bottleneck restricting the large-scale and high-quality application of ITs in asphalt pavement [8]. The interfacial adhesion performance between ITs and asphalt is the core factor determining the macro road performance (such as high-temperature rutting resistance, low-temperature crack resistance, and water damage resistance) of asphalt mixtures [9,10]. Due to the complex mineral composition of ITs and the acidic surface properties of most ITs, the adhesion between ITs and asphalt is usually poor [11]. Under the long-term repeated action of water erosion and vehicle load, the interfacial bonding effect between ITs and asphalt will gradually weaken, leading to early damage phenomena such as peeling, cracking, and potholes of asphalt pavement, which seriously shortens the service life of the pavement.

As the core of asphalt mixture component coordination, interface interaction directly governs its macroscopic pavement performance and long-term durability [12]. In studies on iron tailings as mineral fillers for asphalt mixtures, interface characteristics have become a key research focus [13]. To date, scholars have investigated the iron tailing–asphalt interface, mostly focusing on the mastic system to elucidate interface-induced performance mechanisms. Li et al. [14] evaluated asphalt mastic with iron tailings replacing limestone, showing that interfacial bonding regulates mastic rheology and provides a microscale basis for macroscopic performance analysis. Cui et al. [15] found that iron tailings–asphalt interfacial bonding efficiency is closely related to mastic fatigue resistance, with bonding tightness dictating damage evolution under cyclic loading. Wei et al. [16] revealed that interfacial interaction strength is critical for mastic high-temperature stability and thus mixture rutting resistance.

Regarding interface interaction mechanisms, Wang et al. [17,18] demonstrated that filler surface characteristics significantly affect asphalt mortar high-temperature stability and proposed that interfacial bonding strength determines mortar and mixture performance, providing an interfacial evaluation basis for iron tailings application. Li et al. [19] noted that filler type and moisture alter asphalt mixture interfacial state, affecting mechanical and pavement performance—highlighting the need for interfacial optimization of iron tailings fillers. In summary, existing studies confirm interface interaction's significance, but gaps remain in the quantitative characterization of iron tailing–asphalt interfacial interaction and optimization methods, laying a foundation for further research.

With the rapid development of molecular dynamics (MD) simulation technology and advanced microscopic testing technology, scholars have gradually shifted their research focus from the macro scale to the micro and molecular scales, providing a new technical means for in-depth exploration of the interfacial interaction mechanism between asphalt and mineral aggregates [20,21]. At present, relevant studies on the molecular scale mainly focus on the evaluation of interfacial mechanical properties [22], adhesion [23,24], and water damage resistance [25] between traditional mineral aggregates and asphalt, but there are few studies on the molecular-scale interaction mechanism between ITs (with complex mineral composition) and asphalt. Meanwhile, with the help of advanced microscopic testing technologies such as Fourier transform infrared spectroscopy (FT-IR) and atomic force microscopy (AFM), the research on the interaction between asphalt components and mineral aggregate interfaces is also increasing [26]. Chu et al. [27] studied the interfacial transition zone of asphalt mixture by nanoindentation and AFM, and revealed the influence of different filler types on the microstructure of the interfacial transition zone, but did not involve ITs. Xu et al. [28] used AFM to characterize the evolution of the microscopic properties of asphalt mastic after multiple cycle temperature changes, but did not focus on the interfacial interaction between ITs and asphalt.

In summary, although the application of ITs in asphalt mixtures has been widely studied, there are still obvious research gaps: (1) most existing studies focus on the macro performance evaluation and optimal dosage exploration, and the in-depth research on the interfacial adhesion mechanism between ITs and asphalt is insufficient; (2) the influence of different mineral components in ITs on the interfacial interaction with asphalt, as well as the temperature-dependent law of interfacial interaction, have not been systematically clarified; (3) the internal mechanism of how to effectively enhance the interfacial adhesion performance between ITs and asphalt (such as the modification mechanism of silane coupling agent at the microscale) remains to be further explored; and (4) there are few studies combining MD simulation and microscopic testing technology to reveal the interfacial interaction mechanism between ITs and asphalt from the molecular and microscale.

To fill the above research gaps, this study focuses on the core problem of poor interfacial adhesion between ITs and asphalt, and systematically explores the interfacial interaction mechanism between ITs and asphalt by combining MD simulation, FT-IR, and AFM microscopic testing technology. The research focus of this paper is to clarify the interaction law between different mineral components (SiO_2 , Al_2O_3 , Fe_2O_3 , and CaCO_3) in ITs and asphalt at the molecular and microscale, reveal the temperature-dependent mechanism of interfacial interaction, and explore the modification enhancement mechanism of silane coupling agent on the IT–asphalt interface. The research results can not only enrich the theoretical system of interfacial adhesion between solid waste aggregates and asphalt but also provide scientific theoretical support for the high-value, large-scale application of ITs in asphalt pavement, which is of great theoretical significance and engineering application value.

2. Materials and Methods

2.1. Materials

2.1.1. Physical and Chemical Properties of Mineral Materials

The ITs taken in this study are from Shijiazhuang City, Hebei Province. After investigation, it is found that the mineral composition of ITs in the Shijiazhuang area is mainly quartz, which is acidic. The particle size distribution and mineral composition of ITs in this area are similar to those of ITs in most areas of Shijiazhuang. It can be the most typical ITs. It is universal to select the ITs as the research object. ITs accumulate in large quantities in the local area, occupy a large amount of farmland, pollute water sources, and have a

significant impact on the living environment of residents. Figure 1 is the field sampling photo of the ITs used in the study. In this study, ITs are used to refer to IT sand, and ITs below 0.075 mm are defined as IT filler.



Figure 1. Accumulated IT dam.

There are obvious differences in particle size distribution and morphological characteristics between ITs and natural minerals. In order to observe the difference between ITs and natural minerals, IT sand with a particle size of 0.3–0.6 mm, natural mechanism sand, IT filler with a particle size of <0.075 mm, and limestone filler were magnified by 200 times under an optical digital microscope, as shown in Figure 2.

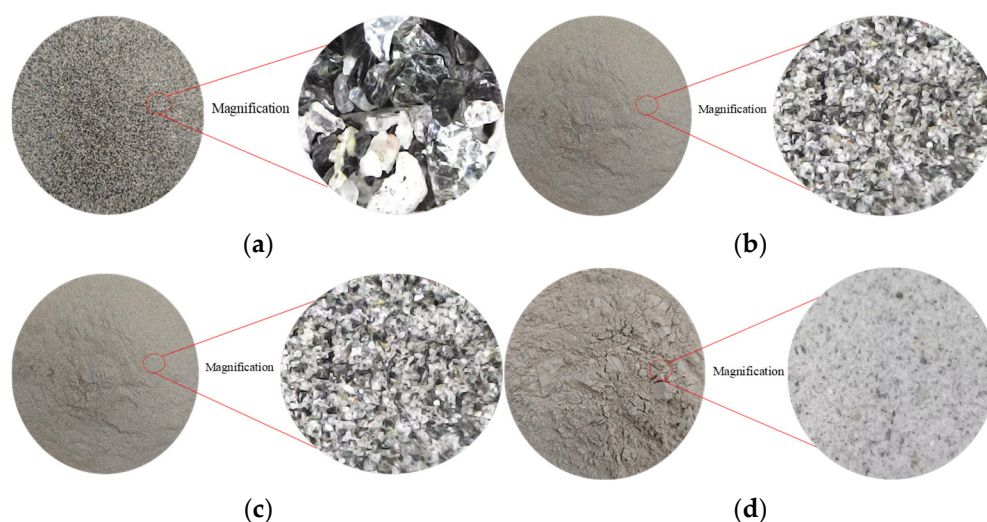


Figure 2. Observation image of ITs and natural mineral under microscope: (a) IT sand (0.3~0.6 mm); (b) manufactured sand (0.3~0.06 mm); (c) IT filler (<0.075 mm); (d) Limestone filler (<0.075 mm).

It can be seen from Figure 2 that at a magnification of 200 times, a large number of bright crystal particles are mixed in the black particles in the microscopic images of IT sand and IT filler. This is because ITs still contain more metal elements, thus showing a metallic luster. In addition, the surface of the IT sand particles is obviously angular, and the shape is broken, while the surface of the machine-made sand particles is rougher, the whole is dark gray, with a small amount of white particles, and the particle shape is relatively regular.

Further, according to the calculation method of the fineness modulus of manufactured sand, the fineness modulus of ITs is 0.95. According to the classification method of sand for construction in the Chinese standard GB/T 14684-2022, it can be judged that the ITs belong

to ultra-fine sand. Then the IT sand is sieved below 0.075 mm, which is called iron tailing filler. The particle size distribution of iron tailings filler and limestone filler was obtained by a laser particle size analyzer, as shown in Figure 3.

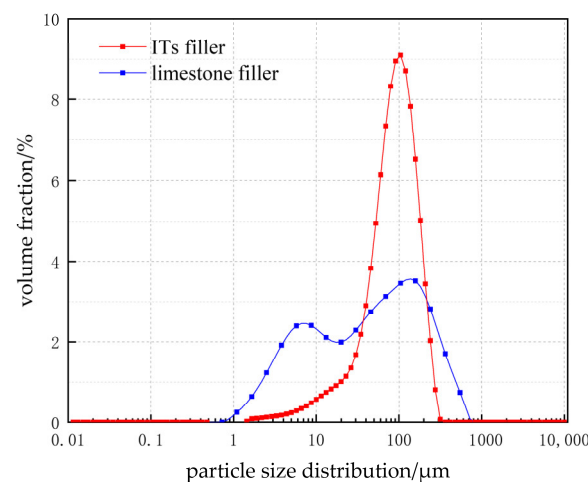


Figure 3. Particle size distribution characteristics of ITs and limestone filler.

The results show that the volume fraction distribution of IT filler shows a single peak characteristic, and its particle size is mainly concentrated near 100 μm . The particle size distribution of limestone ore filler is wide, and the volume fraction peaks appear near 10 μm and 100 μm . The particle size of IT filler is similar to the particle size corresponding to the larger peak value in limestone filler, but a considerable part of the particles in limestone filler are still distributed below 10 μm . The volume proportion of particles with a diameter of 6.6 μm in limestone ore powder is 2.4%, while in IT powder, the proportion of this particle size is only 0.34%. The volume fraction of particles with a diameter of 1 to 10 μm in IT filler is significantly lower than that of limestone filler. Therefore, the overall particle size of limestone filler is finer than that of IT filler, and the specific surface area is also larger.

Table 1 shows the technical index comparison between IT filler and natural limestone filler. It can be seen from the table that the water absorption and mud content of IT sand are high, which meet the requirements of the specification. However, the density and specific surface area of IT filler are lower than those of limestone filler. Therefore, under the premise of the same quality of ore filler and IT filler, the contact area between IT filler and asphalt is small.

Table 1. Technical indexes of iron tail ore and limestone filler.

Test Project	Iron Tailing Filler	Limestone Filler	Method
Specific surface area/(m^2/g)	0.18	0.54	JTG 3432-2024
Hydrophilic coefficient	0.80	0.62	JTG 3432-2024
Apparent density (g/cm^3)	2.36	2.78	JTG 3432-2024
Appearance	No caking	No caking	JTG 3432-2024

The mineral composition of IT filler and limestone filler was determined by X-ray diffraction (XRD) analyzer, the powder samples were dried at $105 \pm 5^\circ\text{C}$, ground to pass a 200-mesh sieve, and tested with Cu K α radiation at 40 kV and 40 mA; the scanning range was set from 5° to 80° (2θ) with a step size of 0.02° , and the scanning speed was controlled at $2^\circ/\text{min}$ to ensure accurate identification of hydration crystalline phases.

The XRD results are shown in Figure 4. The analysis shows that the main mineral composition of the ITs is quartz, accompanied by a small amount of biotite and ankerite.

Quartz is hard and acidic, and has poor adhesion to asphalt. The biotite is weak in weather resistance, low in strength, and easy to oxidize and decompose; iron amphibole is mostly translucent to transparent, and multicolored. In contrast, limestone mineral filler is mainly composed of calcite, which has low strength, alkaline, rough surface and rich texture, which is conducive to adhesion with asphalt. In addition, the limestone filler also contains a small amount of muscovite and quartz, in which muscovite is a sheet structure, colorless to grayish white, with good striping.

