Research Square

Preprints are preliminary reports that have not undergone peer review.
They should not be considered conclusive, used to inform clinical practice,
or referenced by the media as validated information.

Laccase-Assisted Gallic Acid Functionalization of Corn Straw Fibers for Reinforcing SBS-Modified Asphalt Binder: Fiber Toughening, Rheological Performance, and Molecular-Scale Mechanisms

Mingxing Huang
Jilin University
Wenting Dai
Jilin University
Wei Guo
weiguo@cumt.edu.cn
China University of Mining and Technology
Shuo Yuan
China University of Mining and Technology
Gan Cao
China University of Mining and Technology
Kaipeng Gu
Jilin Jianzhu University

Article

Keywords: Corn straw fiber, Functional modification, Fiber toughening, Rheological performance, Molecular-scale mechanism

Posted Date: July 17th, 2026

DOI: https://doi.org/10.21203/rs.3.rs-10036930/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License.
Read Full License

Additional Declarations: No competing interests reported.

Laccase-Assisted Gallic Acid Functionalization of Corn Straw Fibers for Reinforcing SBS-Modified Asphalt Binder: Fiber Toughening, Rheological Performance, and Molecular-Scale Mechanisms

Mingxing Huang [a], Wenting Dai [a], Wei Guo[b,c,d*], Shuo Yuan[b], Gan Cao[b], Kaipeng Gu[c],

a College of Construction Engineering, Jilin University, Changchun, 130026, China;
b School of Mines, China University of Mining and Technology, Xuzhou 221000, China;
c School of Transportation Science and Engineering, Jilin Jianzhu University, Changchun 130000, China;
d Central Research Institute of Building and Construction Co., Ltd., MCC Group, Beijing 100082, China.

Abstract: This study investigated the regulatory effects of differently modified corn straw fibers (CSF)—a green fiber material derived from agricultural waste—on the mechanical properties, rheological characteristics, and molecular structural evolution of asphalt binders. Although plant fibers have been widely used in asphalt modification, natural CSF remains difficult to apply directly because of its insufficient toughness, poor structural stability, and weak interfacial compatibility. To address these limitations, CSF was modified using NaOH, tannic acid, and a gallic acid–laccase synergistic system. The effects and reinforcement mechanisms of different modification methods were revealed through fiber mechanical tests, abrasion tests, SEM, FTIR, XPS, dynamic shear rheological tests, and molecular dynamics simulations. The results showed that the gallic acid–laccase synergistic modification exhibited superior overall performance. Compared with unmodified fibers, the tensile strength, elastic modulus, and elongation at break of the modified fibers increased by 127.51%, 154.00%, and 46.18%, respectively, while the abrasion loss rate decreased by 56.85%. Microscopic characterization indicated that the gallic acid–laccase modification formed a functionalized layer rich in oxygen-containing functional groups on the

fiber surface, enhancing fiber surface activity and interfacial bonding potential. Rheological tests showed that GA-CSF significantly improved the high-temperature stiffness, elastic recovery ability, and resistance to permanent deformation of SBS-modified asphalt. Molecular dynamics simulations further indicated that GA-CSF increased the cohesive energy density and elastic modulus of the system while reducing the free volume fraction, suggesting enhanced intermolecular interactions and restricted molecular motion in asphalt. These findings demonstrate that gallic acid–laccase synergistic modification is an effective method for improving the road applicability of corn straw fibers and the high-temperature performance of asphalt binders.

Keywords: Corn straw fiber; Functional modification; Fiber toughening; Rheological performance; Molecular-scale mechanism.

1. Introduction

Asphalt binder is the key bonding phase in asphalt pavement materials, and its viscoelastic properties, interfacial adhesion, and high-temperature deformation resistance directly affect pavement service stability [1-3]. Fiber reinforcement technology has been widely used to improve the performance of asphalt materials [4]. Owing to their wide availability, low density, low cost, and renewability, plant fibers have gradually become promising alternatives to mineral fibers, polymer fibers, and commercial lignin fibers [5]. Akarsh et al. investigated the application of sisal and coconut coir fibers in stone mastic asphalt (SMA) mixtures and found that natural fibers effectively reduced asphalt drainage while improving the indirect tensile strength, rutting resistance, and fatigue performance of the mixtures [6]. Zhang et al. evaluated the effects of lignin, basalt, polyester, and polyacrylonitrile fibers on the performance of open-graded friction course

(OGFC) mixtures [7]. Their results showed that fibers improved the structural stability of open-graded asphalt mixtures by adsorbing asphalt and strengthening the bonding between aggregates.

Corn straw is a typical lignocellulosic agricultural residue, primarily composed of cellulose, hemicellulose, and lignin, with a porous structure, low density, and inherent fiber skeleton strength. Zhang et al. separated and characterized the cellulose, hemicellulose, and lignin in corn straw, reporting contents of 36.89%, 20.42%, and 17.38%, respectively [8]. Lou et al. applied xylanase and immobilized laccase pretreatment to extract cellulose from corn straw, showing that this method effectively removed non-cellulosic components while enhancing the purity and crystallinity of cellulose [9]. Caixeta Silva et al. investigated the effects of different alkali treatments on corn straw fibers, finding that NaOH treatment improved fiber crystallinity and tensile strength and strengthened the adhesion between fibers and the matrix [10]. Kavitha et al. conducted chemical modifications on Zea mays fibers, further demonstrating that surface treatments could regulate fiber morphology, moisture absorption, and mechanical properties [11]. Collectively, these studies indicate that corn straw fibers possess controllable composition and surface characteristics, providing a theoretical basis for their application as reinforcing materials in asphalt binders.