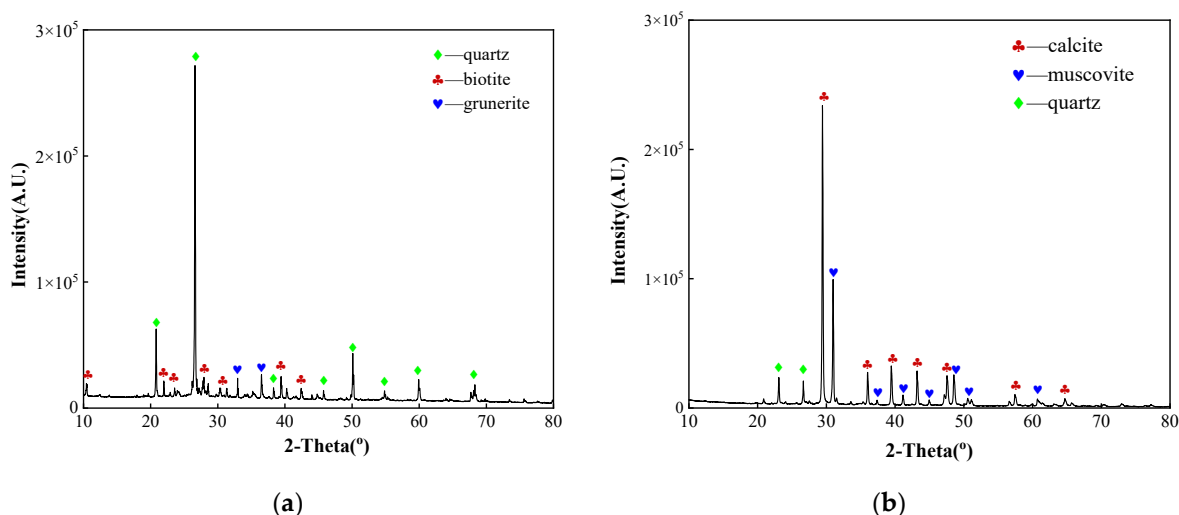


Figure 4. Analysis of mineral composition: (a) IT filler and (b) limestone filler.

The chemical composition of ITs and limestone was analyzed by an X-ray fluorescence spectrometer (XRF). The samples were dried and finely ground to 200-mesh powder beforehand; high-precision fusion bead preparation was adopted to eliminate mineral matrix effects, and the quantitative analysis of main oxide components such as CaO, SiO₂, Al₂O₃ and Fe₂O₃ was completed by XRF.

The test results are shown in Figure 5. It can be seen from Figure 5 that the main components of the ITs are SiO₂, Fe₂O₃, Al₂O₃, and so on. The content of SiO₂ in ITs is the most abundant, reaching 66.7%, which belongs to the typical high silicon ITs. The most common chemical composition of limestone is CaO, accounting for about 65.6% of the total, followed by SiO₂ content of 13.7%, and MgO content of more than 10%. Therefore, limestone is dominated by alkaline oxide CaO, and ITs are dominated by acidic oxide SiO₂, which determines the difference in adhesion with asphalt.

Scanning electron microscopy (SEM) was used to observe the micro-morphology of IT filler and limestone filler, and the results are shown in Figures 6 and 7. Figure 6 shows that the limestone filler is distributed in clusters under SEM, the surface is uneven, the dispersion is poor, and it has rich folds and protrusions. Such surface features can provide a larger bonding area when combined with asphalt. In addition, a large number of micro-pore structures can be seen on the surface of limestone filler, which is helpful for asphalt components to penetrate into the particles through adsorption and capillary action, thus significantly enhancing the bonding performance between asphalt and mineral filler. It can be seen from Figure 7 that the surface of IT filler particles is angular, the particle size distribution is uneven, the shape is relatively broken, and there is no obvious micropore structure.

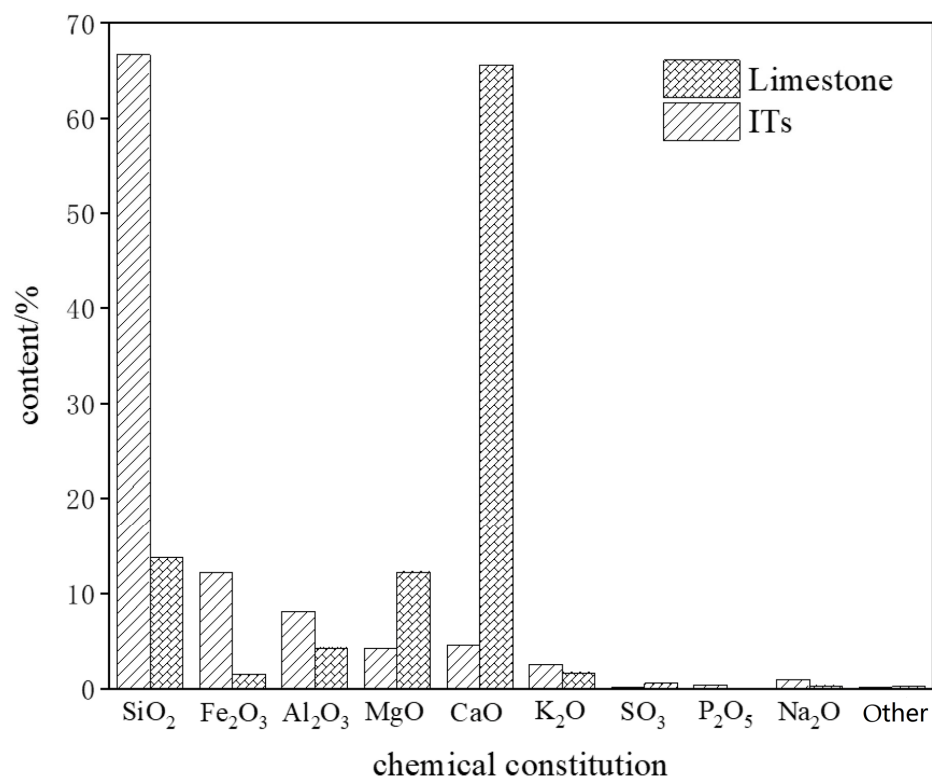


Figure 5. Chemical composition analysis of ITs and limestone.

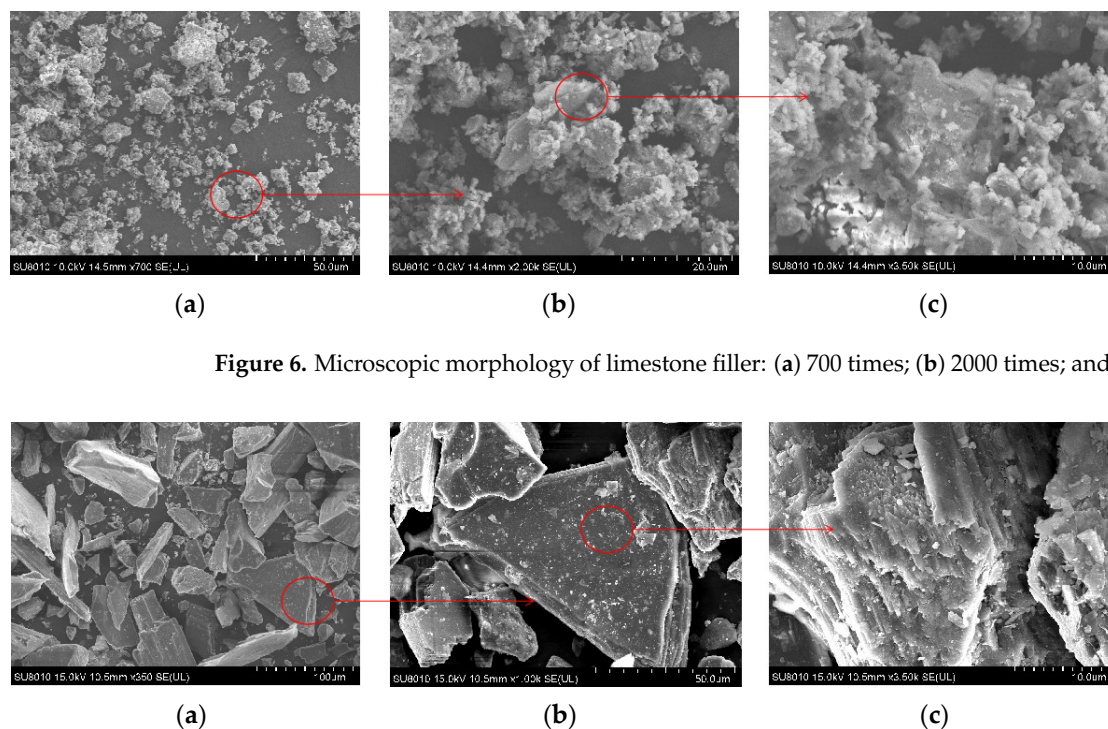


Figure 6. Microscopic morphology of limestone filler: (a) 700 times; (b) 2000 times; and (c) 3500 times.

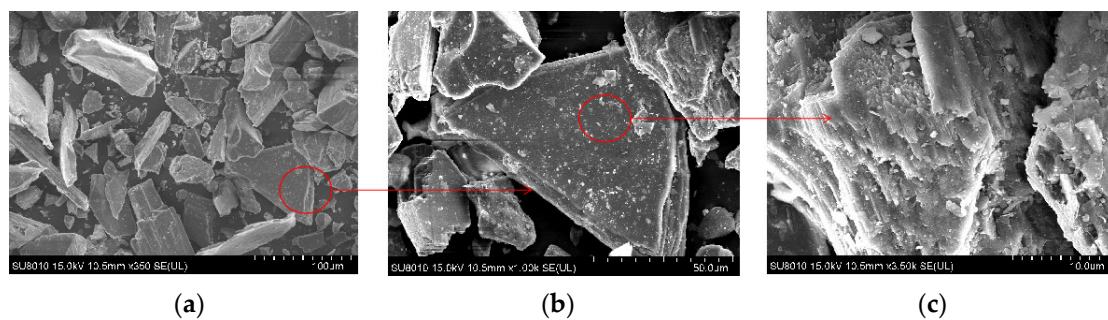


Figure 7. The microstructure of IT filler: (a) 350 times; (b) 1000 times; and (c) 3500 times.

2.1.2. Asphalt Performance

The asphalt selected in this study is 70 # matrix asphalt. According to the test procedures of highway engineering asphalt and asphalt mixture, the basic performance index value of 70 # matrix asphalt is shown in Table 2.

Table 2. 70# matrix asphalt performance index.

Performance Index	Unit	Test Results	Referenced Specifications
Needle penetration (25 °C, 100 g, 5 s)	mm	68.8	T 0604-2011
Softening point	°C	48.0	T 0606-2011
Ductility (5 °C, 5 cm/min)	cm	8.2	T 0605-2011
Ductility (15 °C, 5 cm/min)	cm	146.2	T 0605-2011
Density (25 °C)	g/cm ³	1.028	T 0603-2011

2.2. Research Method

2.2.1. Molecular Dynamics Simulation

Molecular dynamics simulation is a simulation method that tracks the trajectories of atoms or molecules over time and calculates their interactions. It is of great significance to reveal macroscopic phenomena from a molecular perspective. The change in temperature will change the physical state and molecular motion ability of asphalt, thus affecting the interfacial adhesion behavior. According to Newtonian classical mechanics, the microscopic dynamic behavior of each atom is closely related to the total potential energy of the system. Therefore, the force F_i on the i atom is the partial derivative of the system's total potential energy $U(r)$ with respect to that atom, as shown below:

$$F_i = -\nabla U(r) = -\frac{\partial U(r)}{\partial r_i} = -\left\{ \frac{\partial U}{\partial x_i} i + \frac{\partial U}{\partial y_i} j + \frac{\partial U}{\partial z_i} k \right\} \quad (1)$$

In the formula, F_i represents the force of the i th atom (N); $u(r)$ represents the total potential energy of the system (J); r represents atomic displacement (Å); and i , j , and k represent the system space direction vector.

Through molecular dynamics simulation technology, it provides an effective means to study the interaction behavior of the asphalt–mineral interface, and can reveal the interaction mechanism between the IT and asphalt interface at the atomic scale.

The displacement, velocity and acceleration of each atom are calculated by the Verlet algorithm. As a stable classic integration scheme for molecular dynamics, the Verlet algorithm updates particle positions using historical coordinates and instantaneous acceleration, ensuring excellent energy conservation and low numerical error. Based on Newton's classical mechanics theory, the relationship between acceleration and force is obtained, and then the trajectory of each atom is calculated by the total potential energy of the system. The calculation formula is as follows:

$$F_i = m_i a_i = m_i \frac{\partial^2 r_i}{\partial t^2} \quad (2)$$

$$r_i(t + \Delta t) = r_i(t) + \Delta t v_i(t) + \frac{1}{2} \Delta t^2 a_i(t) \quad (3)$$

$$a_i(t + \Delta t) = \frac{F(t + \Delta t)_i}{m_i} \quad (4)$$

$$v_i(t + \Delta t) = v_i(t) + \frac{1}{2} \Delta t [a_i(t) + a_i(t + \Delta t)] \quad (5)$$

In the formula, t represents time (fs); m_i represents the mass of the atom (kg); a_i represents the acceleration of the atom (m/s²); and v_i represents the speed of the atom (m/s).

2.2.2. Surface Micro Parameter Test

In this study, three types of asphalt mastics were prepared, namely iron tailings asphalt mastic (70#-IT), limestone filler modified asphalt mastic (70#-LF-IT), and silane coupling agent KH-550 modified asphalt mastic (70#-IT-KH550).

The detailed preparation procedures are as follows. Firstly, the raw iron tailing sand was sieved through a 0.075 mm square-hole sieve to collect fine iron tailing powder with a particle size below 0.075 mm. The screened powder was dried to a constant weight in an oven to eliminate moisture interference before use.

Prior to mixing, the base asphalt was heated and kept at 166 °C for thermal insulation. The pretreated iron tailing powder was then blended with heated asphalt to prepare the original iron tailings asphalt mastic, with the mass ratio of iron tailing powder to base asphalt strictly controlled at 1:1. The mixture was mechanically stirred continuously at a constant temperature for 30 min to ensure uniform dispersion.

For the limestone filler-modified group, 20% of the iron tailing powder was replaced by limestone filler of equal mass under the same heating temperature and stirring duration. In the KH-550 modification group, the silane coupling agent was added to base asphalt at a mass ratio of 1:100 (KH-550 to asphalt), followed by insulation stirring at the same temperature for 30 min to achieve sufficient modification and homogeneous mixing.

After the preparation of all asphalt mastic samples, the microscopic surface characteristics of different groups were further characterized by atomic force microscopy (AFM). AFM is a non-destructive imaging technology that can obtain information such as surface morphology and surface properties of materials at the microscale. Before preparing the sample, the surface of the glass slide was cleaned with alcohol and placed in a dry and dust-free environment for use. Subsequently, the glass slide and asphalt were put together in a 160 °C oven for heating and heat preservation. After the asphalt was completely melted, an appropriate amount of asphalt was dropped in the center of the glass slide, and the glass slide was placed in the oven for another 5 min. The asphalt was evenly spread on the surface of the glass slide in the molten state to ensure that the test area was smooth. After the paving is completed, the glass slide containing the sample is placed in a special plastic box to avoid the asphalt surface being contaminated. The sample after preparation is shown in Figure 8.



Figure 8. AFM test samples of three types of asphalt mastic.

The surface micro-roughness and adhesion force of the prepared samples were tested in the tapping mode of AFM. The sample surface scanning test area is 20 μm $\times$ 20 μm , the elastic constant K was 45 N/m, and the probe resonance frequency is 325 Hz. Three different test points were taken for each sample, and the surface microscopic parameters

were obtained by scanning the probe with the fluctuation of the sample surface during the scanning process.

2.2.3. Interface Interaction Mechanism Analysis

Fourier transform infrared spectroscopy (FT-IR) was used to analyze the absorption characteristics of molecules on infrared spectroscopy to determine the structure and chemical composition of molecules. The wavelength of infrared spectroscopy is usually between 2.5 μm and 25 μm . The infrared light is absorbed by the molecule and causes molecular vibration and rotation, and the bond angle changes. Different functional groups have characteristic absorption peaks at specific wave numbers to identify the molecular structure. FT-IR uses a Michelson interferometer to generate an interferogram. The interferogram is converted into a spectrogram by the Fourier transform. The horizontal axis is the wave number, and the vertical axis is the absorbance. The absorbance is proportional to the concentration of the substance and can be used for quantitative analysis of the material. Fourier transform infrared spectroscopy has been widely used in materials science, biological science and other fields.

In this study, the peak area ratio was used as a key quantitative index to evaluate the effect of IT filler and limestone filler on the chemical properties of asphalt mastic microstructure. The microscopic chemical changes were analyzed by comparing the peak area ratios of the three characteristic functional groups in the asphalt mastic containing iron-containing tailings filler and limestone filler. Taking the peak area of the whole spectrum in the range of 4000–500 cm^{-1} wave number as the reference, the calculation formula of the peak area ratio of the relevant functional groups is as follows:

$$I_B = A_{1376} / (A_{1376} + A_{1462}) \quad (6)$$

$$I_{Ar} = A_{1600} / \sum A \quad (7)$$

$$I_{S=O} = A_{1032} / \sum A \quad (8)$$

$$\sum A = A_{2920} + A_{2855} + A_{1700} + A_{1600} + A_{1462} + A_{1376} + A_{1032} + A_{862} + A_{723} \quad (9)$$

In the formula, I_B represents the aliphatic functional group index; I_{Ar} represents the aromatic functional group index; $I_{S=O}$ represents the sulfoxide functional group index; A represents the spectral peak area; and $\sum A$ represents the spectral peak area in the range of 4000–500 cm^{-1} .

3. Results and Discussion

3.1. Molecular Simulation of the Interaction Behavior of the IT–Asphalt Interface

3.1.1. Molecular Model Construction

In the Materials Studio software 2018, the asphalt molecular model was constructed by the Amorphous Cell module. In the Calculation interface, the relevant parameters are input according to the number and mass ratio of the molecules corresponding to the 12 components of the asphalt, and the initial density is set to 0.1 g/cm^3 to avoid the entanglement of the molecules in the initial model. The unit cell size of the initial model is 81.49 $\text{\AA} \times 81.49 \text{\AA} \times 81.49 \text{\AA}$.

Subsequently, Geometry Optimization in the Forcite module was used to geometrically optimize the molecular structure of the asphalt until the total energy was minimized. Then, the asphalt molecules were subjected to five cycles of annealing treatment in the temperature range of 300–500 K by the Anneal function. Then, the dynamic calculation of 500 ps is carried out under the NPT ensemble. The simulation temperature is 298 K. The

Nose method is used to control the temperature, the Berendsen method is used to control the pressure, and the time step is set to 1 fs.

The established asphalt molecular model is shown in Figure 9, and the density change curve is shown in Figure 10. After the completion of the calculation, the volume of the asphalt molecular model is significantly reduced, the unit cell size is reduced to $37.87 \text{ \AA} \times 37.87 \text{ \AA} \times 37.87 \text{ \AA}$, and the asphalt density is close to 1.013 g/cm^3 , which is basically consistent with the experimental value.

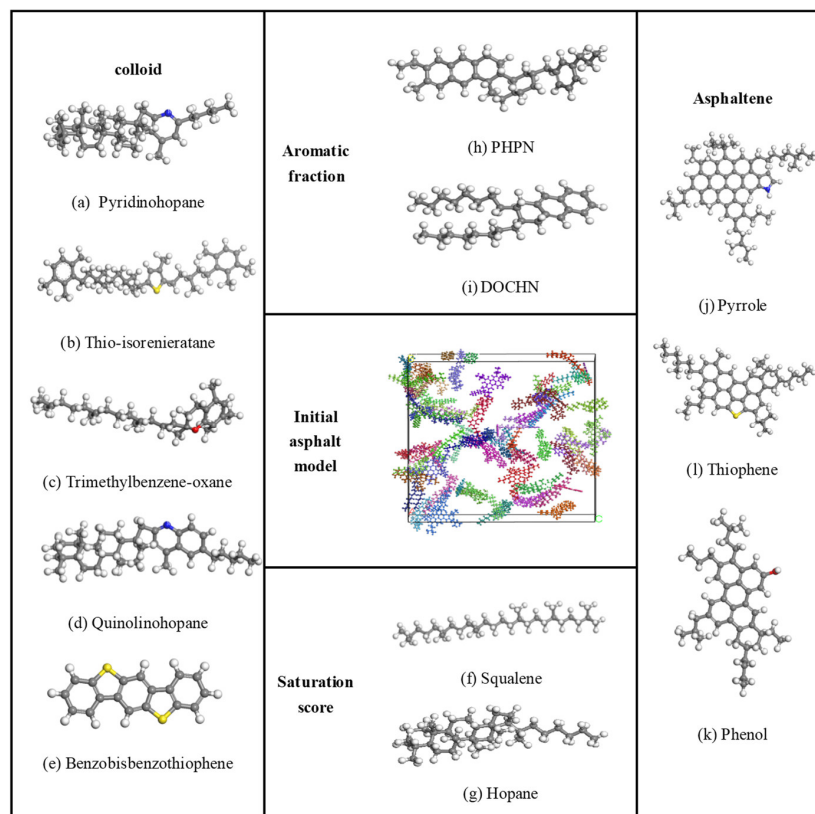


Figure 9. Asphalt molecular model composition.

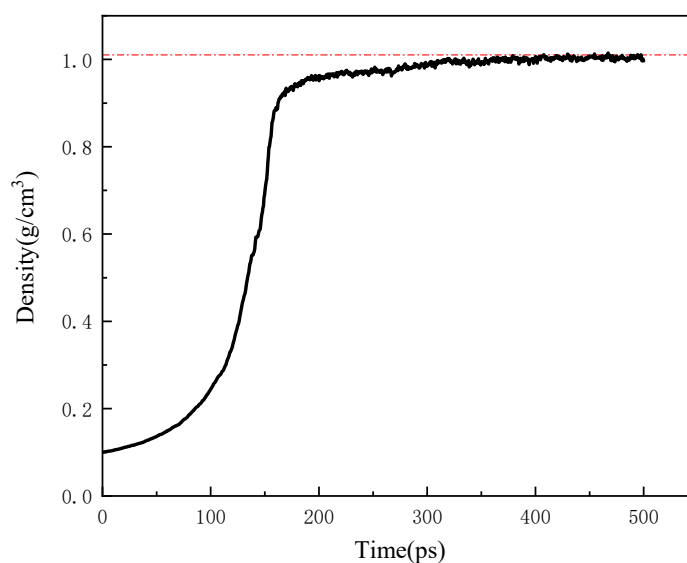


Figure 10. Asphalt molecular density.

To clarify the parameters involved in the analysis, the key indicators are defined as follows: cohesive energy density (CED) refers to the cohesive energy per unit volume of a substance, which is the core basis for calculating the solubility parameter; the solubility parameter, defined as the square root of CED, is used to quantitatively characterize the compatibility between asphalt binder and mineral filler; the radial distribution function (RDF) is a statistical function describing the spatial distribution of atoms and molecules, which is applied to evaluate the interfacial adsorption effect between asphalt molecules and mineral filler surfaces. From a practical engineering perspective, CED and solubility parameter provide a reliable quantitative basis for the selection of mineral fillers (cement, IT powder, and limestone powder) in asphalt mastic, which is crucial for improving the interfacial adhesion performance of asphalt mastic and further enhancing the durability of asphalt pavement. RDF, on the other hand, helps reveal the microscopic adsorption mechanism of asphalt molecules on mineral filler surfaces, which can guide the optimization of asphalt mastic preparation parameters and provide theoretical support for the practical application of IT powder in asphalt pavement engineering.

CED is used to quantitatively evaluate the internal interaction of asphalt molecules. The greater the cohesive energy density, the stronger the intramolecular interaction of asphalt molecules, and the greater the intermolecular force. The cohesive energy density is the arithmetic square root of the solubility parameter. Since the molecular model of asphalt mainly relies on the electrostatic force and van der Waals force to produce interaction, the cohesive energy density and solubility parameter are determined by the van der Waals force and electrostatic force of asphalt. The calculation formula is as follows:

$$CED = \delta^2 \quad (10)$$

$$\delta = \sqrt{\delta_{vdw}^2 + \delta_{ele}^2} \quad (11)$$

In the formula, δ is the solubility parameter $(J/m^3)^{1/2}$; δ_{vdw} and δ_{ele} are the van der Waals force and electrostatic force between pitch molecules, respectively.

The cohesive energy density and solubility parameters of the asphalt molecular model constructed in this paper are shown in Table 3. The simulation results are close to the reference values, which indicates that the test results are relatively accurate and have good reference values.

Table 3. Cohesive energy density and solubility parameters of asphalt.