Existing studies have explored the incorporation of corn stalks and their derivatives into asphalt materials. Li et al. compared the effects of corn stalk fibers, sugarcane bagasse fibers, bamboo fibers, cotton stalk fibers, and lignin fibers on asphalt binder performance, highlighting that differences in microstructure, chemical composition, and thermal stability of plant fibers significantly influence asphalt modification [12]. Hu et al. employed hydrothermal carbon derived from corn stalks for bio-asphalt modification, demonstrating its potential to enhance the high-temperature performance of asphalt [13]. Chen et al. investigated corn stalk biochar in road asphalt,

showing that it could reduce the emission of harmful components in asphalt fumes while maintaining favorable rheological properties [14]. Collectively, these studies indicate that corn stalks can be applied in asphalt modification through fiberization, carbonization, and other approaches, broadening the utilization forms of corn stalks in pavement materials.

However, natural corn straw fibers still struggle to directly meet the reinforcement needs of asphalt binders. Their surface is rich in hydroxyl groups, which exhibit strong hydrophilicity and insufficient compatibility with hydrophobic asphalt matrices. The presence of hemicellulose, waxy substances, pectin, and amorphous components can weaken the thermal stability and interfacial adhesion of the fibers. More critically, natural corn straw fibers have poor toughness and are prone to wear, breakage, and structural damage during cutting, crushing, screening, mixing, and shearing processes [15,16]. Once the effective length and intact morphology of the fibers are difficult to maintain, their bridging, reinforcing, and stress-transferring effects in asphalt will be significantly reduced. Therefore, enhancing the toughness of corn straw fibers is the core issue for their effective application as asphalt reinforcement materials [17,18].

Addressing the challenges of natural fibers, including strong hydrophilicity, weak interfacial compatibility, and insufficient structural stability, various surface modification strategies have been explored. Kamaruddin et al. demonstrated that NaOH treatment can remove hemicellulose, lignin, wax, and surface impurities from natural fibers, reducing water absorption and improving interfacial bonding with the matrix [19]. Shifa et al. reported that alkali treatment of wheat straw fibers significantly enhanced the tensile and flexural strengths of epoxy composites [20]. Caixeta Silva et al. further confirmed that alkali treatment can regulate the functional performance of corn straw fibers in composites [21]. Stepczyńska et al. modified flax fibers with tannic acid, highlighting that polyphenol treatments improve interfacial adhesion in biocomposites [22]. Li et

al. performed hydrophobic modification of cellulose nanofibers using gallic acid, demonstrating its effectiveness in regulating surface properties [23]. Collectively, these studies indicate that surface modification is a key approach to enhancing the engineering applicability of natural fibers. However, most existing research focuses on macroscopic changes in asphalt properties after fiber incorporation, with limited systematic understanding of how fiber modification influences bulk toughness, resistance to mechanical damage during processing, evolution of surface functional groups, and the stability of asphalt molecular structure.

Based on the identified research gaps, this study proposes a strategy for toughening and modifying corn straw fibers (CSF) to enhance asphalt binders. CSF were treated with NaOH, tannic acid, and a gallic acid–laccase (GA) system, and the effects of different modifications on fiber toughness, structural stability, and surface chemistry were systematically compared. Fiber mechanical performance and damage resistance were evaluated through tensile and wear tests, while changes in surface morphology, functional groups, and elemental composition were characterized using SEM, FTIR, and XPS. Subsequently, the influence of GA-CSF on the high-temperature viscoelastic behavior and resistance to permanent deformation of SBS-modified asphalt was assessed via dynamic shear rheometry, and molecular dynamics simulations were conducted to elucidate the regulatory mechanism of GA-CSF on intermolecular interactions, free volume, and mechanical response within the asphalt system. The results provide both experimental evidence and molecular-scale insights for the toughening and modification of corn straw fibers, supporting their application in sustainable asphalt binders.

2. Materials and Sample Preparation

2.1 Materials

2.1.1 Corn stover fiber

Corn stover fiber (CSF) was obtained from discarded corn straw collected after harvesting in a corn-growing area of Changchun, Jilin Province, China. The raw material was processed through natural cleaning, husk–pith separation, cutting, mechanical crushing, and screening, as shown in Fig. 1. The resulting fibers were light-yellow short fibers with a length of no more than 6 mm, a moisture content of 4.5%, an ash content of 5.2%, and a pH of 7.5.

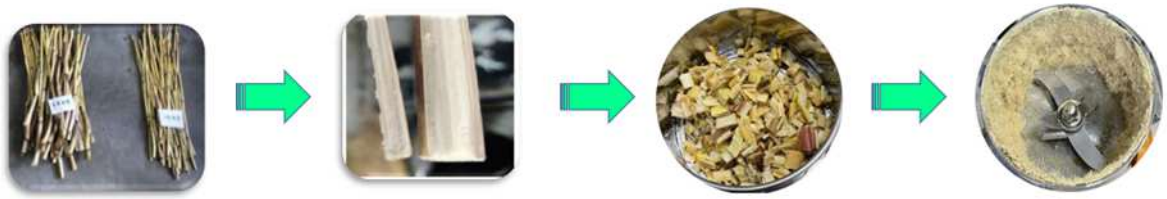


Fig. 1. Corn Stover Fiber Preparation Process

2.1.2 Modifiers

The modifiers employed in this study include sodium hydroxide (NaOH), tannic acid (TA), and a gallic acid–laccase system (GA), all commonly used for surface modification of corn straw fibers. Their chemical characteristics and functional roles are as follows:

（1） Sodium hydroxide (NaOH)

NaOH is a strong alkali that selectively removes hemicellulose, lignin, pectin, and impurities from the fiber surface. This disrupts the original dense structure, improving fiber surface quality and facilitating subsequent functionalization.

（2） Tannic acid (TA)

TA is a natural polyphenol rich in phenolic hydroxyl groups, capable of forming hydrogen bonds with fiber surface hydroxyl groups. It enhances structural stability through physical adsorption and chemical interactions, and improves bonding between fiber bundles.

（3） Gallic acid–laccase system (GA)

Gallic acid, a natural polyphenol with three hydroxyl groups and one carboxyl group, reinforces the internal and surface structures of fibers via hydrogen bonding and condensation reactions. Laccase, a multicopper oxidase, catalyzes the oxidation of gallic acid to quinone intermediates, accelerating polymerization and crosslinking to form a dense functional layer. This process simultaneously improves fiber toughness and wear resistance. The main technical parameters are summarized in Table 1.

Table 1 Main technical parameters of the modifiers

Modifier	Purity (%)	pH or Enzyme Activity	Key Parameters
NaOH	≥96.0	—	Melting point 318.4 °C; insoluble matter ≤0.05%
Tannic acid	≥98.0	pH≈2.5	Total phenolic hydroxyl content ≥10.0 mmol/g
Gallic acid	≥99.0	pH≈3.0	Ash content ≤0.1%
Laccase	≥85.0	Enzyme activity ≥10000 U/g	Optimal pH ≈ 4.5; optimal temperature 60 °C

2.1.3 SBS-modified asphalt

In this study, I-B type SBS-modified asphalt was selected as the binder matrix. Its main technical indicators are summarized in Table 2. Characterizing the fundamental properties of SBS-modified asphalt provides a baseline for assessing the performance of the raw material and serves as a reference for evaluating the effects of GA-CSF on the rheological behavior of asphalt binders.

Table 2 Technical properties of SBS-modified asphalt binder

Test Item	Unit	Test Result	Technical Requirement

Penetration at 25 °C, 100 g, 5 s	0.1mm	103	≥100
Penetration Index (PI)	/	-0.90	≥-1.2
Softening Point	℃	48.5	≥45
Mass change after TFOT	%	-0.495	-1.0～+1.0
Penetration ratio after TFOT	%	58.2	≥55
Ductility at 5 °C	cm	33	≥30

2.2 Sample Preparation

2.2.1 Preparation of modified corn straw fiber

To prepare modified corn stover fibers, this study employed NaOH, tannic acid (TA), and gallic acid–laccase (GA) systems. NaOH-CSF was obtained by immersing fibers in 5% NaOH solution at a solid-to-liquid ratio of 1:20 (g: mL), stirring at 500 r/min for 30 min at room temperature, followed by filtration, washing until neutral, and drying at 60 °C for 4 h. TA-CSF was prepared using 10% tannic acid solution under the same solid-to-liquid ratio, stirred in the dark at room temperature for 60 min, allowed to stand for 2 h, then filtered, washed, and dried. GA-CSF was prepared by treating fibers with 5% gallic acid solution and laccase (10 U/g fiber) at a solid-to-liquid ratio of 1:20 (g: mL) at 45 °C for 2 h under 300 r/min stirring, followed by filtration, washing until neutral, and drying at 60 °C for 4 h. The detailed preparation process of modified corn stover fibers is illustrated in Figure 2. For clarity, untreated corn stover fibers, sodium hydroxide-modified fibers, tannic acid-modified fibers, and gallic acid–laccase-modified fibers are denoted as CSF, NaOH-CSF, TA-CSF, and GA-CSF, respectively.

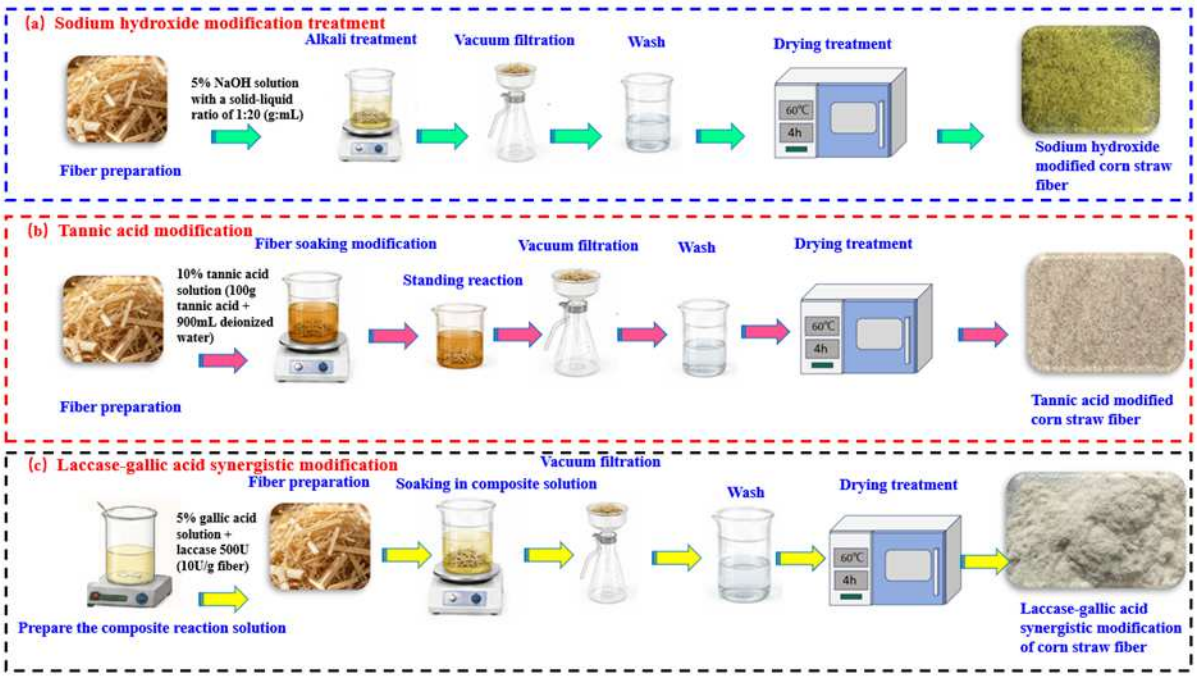


Fig. 2. Preparation process of modified corn stover fibers

2.2.2 Preparation of Asphalt Modified with Corn Stover Fibers

Based on preliminary screening of fiber bulk properties and wear resistance, gallic acid–laccase modified fibers (GA-CSF) exhibited optimal strength, toughness, and structural stability. Therefore, subsequent asphalt preparation and rheological evaluations focused on GA-CSF to investigate its reinforcing effect in SBS modified asphalt. Fiber contents were set at 0.6%, 0.8%, and 1.0% by mass of asphalt, referencing commonly used levels for natural plant fibers. This range ensures effective fiber adsorption and reinforcement while avoiding agglomeration due to excessive fiber content. To minimize moisture-related adverse effects, fibers were dried in a vacuum oven at 60°C for 24 hours until constant weight, maintaining moisture below 0.5%. SBS modified asphalt is denoted as SBS, while corn straw fiber-reinforced asphalt with 0.6%, 0.8%, and 1.0% fiber content are denoted as GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0%, respectively. The preparation procedure is as follows:

（1） Asphalt Preheating

A predetermined amount of SBS asphalt was placed in a beaker and heated at 180°C for 60 minutes to ensure complete melting and good fluidity.

（2） Fiber Addition and Preliminary Dispersion

The molten asphalt was stirred at 500 r/min using a low-speed high-shear mixer. GA-CSF was added gradually in portions not exceeding 1/5 of the total mass, with 30-second intervals between additions to prevent fiber agglomeration.

（3） High-Speed Shearing

After all fibers were added, the rotational speed was increased to 5000 r/min and shearing continued for 40 minutes. The system temperature was maintained at 160±5°C using an oil bath to ensure suitable asphalt viscosity and avoid thermal degradation.

（4） Defoaming and Stabilization

The rotational speed was reduced to 1500 r/min and stirred for an additional 20 minutes to remove entrapped air and promote fiber–asphalt integration, enhancing system stability.

（5） Sample Storage

The modified asphalt was transferred hot to clean, sealed containers, cooled to room temperature, and heat-preserved in a 60°C oven for 2 hours. Samples were stored away from sunlight and contaminants until testing.

3. Methods and laboratory tests

3.1 Fiber Basic Performance Testing

3.1.1 Fiber tensile test

A Shenzhen Sansi UTM4103 tensile testing machine was used to evaluate the tensile properties of the fibers. The instrument has a force measurement range of 0–500 N with an

accuracy of ±0.5% FS. The tensile rate can be adjusted from 1 to 500 mm/min, and the displacement measurement accuracy is ±0.01 mm. The test was conducted in accordance with GB/T 14337-2008, Test Method for Tensile Properties of Chemical Fiber Staple Fibers.

3.1.2 Fiber wear detection

To evaluate the wear resistance of corn stover fibers during mixing, the wear loss rate of different modified fibers was determined using a method simulating asphalt mixture mixing conditions, following the testing concept of GB/T 26743—2011. In the test, fiber samples were weighed at a fiber content of 4% and mixed with aggregates in a mixing pot for 90 s. After mixing, aggregates larger than 9.5 mm were removed, and the sieve residue within the 0.15–0.30 mm particle size range was retained. The residual fiber mass after mixing was calculated by subtracting the aggregate mass within the same particle size range, and the wear loss rate was then determined.

3.2 Microstructural and chemical characterization of corn stover fibers

3.2.1 SEM electron microscopy scanning

Fiber samples—unmodified, tannic acid-modified, gallic acid–laccase-modified, and sodium hydroxide-modified—were cut into 5 mm segments and evenly mounted on the sample stage. After gold sputtering, the samples were observed in the SEM chamber at magnifications of 500×, 1000×, and 3000× under an accelerating voltage of 5.0 kV. Micrographs from different fields of view were captured to analyze fiber surface characteristics, including roughness, etching, presence of surface deposits, and structural integrity.

3.2.2 FTIR test

FTIR analysis was conducted on unmodified, sodium hydroxide-modified, tannic acid-modified, and gallic acid–laccase-modified corn straw fibers using a Bruker INVENIO Fourier Transform Infrared Spectrometer (Germany). The measurements were performed over a

wavenumber range of 4000–400 cm^{-1} with a spectral resolution of 1 cm^{-1}, 32 scans, and a signal-to-noise ratio of 60,000:1. Samples were prepared using the KBr pellet method, in which fibers were mixed and ground with dried KBr at a ratio of 1:100, then pressed into pellets to ensure stable and reliable spectra. To compare relative changes in characteristic functional groups across samples, baseline correction and normalization were applied, and the integral method was used to calculate the relative peak areas of the main characteristic peaks. Since the spectra were normalized, peak areas are expressed as relative integrated areas, serving for semi-quantitative comparison among samples.

3.2.3 XPS analysis

In this study, Thermo Scientific K-Alpha+ X-ray photoelectron spectrometer was used to test unmodified corn straw fibers, sodium hydroxide-modified corn straw fibers, tannic acid-modified corn straw fibers, and gallic acid-laccase-modified corn straw fibers. The sample preparation method employed was the dry pellet method. The fiber samples were dried at 50°C for 2 hours, then ground into powder, and evenly pressed onto conductive tape to ensure a flat surface and no impurity contamination. The test results were obtained as the average of three characteristic points to ensure data reliability.

3.3 Rheological test of asphalt binder

To evaluate the influence of gallic acid–laccase-modified corn straw fiber (GA-CSF) on the macroscopic rheological behavior of asphalt binders, dynamic shear rheological tests were conducted using an Anton Paar SmartPave 102 rheometer. Frequency sweep tests were performed at 35, 45, 55, and 65 ℃ over a frequency range of 0.2–30 Hz, with a strain of 0.1% to ensure measurements remained within the linear viscoelastic region. Multiple stress creep recovery (MSCR) tests were carried out following AASHTO TP 70, employing parallel plates with a

diameter of 25 mm and a gap of 1 mm. Tests were conducted at stresses of 0.1 kPa and 3.2 kPa at 64 ℃, with 10 loading–recovery cycles at each stress level. Temperature sweep tests were performed in accordance with AASHTO T 315-12, using an angular frequency of 10 rad/s and temperatures of 46, 52, 58, 64, and 70 ℃ [24].