Characteristic Attribute	70#Asphalt Binder	Reference Value
Cohesive energy density (J/cm^3)	316	234–529 [29]
Solubility parameter $(J/m^3)^{1/2}$	17.7	13.3–22.5 [29]

Through XRF chemical composition analysis, it can be seen that the main chemical component of the ITs is SiO_2 , accounting for 66.7%. In addition, the ITs also contain a certain amount of Fe_2O_3 and Al_2O_3 . SiO_2 is a kind of atomic crystal mainly composed of covalent bonds. It forms a complex three-dimensional network structure by silicon–oxygen tetrahedron. SiO_2 has a variety of crystal structures, such as α -quartz and β -quartz. Among them, α -quartz belongs to the trigonal system, which is the most common natural quartz mineral, trigonal system, and a common hexagonal columnar crystal. In this paper, α -quartz was selected as the main mineral component of ITs. Taking the SiO_2 cell as an example, the (0 0 1) crystal plane was selected to cut out the surface atomic structure, the cell thickness was set to 25, and the supercell structure was established. Then, the geometric optimization was carried out by the energy minimization algorithm, and the vacuum layer was established on the surface. In addition, considering the influence of Al_2O_3 and Fe_2O_3

on the interfacial adhesion of asphalt–IT, the supercell of Al_2O_3 and Fe_2O_3 crystals was established. Fe_2O_3 and Al_2O_3 are not typical ionic crystals, but complex compounds with ionic bonds and covalent bonds. The crystal structure and physical properties of Fe_2O_3 and Al_2O_3 reflect the interaction of ionic bonds and covalent bonds.

At the same time, the main mineral component of limestone is calcite, CaCO_3 . Calcite belongs to the trigonal system. Common rhombohedral, columnar or plate-like crystals have complete rhombohedral cleavage and are easy to crack along the cleavage plane. CaCO_3 is a typical ionic crystal. The common cleavage plane (2 1 4) of calcite is selected to establish the supercell structure of CaCO_3 . The four main components of ITs and limestone are shown in Figure 11. The interface adhesion model between minerals and asphalt molecules was established by the Build layer instruction in MS software, as shown in Figure 12.

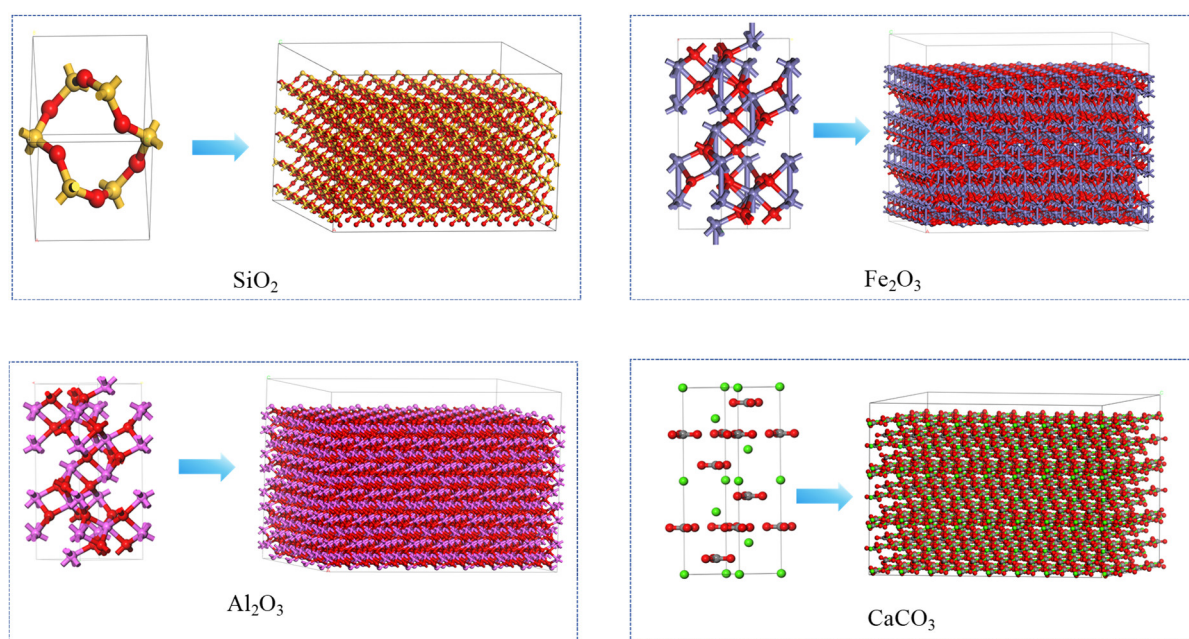


Figure 11. Main components of ITs and limestone.

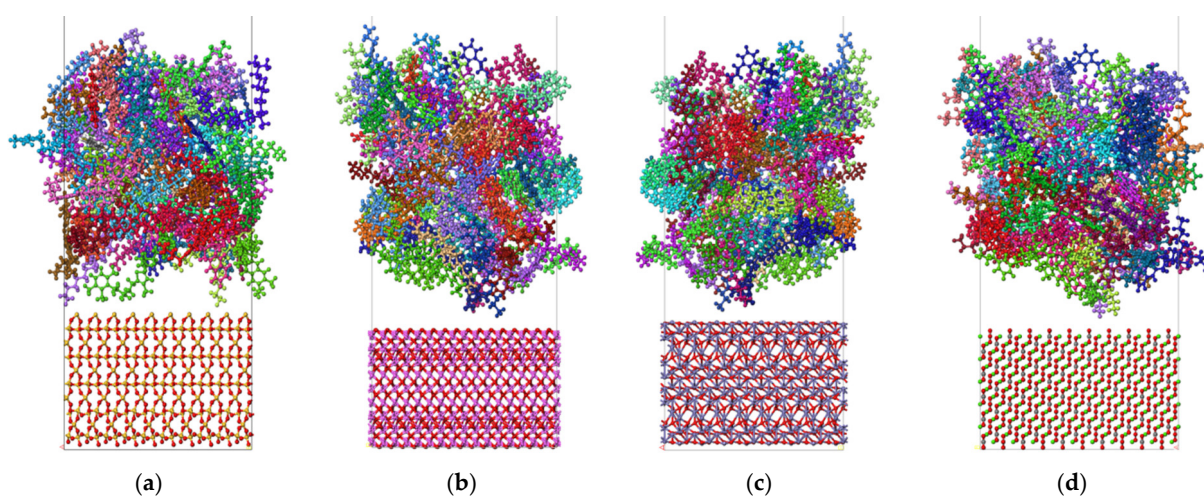


Figure 12. Interface adhesion model of asphalt and main mineral components: (a) asphalt– SiO_2 ; (b) asphalt– Fe_2O_3 ; (c) asphalt– Al_2O_3 ; and (d) asphalt– CaCO_3 .

The interface models of mineral and asphalt molecules in the diagram are asphalt– SiO_2 , asphalt– Fe_2O_3 , asphalt– Al_2O_3 and asphalt– CaCO_3 . The middle and lower layers of

the model are mineral molecules, and the upper layer is asphalt molecules. The model size is $30 \text{ \AA} \times 30 \text{ \AA} \times 120 \text{ \AA}$, the distance between asphalt and mineral aggregate is $2 \text{ \AA}$, and a $50 \text{ \AA}$ vacuum layer is set on the upper layer of asphalt molecules to eliminate the influence of periodic boundary conditions.

3.1.2. Analysis of the Interaction Mechanism of the IT–Asphalt Molecular Interface

In order to study the effect of temperature on the interfacial interaction between asphalt and ITs, four research temperatures of $0 \text{ }^{\circ}\text{C}$ (273 K), $15 \text{ }^{\circ}\text{C}$ (288 K), $25 \text{ }^{\circ}\text{C}$ (298 K), $35 \text{ }^{\circ}\text{C}$ (308 K) and $50 \text{ }^{\circ}\text{C}$ (323 K) were set up in this paper. The evolution of interfacial interaction between asphalt and minerals at different temperatures was quantitatively revealed by radial distribution function, relative concentration and root mean square displacement.

The radial distribution function (RDF) is an essential and widely used parameter in molecular dynamics (MD) simulation to characterize the microscopic spatial distribution of molecules at the interface between materials. In this study, the RDF equation is fully applicable to analyzing the IT–asphalt interface, as it can effectively statistically characterize the distance distribution between asphalt molecules and typical mineral components (SiO_2 , Al_2O_3 , Fe_2O_3 , CaCO_3) of ITs at the equilibrium state, thereby accurately evaluating the interfacial adsorption effect. As a conventional and mature equation in MD simulation, the RDF equation has been widely applied in interfacial interaction studies of asphalt and mineral aggregates, which fully verifies its reliability and feasibility. In this research, the RDF equation is used to quantify the adsorption density of asphalt molecules on the mineral surface through peak intensity analysis, which can effectively reflect the strength of interfacial interaction, proving that the RDF equation is suitable for the research purpose of this study, as shown in Formula (12):

$$g(r) = \frac{1}{4\rho\pi r^2\delta r} \frac{\sum_{t=1}^K \sum_{j=1}^N \Delta N(r \rightarrow r + \delta r)}{N \times K} \quad (12)$$

In the formula, ρ is the density of the system, N is the number of atoms, K is the time step, r is the radial distribution function distance, and ΔN is the number of atoms.

Figure 13 shows significant differences in asphalt molecule aggregation on the surfaces of different ITs main minerals, SiO_2 , Al_2O_3 , Fe_2O_3 , and CaCO_3 , at varying temperatures, which are closely related to the minerals' distinct physico-thermal properties and mineralogical compositions. These properties have been partially reported in limited existing studies. Specifically, the asphalt radial distribution function RDF peak is the highest on CaCO_3 , the lowest on SiO_2 , and similar on Fe_2O_3 and Al_2O_3 . This indicates the strongest asphalt– CaCO_3 adsorption and highest aggregation, as well as the weakest asphalt– SiO_2 adsorption and poorest aggregation, consistent with the known mineralogical characteristics such as alkaline CaCO_3 and acidic SiO_2 in relevant studies.

As shown in Figure 13a, the asphalt– SiO_2 interface RDF peak is the largest at $0 \text{ }^{\circ}\text{C}$ with high aggregation. With increasing temperature, the peak gradually decreases and broadens, accompanied by reduced aggregation and weakened asphalt molecular interaction. This is attributed to SiO_2 's physico-thermal properties. Low temperatures limit asphalt molecular mobility, leading to insufficient surface contact and a low RDF peak. High temperatures cause a disordered arrangement, destroying the interface structure and further reducing the peak. This observation is consistent with limited existing research on asphalt–mineral interface temperature dependence.