3.4 Molecular simulation

In this study, Materials Studio 2020 was employed to construct a molecular model of SBS-modified asphalt, while the cellulose structure was used to represent the primary skeleton of corn straw fibers. Based on this, a model of gallic acid–laccase-modified corn straw fiber (GA-CSF) was established by incorporating gallic acid grafting structures to capture the chemical characteristics of the fiber surface after modification. To investigate the effect of GA-CSF on the molecular behavior and mechanical properties of asphalt binders, molecular models of modified asphalt containing GA-CSF at 0.6%, 0.8%, and 1.0% by mass of asphalt were constructed, followed by molecular dynamics simulations.

3.4.1 Gallic acid modified corn straw fiber model

Based on previous studies, cellulose constitutes the main structural component of corn straw, largely determining its mechanical load-bearing capacity and reinforcement potential. Since cellulose predominantly forms the fiber skeleton and mediates interfacial interactions, it was selected in this study to represent the primary load-bearing framework of corn straw fibers. Gallic acid functionalization was then incorporated to model the modified fiber surface, enabling investigation of its influence on asphalt molecular behavior. The model, illustrated in Figure 3, is a simplified representation primarily aimed at elucidating the trends of molecular interactions between modified fibers and asphalt systems.

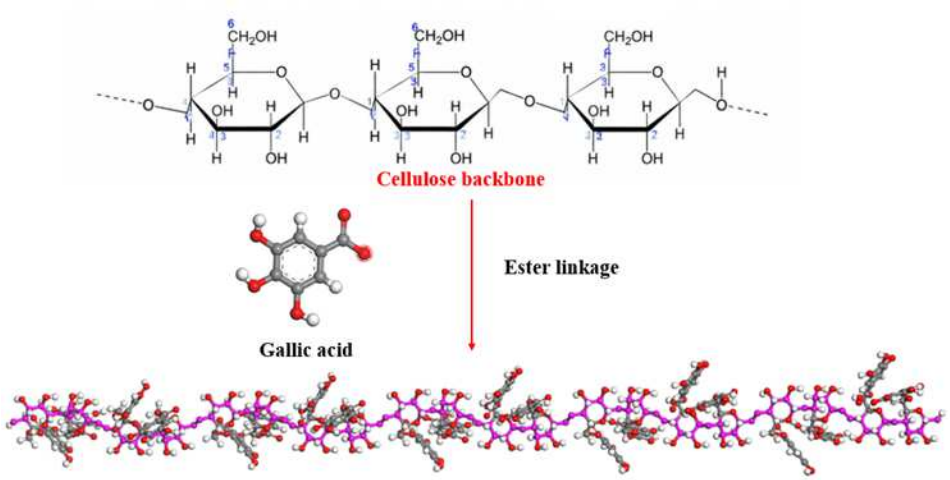


Fig. 3. Gallic acid–modified corn straw fiber model

3.4.2 Asphalt molecule

Based on the SARA fractionation, asphalt binder can be regarded as a complex organic system comprising saturates, aromatics, resins, and asphaltenes. To represent the compositional characteristics of SBS modified asphalt and its molecular interactions with gallic acid–treated fibers, representative molecules of each component were selected to construct an asphalt molecular model. The Amorphous Cell module in Materials Studio was used to generate an amorphous asphalt system with an initial density of 0.1 g/cm³ to minimize molecular overlap and local packing. Geometry optimization was performed using the Forcite module under the Compass II force field, with 50,000 iterations and the Smart method. The optimized asphalt molecules were then annealed with the Nose thermostat and Andersen barostat at a time step of 1 fs. Following geometric optimization and annealing, the system underwent a 300 ps NPT equilibrium simulation at 298.15 K and 1 atm, with a time step of 1 fs and trajectories saved every 1000 steps, followed by a 200 ps NVT simulation at 298.15 K. Once the system energy and volume stabilized, the equilibrium configuration was selected for subsequent analysis [25,26]. The molecular structures

of each asphalt component are shown in Figure 4, and the assembled asphalt model is presented in Figure 5.

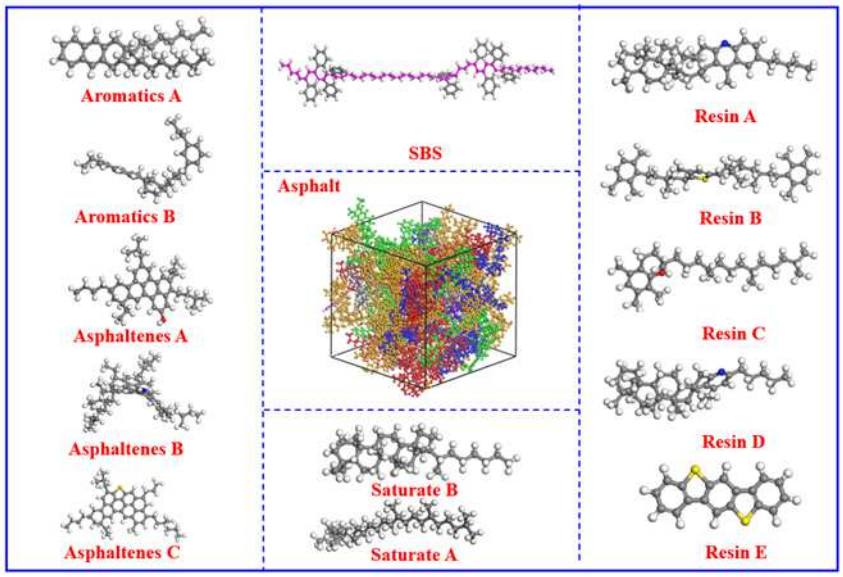


Fig. 4. Molecular models of asphalt components

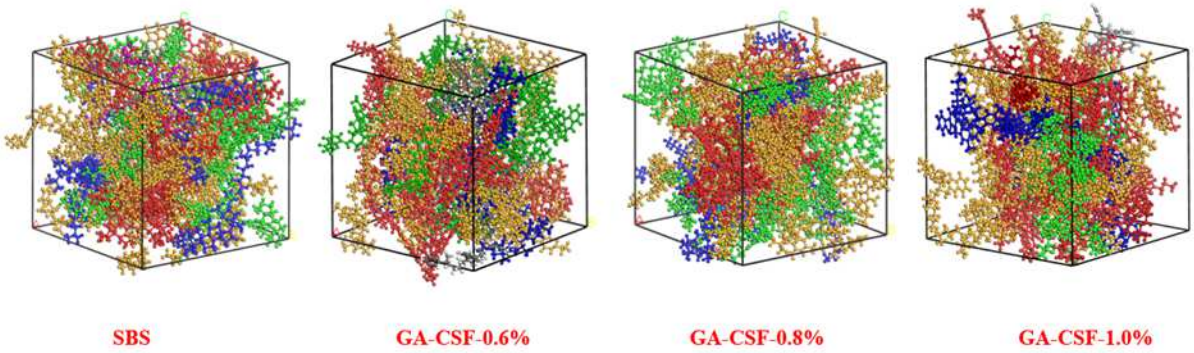


Fig. 5. Molecular model of GA-CSF-modified asphalt

3.5 Validation of molecular models

To verify the rationality and stability of the constructed asphalt molecular models, the thermodynamic and structural characteristics of different systems were evaluated based on molecular dynamics simulations. Key parameters, including system density and the radial

distribution function, were analyzed to determine whether the models reached equilibrium and to further confirm their suitability for subsequent molecular behavior analysis.

3.5.1 Density

As shown in Figure 6, the density of the four modified asphalt systems increased rapidly during the initial stage of simulation, gradually stabilized, and reached equilibrium after 200 ps. This indicates that the internal free volume of the models was effectively reduced and the molecular arrangement became reasonable. After equilibrium, the density of the SBS asphalt model was 0.950 g/cm³, while the densities of GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% modified asphalt were 0.973, 1.030, and 1.038 g/cm³, respectively. The increasing GA-CSF content corresponded to a rising system density, suggesting that the introduction of GA-CSF enhanced molecular packing and overall compactness of the asphalt system. Minor fluctuations in the final densities of each model remained within the typical density range of asphalt binders, confirming that the constructed molecular models were stable and reliable.

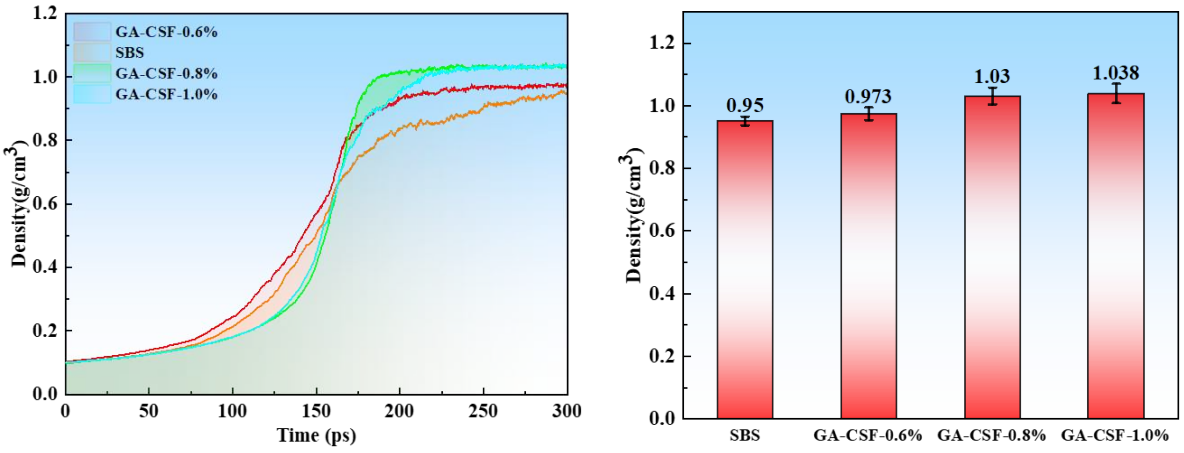


Fig. 6. Density of asphalt models

3.5.2 RDF

The radial distribution function (RDF) was used to characterize the local ordering and spatial distribution of the asphalt molecular systems. As shown in Figure 7, the (g(r)) values of the four

asphalt systems were zero within 0.9 Å, indicating the absence of significant atomic overlap or unreasonable close contacts, and confirming the rationality of the optimized initial structures. At (r = 1.11) Å, a pronounced peak appeared in all four asphalt systems, indicating stable short-range interactions within the systems. As the distance increased, the RDF curves gradually decayed and approached 1, suggesting the absence of long-range ordered arrangements, which is consistent with the amorphous nature of asphalt binders. The peak positions of the different modified asphalt models were nearly identical, whereas the peak intensities differed, indicating that GA-CSF altered local molecular packing without disrupting the overall structural rationality of the system.