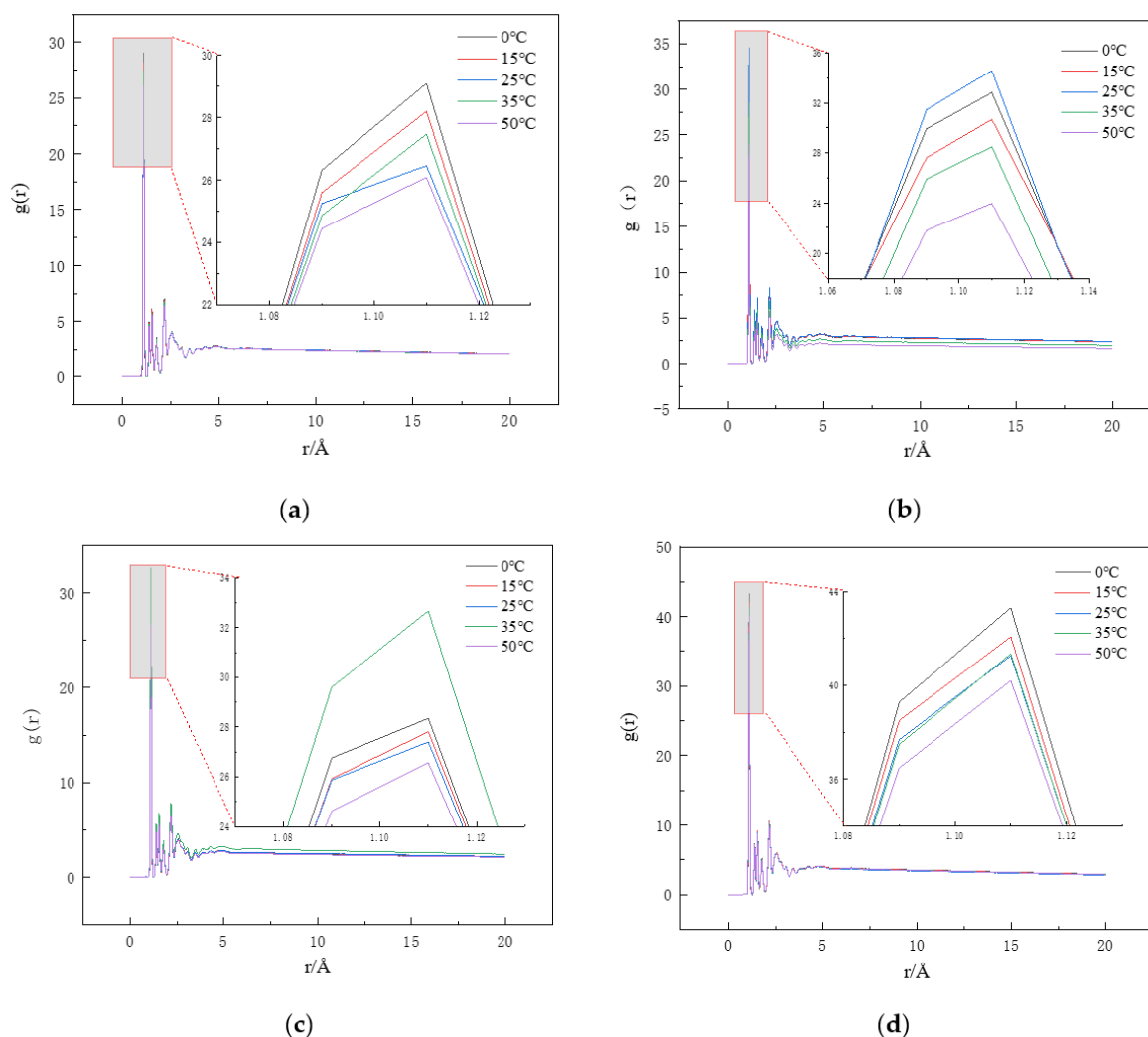


Figure 13. Radial distribution function of asphalt on mineral surface at different temperatures: (a) SiO_2 ; (b) Al_2O_3 ; (c) Fe_2O_3 ; and (d) CaCO_3 .

Figure 13b,c shows that the temperature response of its' main iron-bearing minerals, Al_2O_3 and Fe_2O_3 , differs from that of SiO_2 . Their asphalt RDF peaks appear at 25 °C and 35 °C, respectively, indicating the highest asphalt aggregation under this temperature range due to the synergistic effect of temperature and mineral adsorption. Medium temperatures grant asphalt molecules moderate mobility, enabling full diffusion and orderly arrangement on Al_2O_3 and Fe_2O_3 surfaces by virtue of their unique physico-thermal properties, thus maximizing RDF peaks. This finding supplements the limited existing research on IT mineral–asphalt interfacial interaction under different temperatures.

Figure 13d shows that with increasing temperature, asphalt molecule density near CaCO_3 decreases, the RDF peak weakens, and peak width increases, indicating a gradual dispersion trend. This is closely related to CaCO_3 's thermal stability and surface adsorption capacity, consistent with its basic physico-thermal characteristics in relevant mineralogy studies. Overall, the results reflect the influence of IT minerals' physico-thermal properties and mineralogical composition on asphalt–mineral interfacial interaction, providing a new reference for the limited existing research on ITs in asphalt mastic.

Figure 14 shows significant differences in the concentration distribution of asphalt on the surfaces of four main IT minerals at varying temperatures. The concentration of asphalt molecules on the surface of CaCO_3 is generally higher than that of the other three minerals, indicating that CaCO_3 has the strongest adsorption effect on asphalt molecules.

The concentration of asphalt molecules on the surface of SiO_2 is the lowest, which means its interaction with asphalt is the weakest. With the increase in temperature, the concentration of asphalt molecules on the surface of each mineral shows a downward trend, and this trend is particularly obvious on the surfaces of SiO_2 and CaCO_3 . At 25 °C, the concentration peak of asphalt molecules appears at a closer distance on the surface of SiO_2 , indicating that the adsorption of SiO_2 on asphalt is relatively enhanced at this temperature. As the longitudinal distance increases, the concentration gradually decreases. The average concentration of asphalt molecules on the surface of SiO_2 is the highest at 0 °C, which reflects the strong aggregation of asphalt at low temperatures. At 50 °C, the relative concentration is lower, and the peak value is wider, indicating that asphalt molecules are more dispersed on the mineral surface at high temperatures. On the surfaces of Al_2O_3 and Fe_2O_3 , the average concentration of asphalt molecules reaches the highest in the temperature range of 25 to 35 °C. This indicates that the surface interaction between asphalt molecules and these two iron-bearing minerals is best under medium temperature conditions. Such temperature conditions are conducive to the formation of stronger interfacial bonding and a stable interface layer, thereby enhancing the interfacial interaction between asphalt and IT minerals. These findings are consistent with the basic physico-thermal characteristics of main IT minerals reported in relevant mineralogy studies and supplement the limited existing research on ITs application in asphalt mastic.

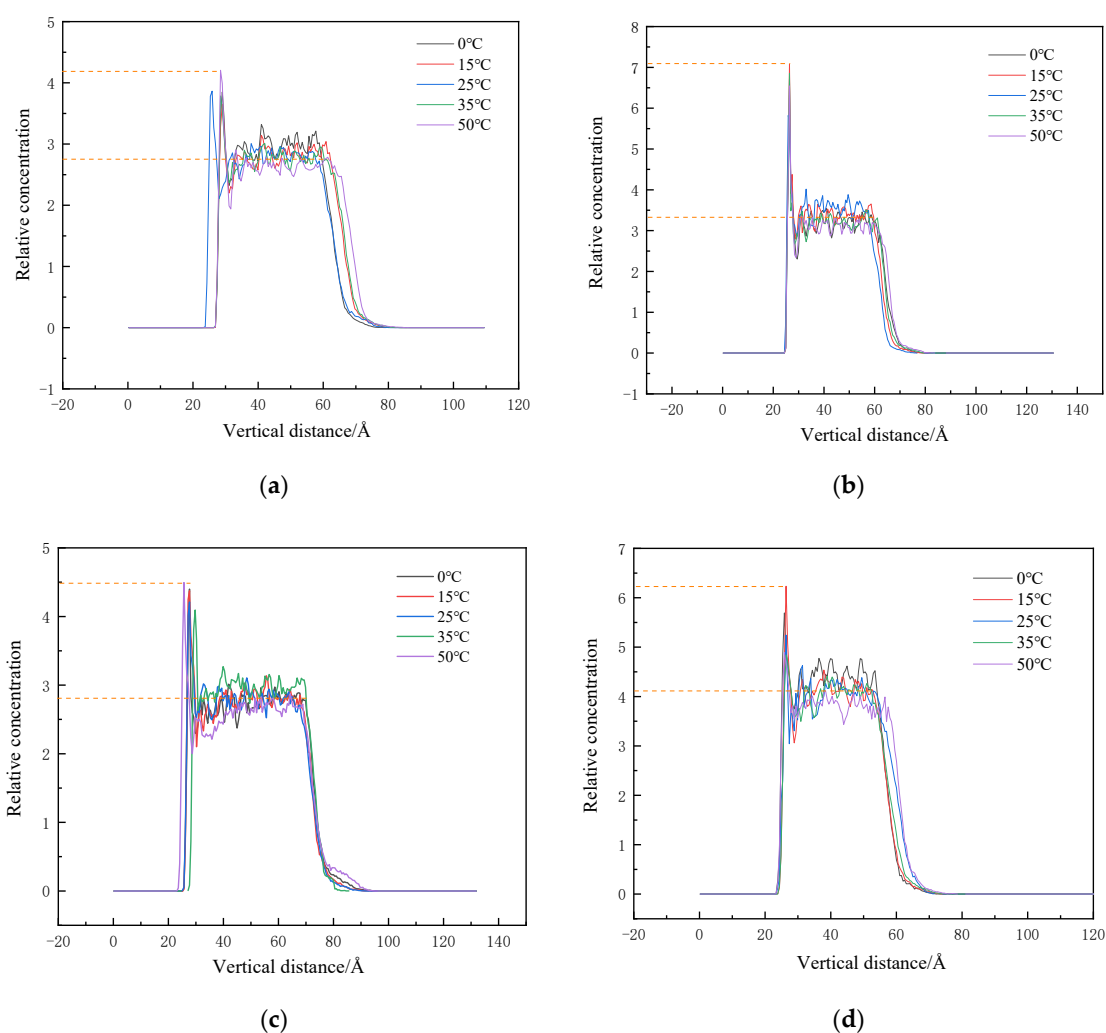


Figure 14. Relative concentration distribution of asphalt on different mineral surfaces at different temperatures: (a) SiO_2 ; (b) Al_2O_3 ; (c) Fe_2O_3 ; and (d) CaCO_3 .

The diffusion coefficient equation based on the Einstein relation is a standard and widely accepted method in MD simulation to evaluate molecular mobility at the interface [30]. This equation is fully applicable to this study, as it can effectively quantify the mobility of asphalt molecules on the surface of IT minerals, and further reflects the strength of interfacial interaction—a lower diffusion coefficient indicates stronger interfacial adsorption and better bonding effect [31]. The Einstein relation-based diffusion coefficient equation has been extensively applied in the study of asphalt molecular diffusion behavior at mineral interfaces, which fully verifies its feasibility and reliability. In this research, this equation is used to analyze the diffusion behavior of asphalt molecules on different IT mineral surfaces, which can effectively reveal the intrinsic mechanism of interfacial interaction, proving that the diffusion coefficient equation is fully capable of meeting the research needs of this study. The mean square displacement calculation formula is as follows:

$$MSD(t) = \sum_{i=1}^n \langle |r_i(t) - r_i(0)|^2 \rangle \quad (13)$$

In the formula, n is the number of atoms; $r_i(t)$ is the coordinate of the atom at time t ; $r_i(0)$ is the coordinate of the atom at the initial time; and $|r_i(t) - r_i(0)|$ is the mean square displacement, where the angle bracket indicates that the MSD of all systems is averaged.

The diffusion coefficient is 1/6 times the slope of the root mean square displacement over a period of time, as shown in the following equation:

$$D = \frac{1}{6N} \lim_{t \rightarrow \infty} \frac{d}{dt} \sum_{i=1}^N \langle |r_i(t) - r_i(0)|^2 \rangle \quad (14)$$

The diffusion law of asphalt on the surface of four mineral components at different temperatures was analyzed by root mean square displacement, as shown in Figure 14.

Figure 15 shows the diffusion ability and movement characteristics of asphalt on the mineral surface at different temperatures. The middle part of the curve with a relatively stable diffusion rate is linearly fitted, and the slope of the fitting line indirectly reflects the diffusion performance of asphalt during this period. As shown in Figure 15a, at high temperatures, asphalt molecules exhibit high diffusion ability. The molecular movement is violent, fluidity is significantly enhanced, molecular diffusion speed is accelerated, the mean square displacement (MSD) increases rapidly with time, and the slope of the fitting line becomes larger. At this time, the kinetic energy of asphalt molecules is higher, and the interaction with minerals is stronger. At low temperatures, asphalt molecules are in a “frozen” state in local areas, with a slow diffusion rate, little change in MSD with time, a low slope of the fitting line, limited movement ability of asphalt molecules, slow molecular diffusion and low diffusion speed.

It can be seen from Figure 15b,c that the slope of asphalt molecules on the surfaces of Fe_2O_3 and Al_2O_3 is the lowest in the temperature range of 15~25 °C. This indicates that asphalt molecules are strongly adsorbed by these two iron-bearing minerals at this temperature range, and the movement of asphalt molecules at the interface is properly constrained. The diffusion behavior tends to be stable, resulting in the smallest slope of the fitting line. At this time, asphalt molecules will not be completely restricted, as at low temperatures, nor will they move violently as at high temperatures, and the diffusion state is the most stable.

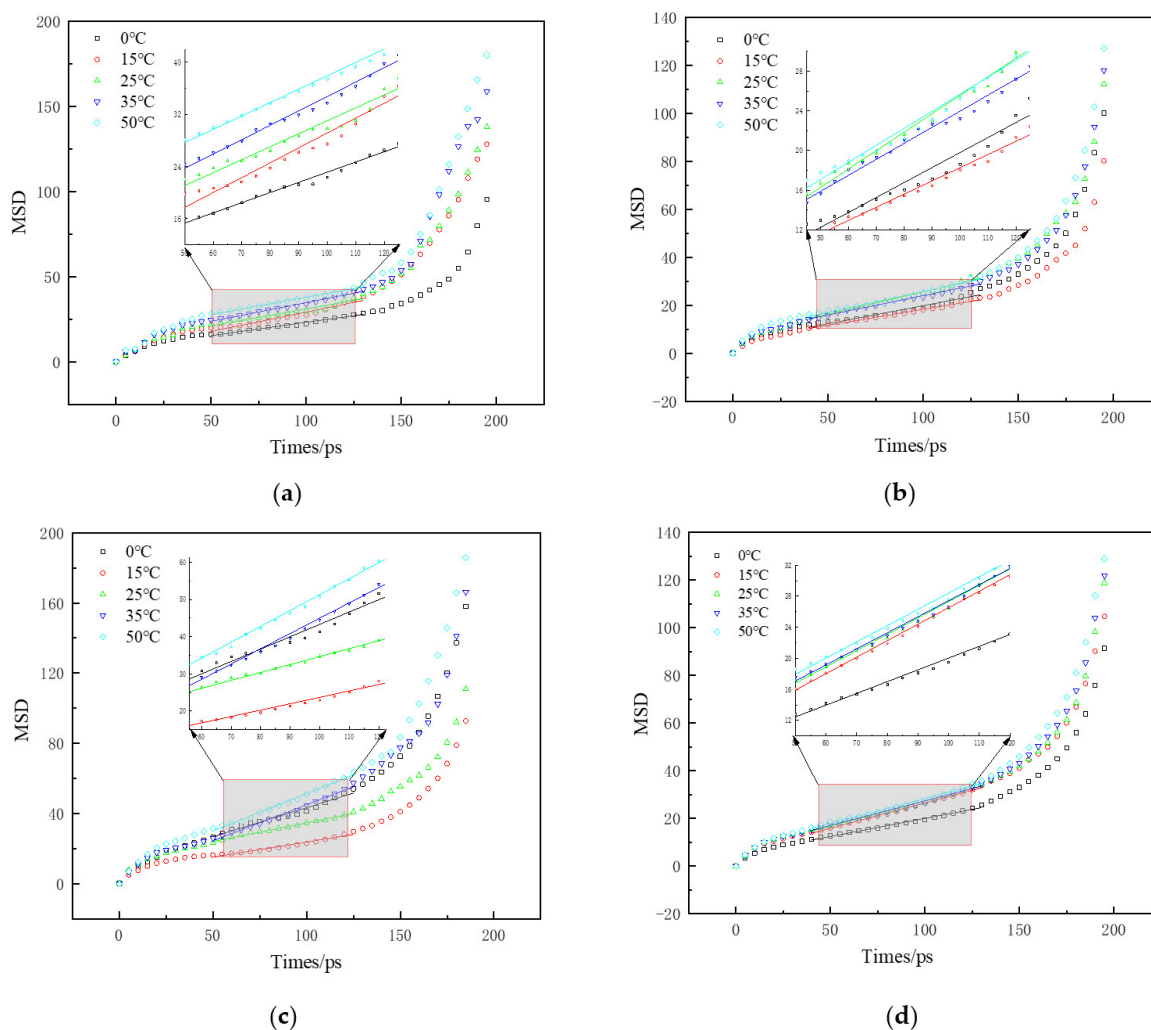


Figure 15. Diffusion law of asphalt on different mineral surfaces at different temperatures: (a) SiO₂; (b) Al₂O₃; (c) Fe₂O₃; and (d) CaCO₃.

From Figure 15d, it can be seen that the MSD peak values of asphalt on the surfaces of CaCO₃ and Al₂O₃ are small, and the slope of the middle section of the curve is low. This indicates that CaCO₃ and Al₂O₃ have strong adsorption capacity for asphalt molecules; the interaction between asphalt molecules and these two minerals is strong, and the diffusion movement of asphalt molecules is effectively constrained, so the diffusion speed is slow and the MSD changes little with time. These findings are consistent with the basic physico-thermal characteristics of the corresponding minerals reported in relevant mineralogy studies, and further supplement the limited research on the interaction between IT minerals and asphalt.

3.1.3. Study on the Interaction Enhancement Mechanism of the IT–Asphalt Interface Modification Mechanism of Silane Coupling Agent

In this study, silane coupling agent KH-550 was selected as the reinforcing material of the IT–asphalt interface. Silane coupling agent can connect asphalt and aggregate through its organic end and the inorganic end, respectively, so as to improve the interfacial adhesion between asphalt and ITs. At present, the common application method is to directly add the silane coupling agent to asphalt for modification. However, due to the high water content of the silane coupling agent, it is easy to produce a foaming phenomenon in water, and its hydrolysis process will release a large amount of ethanol, which not only has safety risks but also may affect the basic performance of asphalt. Therefore, this section uses silane

coupling agent hydrolysis to prepare a filler for modified asphalt to improve the problem of insufficient adhesion between ITs and asphalt.

In this section, the effect of silane coupling agent KH-550 hydrolysate on the interaction behavior of the IT–asphalt interface was studied based on the molecular dynamics method. Figure 16a is the chemical formula of the hydrolysis product of silane coupling agent KH-550, which is 3-aminopropyltriethoxysilanol $\text{H}_2\text{NC}_3\text{H}_6\text{Si}(\text{OH})_3$, and its modification mechanism is shown in Figure 16b.

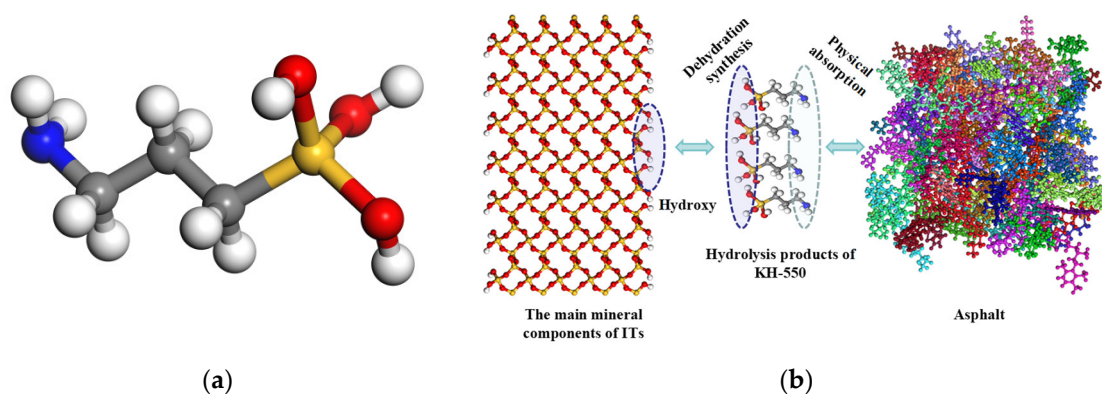


Figure 16. Schematic diagram of strengthening mechanism of KH-550 on IT–asphalt interface: (a) hydrolysis product of silane coupling agent KH-550 and (b) modification method.

From Figure 17a, it can be seen that the peak value of the radial distribution function of the asphalt on the surface of the ITs becomes larger and the aggregation degree of the asphalt becomes higher due to the action of the hydrolysis molecules of the silane coupling agent on the interface between the asphalt and the ITs molecules, indicating that the attraction of the modified ITs to the asphalt is obviously stronger. It can be seen from Figure 17b that the slope of the root mean square displacement curve of the modified asphalt molecule becomes significantly larger, indicating that the diffusion of the asphalt molecule on the surface of the ITs becomes stronger, and the asphalt has a tendency to accelerate to the surface of the ITs. From Figure 17c, it can be seen that the concentration of modified asphalt on the surface of ITs has also changed to a certain extent. The longitudinal distance of the peak value of asphalt on the surface of ITs is advanced, indicating that the adsorption of ITs on asphalt becomes larger, so that the distance of asphalt distribution on the surface of minerals is further reduced, and the bonding is closer. Based on the radial distribution function, root mean square displacement and relative concentration, it was confirmed that the silane coupling agent KH-550 effectively enhanced the interfacial interaction between ITs and asphalt.

Modification Mechanism of Limestone

Because alkaline minerals such as limestone mineral filler mainly contain calcium carbonate and the surface is rich in calcium ions, the acidic components in the asphalt react with the calcium ions on the surface of the mineral to form a stable calcium carboxylate. In addition, the surface energy of alkaline minerals is high, and van der Waals force and electrostatic interaction enhance the infiltration and adhesion of mineral surface and asphalt. Therefore, alkaline minerals can improve the interfacial interaction between acid ITs and asphalt through the synergistic effect of chemical bonding and high surface energy. In this section, the surface of acidic ITs was alkaline-treated, and a layer of calcite was constructed on the surface of quartz, the main mineral component of ITs, to study the effect of alkaline minerals on the interfacial interaction behavior of IT–asphalt. The $\text{SiO}_2\text{--CaCO}_3\text{--asphalt}$ model is shown in Figure 18.

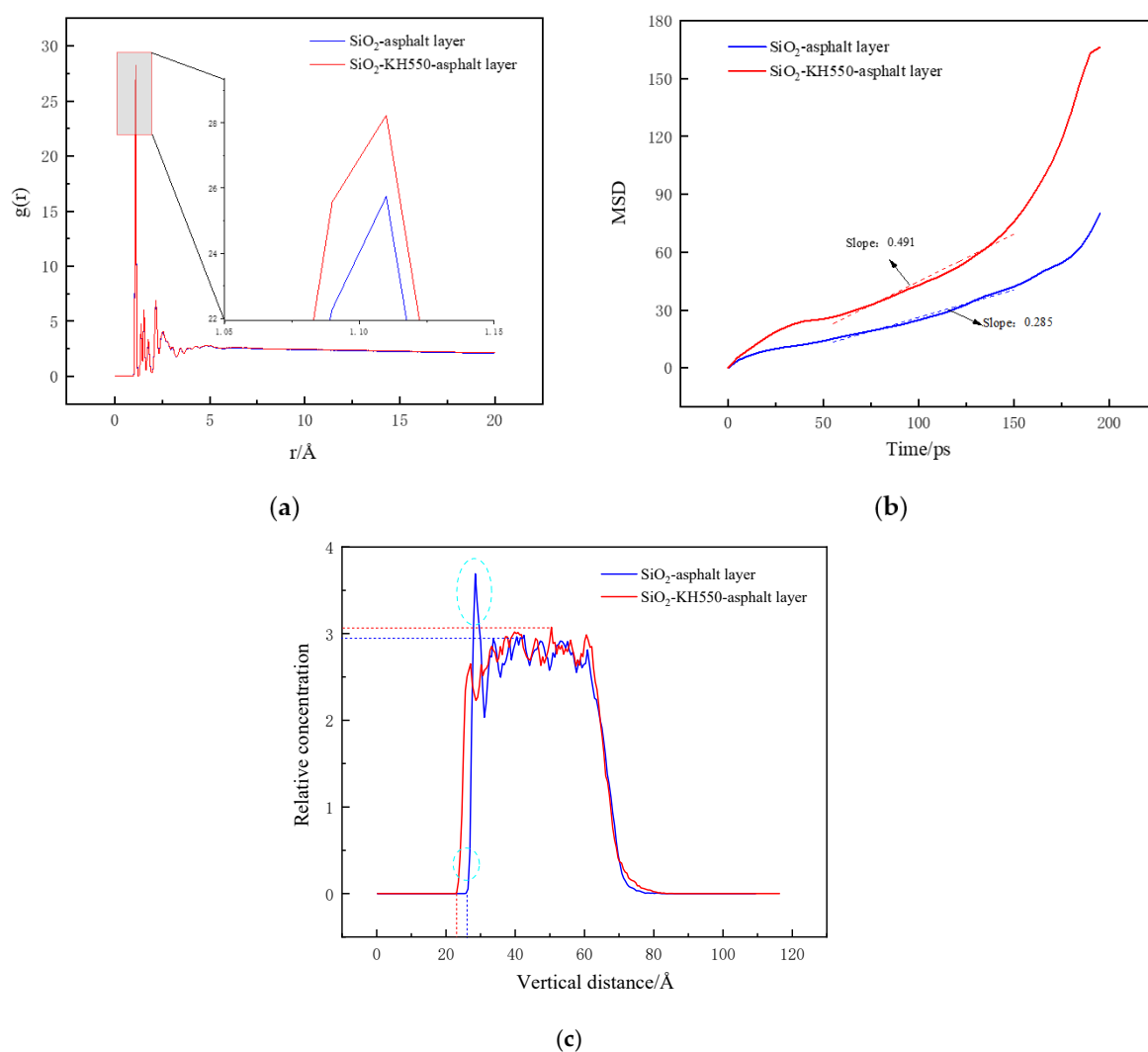


Figure 17. Interaction analysis of IT-asphalt interface before and after silane coupling agent modification: (a) RDF; (b) MSD; and (c) relative concentration.