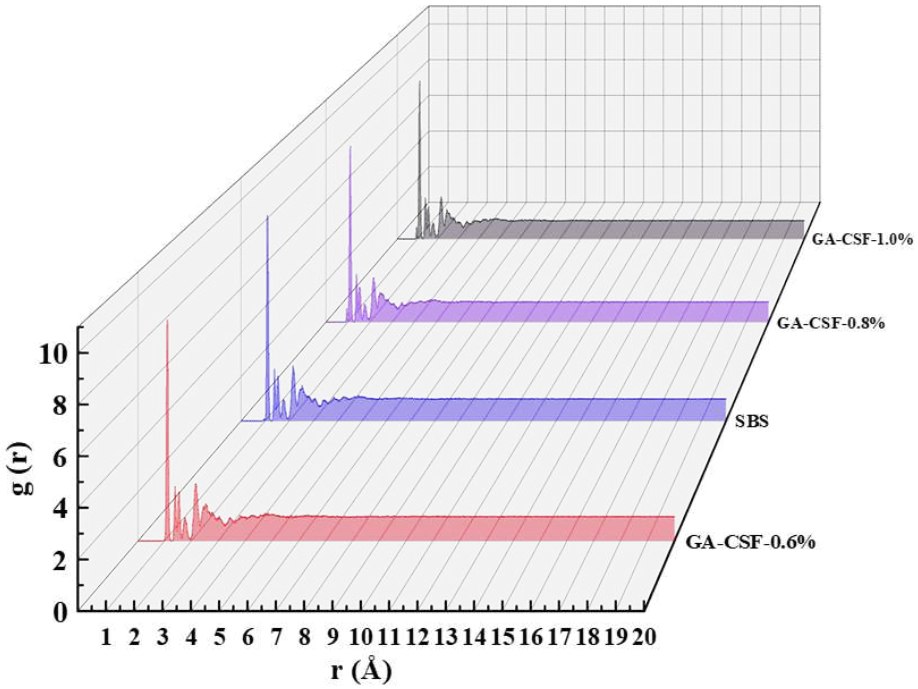


Fig. 7. RDF curves of the four asphalt binders

3.6 Molecular simulation evaluation metrics

To characterize the intermolecular interaction strength and component compatibility in the corn straw modified asphalt system, this study calculated the cohesive energy density and

solubility parameter [27,28]. The cohesive energy density reflects the energy level corresponding to molecular aggregation per unit volume in the system and can be used to evaluate the strength of intermolecular attraction. The solubility parameter, derived from the cohesive energy density, can be used to analyze the thermodynamic matching degree between different components. The relevant calculation formulas are as follows.

$$\delta = \frac{E_{coh}}{V} = \sqrt{E_{CED}} \tag{1}$$

In the formula, δ represents the solubility parameter of asphalt; V is the total volume of the system;E_{coh} refers to the energy accumulated within the asphalt system; and E_{CED} is the cohesive energy density.

In this study, Materials Studio 2020 was used to analyze and calculate the free volume of asphalt molecules using the Atom Volumes and Surfaces tools [29,30].

$$FFV = \frac{v_{free}}{v_{total}} = \frac{v_{free}}{v_{free} + v_{occupied}} \tag{2}$$

In the equation, FFV represents the free volume fraction in the system; v_{free} denotes the free volume (Å³) in the model system, and $v_{occupied}$ represents the occupied volume (Å³) in the system.

4. Results and Discussion

4.1 Analysis of Basic Performance Test Results of Fibers

In this study, a constant-rate extension tensile tester was used to measure the breaking strength and elongation at break of unmodified and modified corn stover fibers. The purpose was to evaluate the effects of different modification systems on the tensile properties and toughening behavior of the fibers. The test results are presented in Table 3.

Table 3 Tensile test results of modified corn stover fibers

Fiber type	Maximum force (N)	Tensile strength (MPa)	Elastic modulus (MPa)	Elongation at break (%)
CSF	3.42	75.67	3821.51	2.11
TA-CSF	4.33	125.15	5342.43	1.90
GA-CSF	8.45	172.16	9706.60	3.08
NaOH-CSF	1.37	27.90	4664.07	0.59

As shown in Table 3, the laccase–gallic acid synergistic modification exhibited the most pronounced improvement in the mechanical properties of corn stover fibers. The tensile strength, elastic modulus, and elongation at break increased by 127.51%, 154.00%, and 46.18%, respectively, indicating that this treatment simultaneously enhanced the load-bearing capacity and toughness of the fibers. This improvement may be attributed to the hydrogen-bonding network formed by the polyhydroxyl structure of gallic acid and the enzymatic crosslinking effect. Tannic acid modification increased the tensile strength and elastic modulus by 65.38% and 39.80%, respectively, but reduced the elongation at break by 9.50%, suggesting that its reinforcing effect was accompanied by a certain degree of embrittlement. In contrast, NaOH treatment decreased the tensile strength and elongation at break by 63.13% and 71.70%, respectively, indicating that alkali erosion damaged the structural integrity of the fibers.

During service, corn stover fibers are subjected to mechanical actions such as aggregate friction, shearing, and compression, which may cause surface abrasion, fiber shortening, and fracture, thereby weakening their bridging and reinforcing effects in asphalt systems. Therefore, wear resistance is an important indicator for evaluating the structural stability and engineering

applicability of corn stover fibers. The wear resistance results of different corn stover fibers are shown in Figure 8.