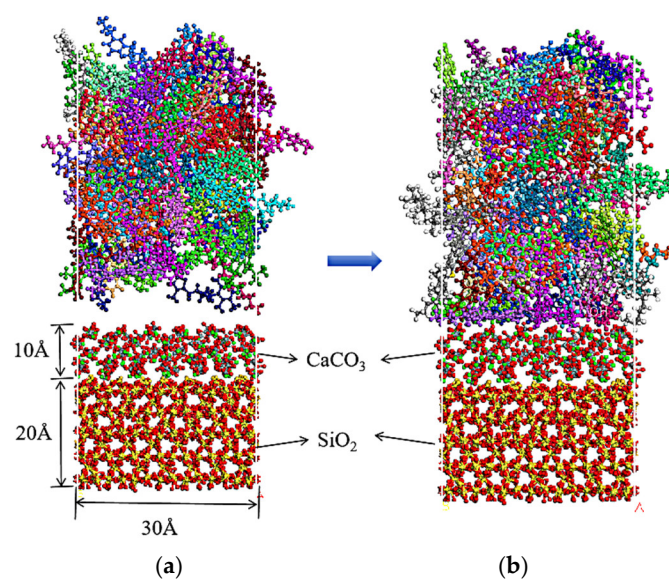


Figure 18. Analysis of IT-asphalt interface interaction modified by CaCO_3 : (a) before dynamic equilibrium and (b) after dynamic equilibrium.

The influence of alkaline minerals on the interaction behavior of the IT–asphalt interface was analyzed by the radial distribution function, root mean square displacement, and relative concentration change in asphalt molecules on the modified mineral surface. The simulation results are shown in Figure 18.

It can be seen from Figure 19a that after the surface of SiO_2 is modified by alkaline mineral CaCO_3 , the peak value of the radial distribution function of asphalt molecules on the surface of ITs becomes larger, and the aggregation of asphalt molecules becomes higher, indicating that the modified minerals have a stronger adsorption effect on asphalt. From Figure 19b, it can be seen that the slope and peak value of the root mean square displacement curve of asphalt on the surface of ITs are larger after modification, indicating that the diffusivity of asphalt molecules becomes stronger. With the increase in the simulation time step, the slope of the curve gradually becomes larger. The modified mineral has a stronger adsorption effect on asphalt, so that asphalt molecules have a tendency to accelerate the migration to the surface of ITs; it can be seen from Figure 19c that the concentration of asphalt molecules on the surface of alkaline ITs is significantly higher than that before modification, indicating that the adsorption of asphalt by modified ITs is enhanced. At this time, the distribution of asphalt molecules on the surface of ITs is more intensive. However, due to the increase in the thickness of the mineral layer after modification, the red curve in the figure shows an overall shift to the right.

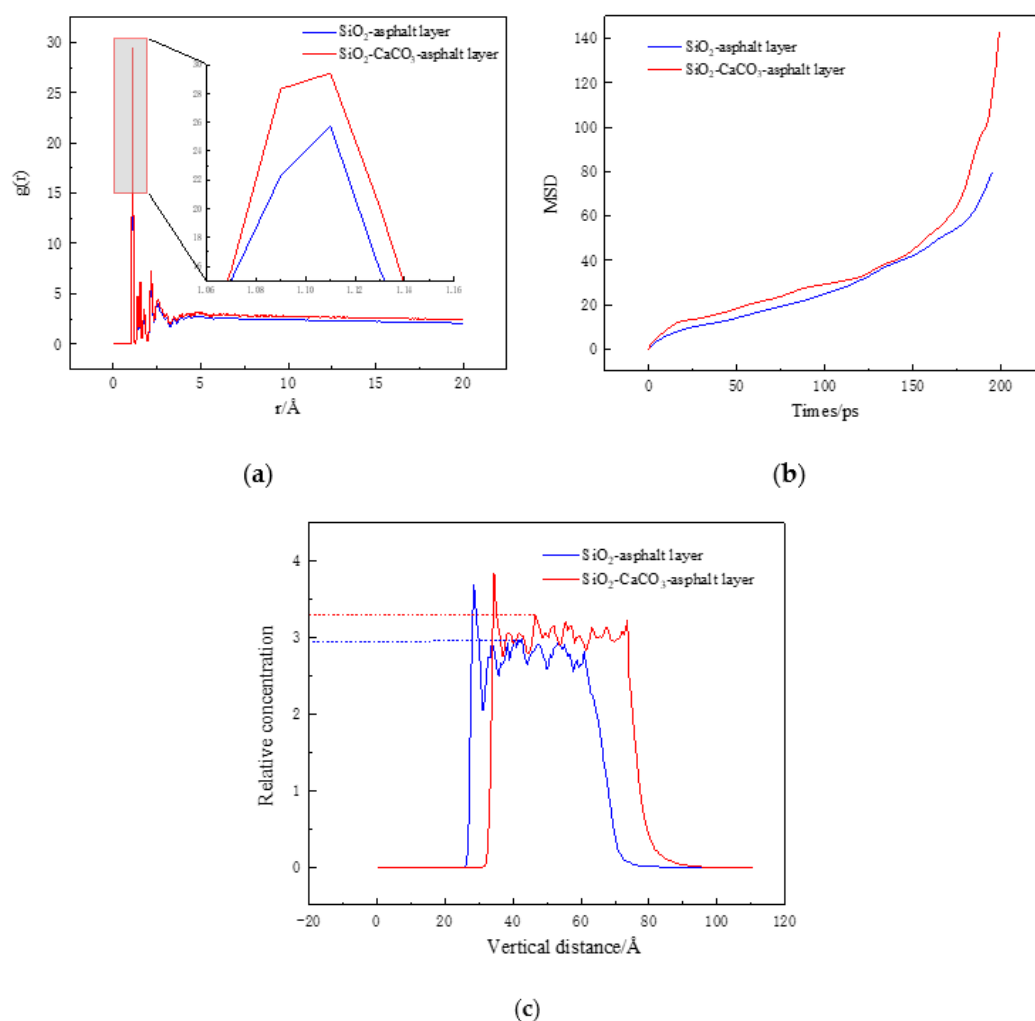


Figure 19. Interaction analysis of IT–asphalt interface before and after limestone mineral filler modification: (a) RDF; (b) MSD; and (c) relative concentration.

3.2. Study on the Interaction Mechanism of the Microscale IT–Asphalt Interface

3.2.1. Study on Surface Microscopic Parameters of IT–Asphalt Mastic

The surface roughness, micro-modulus and adhesion of the IT–asphalt mastic were tested by atomic force microscopy (AFM). For AFM testing, three observation positions on each sample were deliberately screened and selected with typical microstructural characteristics, ensuring representative morphological analysis. The bee structure distribution can be visually distinguished clearly, while quantitative roughness data are directly displayed alongside the AFM images, supporting reliable comparison without additional statistical processing. The bee-shaped structures in asphalt are usually rich in asphaltenes, and the bee structures are accompanied by continuous phases. This area is dominated by saturated phenols and other oils. The size, number and total area of the bee structures are closely related to the micro-surface parameters of asphalt. The surface roughness and adhesion images of asphalt mastic are shown in Figures 20 and 21.

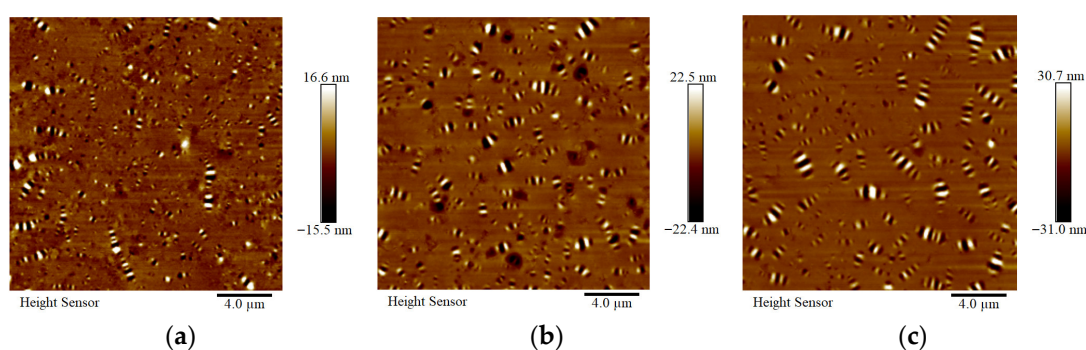


Figure 20. Surface roughness of asphalt mastic: (a) 70#-IT; (b) 70#-LF-IT; and (c) 70#-KH550-IT.

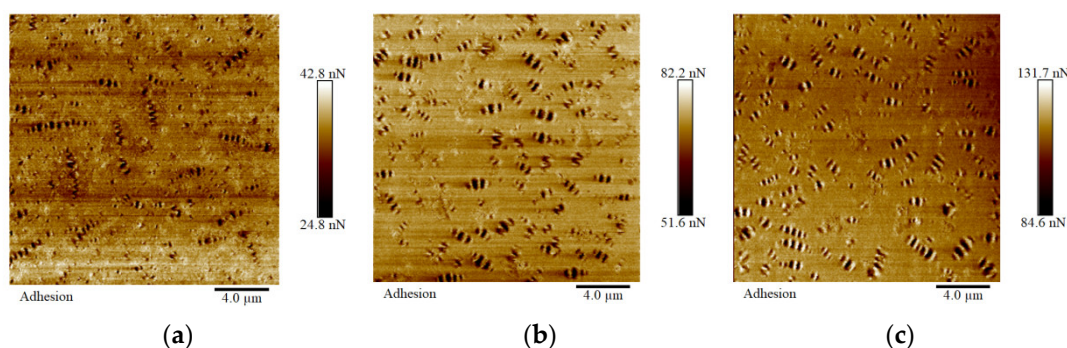


Figure 21. Microscopic adhesion of asphalt mastic surface: (a) 70#-IT; (b) 70#-LF-IT; and (c) 70#-KH550-IT.

It can be seen from Figure 20a that the bee structures present thin, short stripes, with individual lengths mostly ranging from 0.3 to 1.0 μm and small stripe spacing. The overall morphology is fragmented and delicate, which is a typical feature of the natural aggregation of asphaltene/wax crystals in unmodified asphalt. The bee structures of Figure 20b (limestone filler-modified group) are obviously thickened and elongated, with individual lengths increasing to 0.5–1.5 μm . Partial fusion and agglomeration of structures are observed, along with increased stripe spacing. The morphology transforms from fragmentation to coarsening, reflecting the physical filling and dispersion effect of mineral filler on asphalt components. The bee structures of Figure 20c (KH550-modified group) exhibit large, long-strip shapes, with individual lengths generally exceeding 1.0 μm (up to 2.0 μm). The stripes are regular with clear outlines, and the fluctuation amplitude is far greater than the other two groups, resulting from the uniform dispersion and ordered

aggregation of asphaltene after chemical modification by silane coupling agent. The unmodified 70#-IT group presents fragmented and dense bee structures with the smallest surface fluctuation ($-15.5\text{ nm}\sim+16.6\text{ nm}$), corresponding to the lowest surface roughness; the limestone-modified group shows coarsened bee structures with local agglomeration and increased fluctuation ($-22.4\text{ nm}\sim+22.5\text{ nm}$), with medium roughness; the KH550-modified group exhibits large-sized, long-strip bee structures with uniform distribution and significantly improved fluctuation ($-31.0\text{ nm}\sim+30.7\text{ nm}$), with the highest surface roughness. The bee structure of IT-asphalt mastic is the least, and the surface roughness is the smallest. The surface roughness of IT-asphalt mastic modified by silane coupling agent is the highest. The higher the surface roughness of asphalt mastic, the better the adhesion between asphalt mastic and aggregate. The order of surface roughness is $70\#\text{-KH550-IT} > 70\#\text{-LF-IT} > 70\#\text{-IT}$. It can be found that the surface roughness of IT-asphalt mastic modified by KH-550 is significantly improved, and the distribution of bee structure is larger and more uniform, indicating that the addition of silane coupling agent promotes the dispersion of asphaltene in asphalt mastic, optimizes the winding adsorption capacity of ITs and asphalt, improves the surface roughness of IT-asphalt mastic, and is conducive to improving its adhesion to coarse aggregate.

It can be seen from Figure 21 that the micro-adhesion force of asphalt mastic is quite different. Figure 21a shows that the overall adhesion force is at the lowest level, with a maximum value of only 42.8 nN and a minimum of 24.8 nN. The image shows a uniform light brown tone, indicating that the unmodified asphalt mastic has weak interfacial interaction and low surface adhesion. Figure 21b shows that the adhesion force is significantly improved compared to the control group, with a range of 51.6–82.2 nN and an amplitude of 30.6 nN. The image presents obvious bright and dark contrast, with scattered high-adhesion regions, reflecting the limited enhancement effect of physical modification by mineral filler. Figure 21c shows that the adhesion force reaches the highest level, with a maximum of 131.7 nN and a minimum of 84.6 nN, and the amplitude increases to 47.1 nN. The overall image is bright, with a large number of high-adhesion regions, showing a substantial improvement in surface adhesion after chemical modification. This order is completely consistent with the macroscopic adhesion test results, which directly verify that the improvement of micro-adhesion force is the micro-mechanism for enhancing the interfacial adhesion, water damage resistance, and fatigue performance of the asphalt mixture. The micro-adhesion force measured by AFM has a good linear correlation with the macroscopic asphalt–aggregate interfacial adhesion work. The order of micro-adhesion force of the three groups is $70\#\text{-KH550-IT} > 70\#\text{-LF-IT} > 70\#\text{-IT}$. The bee structure on the surface of IT-asphalt mastic is not obvious, and the micro-adhesion force is the smallest. The micro-adhesion force of the IT-asphalt mastic modified by silane coupling agent is the largest, and the highest adhesion force is 3.07 times that of the IT-asphalt mastic. The micro-adhesion force of the IT-asphalt mastic modified by limestone is 1.92 times that of the IT-asphalt mastic. The research shows that there is a good linear function relationship between the micro adhesion force and the interfacial adhesion work of asphalt aggregate [32]. Therefore, the greater the micro adhesion force is, the stronger the interfacial interaction between asphalt and aggregate is. Therefore, mineral filler and silane coupling agent can improve the interfacial adhesion characteristics of IT-asphalt mixture by improving the micro-adhesion force on the surface of the asphalt mastic.

3.2.2. Interaction Mechanism of IT–Asphalt Interface

The functional group index values of 70# asphalt binder, IT-asphalt mastic and limestone filler-asphalt mastic were obtained by Fourier infrared spectroscopy test and analysis, as shown in Figure 22.

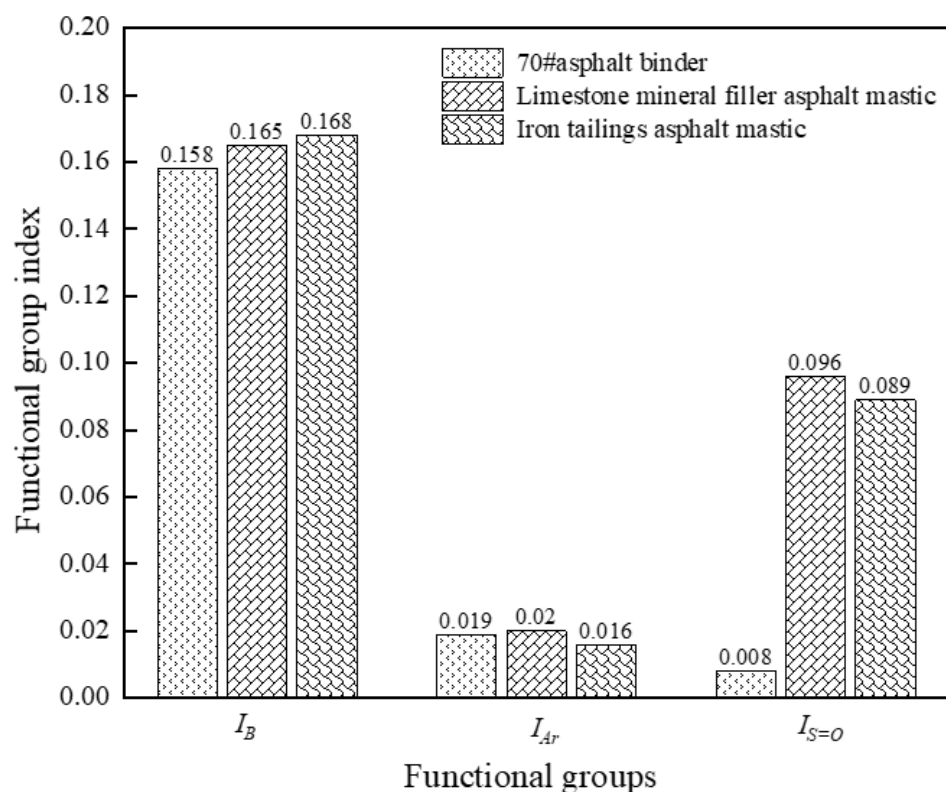


Figure 22. Functional group index of asphalt and asphalt mastic.

From Figure 22, it can be seen that saturated aliphatic groups dominate the composition of asphalt, while aromatic and sulfoxide groups are relatively few. When limestone filler and IT filler were mixed into asphalt mastic, the aliphatic functional group index showed an upward trend, while the aromatic functional group index remained relatively stable and did not fluctuate significantly. The index of the sulfoxide functional group of the asphalt mastic has been significantly improved. The sulfur atoms in the sulfoxide group form coordination bonds with the metal ions on the surface of the aggregate. The higher the sulfoxide index, the better the adhesion. The sulfoxide index of limestone filler is slightly higher than that of IT–asphalt mastic, indicating that the adhesion of limestone filler to asphalt mastic is higher.

The infrared spectral images of IT filler, limestone filler, 70 # matrix asphalt, and its prepared IT–asphalt mastic and limestone asphalt mastic were analyzed, as shown in Figure 23.

From Figure 23, it can be found that both figures are Fourier transform infrared (FT-IR) spectra of asphalt mastics and their raw materials, covering the wavenumber range of 4000–500 cm^{-1} . The analysis focuses on the characteristic absorption peaks, their chemical assignments, and the interaction mechanisms between asphalt and fillers (IT/limestone filler).

The FT-IR spectra of IT–asphalt mastic, 70# asphalt, and ITs are shown in Figure 23a. The characteristic peaks at 2922 cm^{-1} , 2856 cm^{-1} , and 1441 cm^{-1} correspond to the stretching and bending vibrations of aliphatic hydrocarbons ($-\text{CH}_2-$ and $-\text{CH}_3$) in asphalt, which are fully retained in the mastic, indicating that the chemical structure of asphalt remains stable after mixing with ITs. The peak at 1020 cm^{-1} is the characteristic absorption of Si-O-Si in ITs, which is clearly observed in the mastic. No significant shift or new peak is detected, proving that the interaction between ITs and asphalt is dominated by physical adsorption.

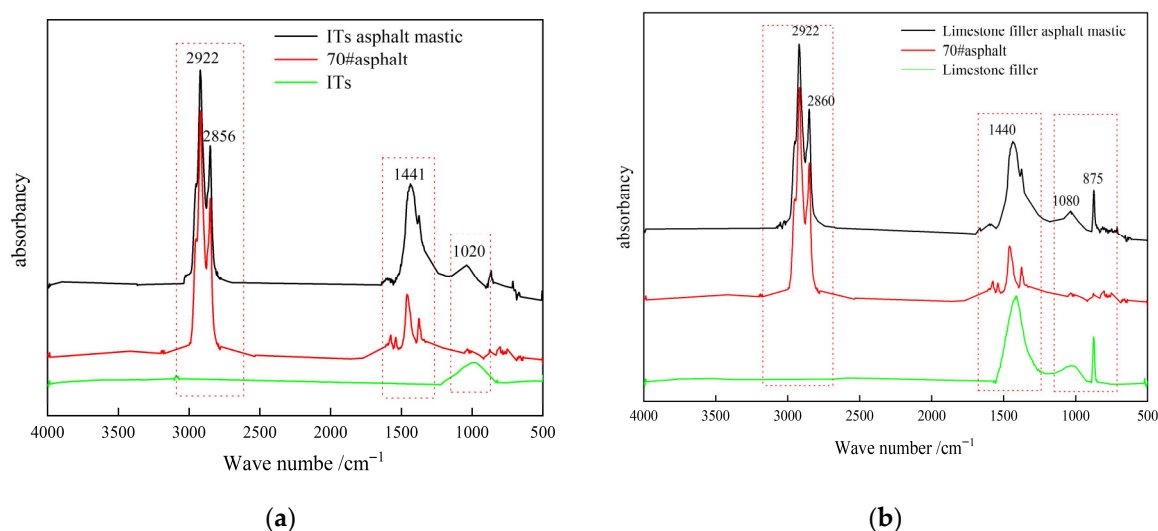


Figure 23. Infrared spectrum analysis of asphalt mastic: (a) ITs, asphalt, and IT–asphalt mastic; (b) limestone filler, asphalt, and limestone filler–asphalt mastic.

For the limestone filler system, the characteristic peaks of asphalt (2922 cm⁻¹, 2856 cm⁻¹, and 1440 cm⁻¹) are also fully retained. The unique peak at 875 cm⁻¹ of CaCO₃ in limestone filler is clearly visible in the mastic, and no chemical reaction peak is observed, indicating that the interaction between limestone filler and asphalt is also physical adsorption. These results confirm that FT-IR can only reflect the physical interaction between filler and asphalt, and the enhancement of interfacial adhesion requires further analysis by molecular dynamics simulation.

4. Conclusions

From the perspective of engineering practical applications, this paper systematically investigates the interfacial interaction characteristics between iron tailings (ITs) and asphalt by combining molecular dynamics simulation, atomic force microscopy (AFM) and Fourier transform infrared spectroscopy (FT-IR). The core research findings and practical engineering implications are summarized as follows.

(1) Molecular dynamics simulation results show that asphalt molecules exhibit higher aggregation degree, surface enrichment concentration and diffusion activity on Al₂O₃ and Fe₂O₃ surfaces compared to the SiO₂ surface. In practical engineering, a moderate content of Al₂O₃ and Fe₂O₃ in ITs can effectively strengthen the interfacial bonding performance between the IT aggregate and asphalt binder. Temperature exerts a differentiated influence on interfacial interaction: the bonding effect between asphalt and Al₂O₃/Fe₂O₃ reaches the optimal state at medium service temperature, while the interfacial adhesion of asphalt on CaCO₃ and SiO₂ surfaces gradually weakens with the increase in temperature. This conclusion provides a theoretical basis for the component optimization of IT raw materials in asphalt mixture proportion design.

(2) AFM microscopic tests confirm that alkaline mineral components can refine and densify the bee-like structure of IT–asphalt mastic and increase its surface roughness, which is conducive to improving the anti-stripping performance of the interface. After modification with silane coupling agent KH550, the surface micro-adhesion force of IT–asphalt mastic increases to 3.14 times that of the original material. The microscopic modification mechanism revealed in this study can guide the surface modification construction of ITs to improve the interfacial durability of asphalt pavement in actual road engineering.

(3) FT-IR analysis reveals that the characteristic absorption peaks of the iron tailings asphalt mastic are merely a simple superposition of those of neat asphalt and iron tailing

powder. This finding suggests that the interfacial bonding between unmodified iron tailing powder and asphalt mainly relies on physical adsorption, with no chemical reactions or new chemical bond formation occurring at the interface. Furthermore, combined with molecular dynamics simulation results, it is further confirmed that both limestone filler and silane coupling agent can enhance the interfacial interaction between asphalt and iron tailing powder, while the silane modifier exhibits a more prominent optimization effect at the microscale. Such differences in modification mechanisms may provide a favorable reference for selecting targeted modification strategies to improve the interfacial stability of iron tailings asphalt mastic under complex service environments.

This study still has certain limitations. It only investigates the microscopic interfacial mechanism under laboratory conditions, without exploring the long-term durability evolution of IT-asphalt materials in complex service environments. In addition, the molecular simulation adopts ideal parameters and lacks verification with practical engineering performance.

Future research will focus on the long-term durability deterioration mechanism of ITs modified asphalt materials, construct refined mineral models of iron tailings, optimize simulation parameters to fit actual engineering conditions, and conduct field application tests to promote the practical popularization of iron tailings in pavement engineering.

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