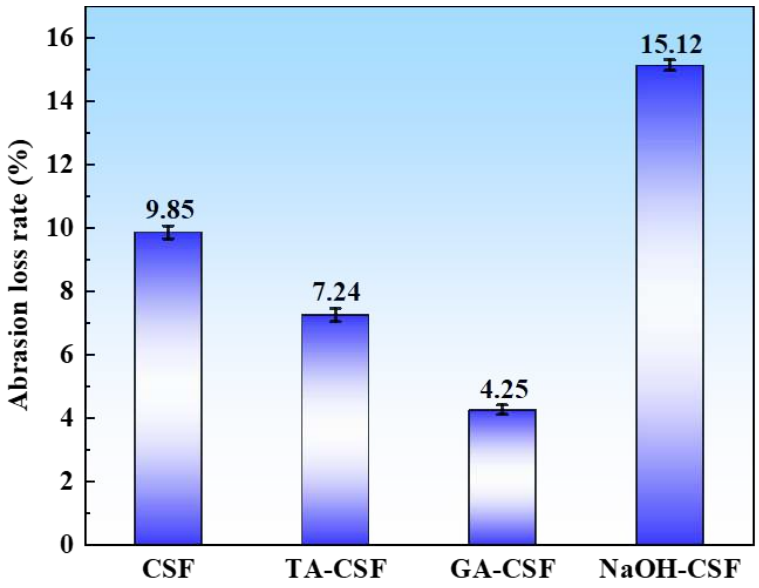


Fig. 8. Wear resistance test results of different corn stover fibers

As shown in Figure 8, corn stover fibers exhibited markedly different wear resistance after different modification treatments. The abrasion loss of unmodified CSF was 9.85%, indicating that its natural lignocellulosic structure was prone to abrasion and fracture under aggregate friction and mixing shear. After tannic acid modification, the abrasion loss decreased to 7.24%, suggesting that polyphenol molecules improved the bonding state of the fiber surface and enhanced structural stability. Laccase–gallic acid-modified fibers showed the lowest abrasion loss of 4.25%, representing a 56.85% reduction compared with unmodified fibers and indicating the best wear resistance. This result suggests that enzymatic polyphenol crosslinking contributed to improved surface protection and bulk integrity of the fibers. In contrast, NaOH-modified fibers exhibited an increased abrasion loss of 15.12%, indicating that alkali etching may weaken the lignocellulosic skeleton and make the fibers more susceptible to breakage under mechanical action.

4.2 Microstructural and chemical characterization of corn stover fibers

4.2.1 SEM Characterization of Corn Stover Fibers

Figure 9 SEM characterization of four corn stover fibers.

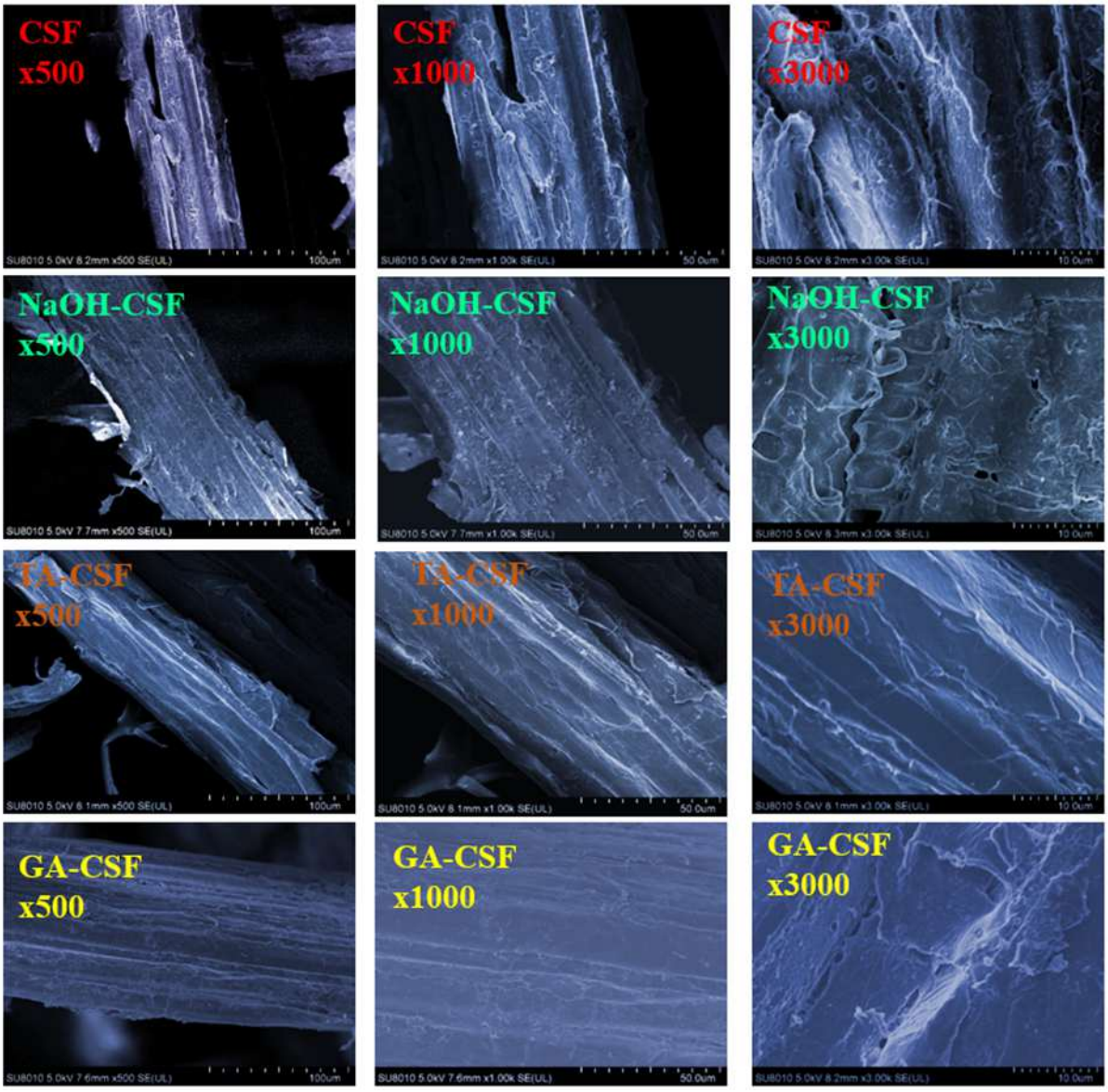


Fig. 9. SEM images of different corn stover fibers

As shown in Figure 9, fiber surface morphology is strongly influenced by the modification method. Unmodified CSF retains a smooth, continuous strip-like structure with shallow grooves, limiting mechanical interlocking with asphalt. NaOH-CSF shows pronounced etching, holes, cracks, and partial detachment, indicating increased roughness but compromised structural

integrity. TA-CSF exhibits uniform fine protrusions and thin layer attachments while maintaining fiber integrity, suggesting mild surface enhancement. GA-CSF presents a dense, continuous micro–nano granular network with no severe erosion, forming a stable functional layer that increases surface area, adhesion sites, wear resistance, and fiber–asphalt interfacial bonding.

4.2.2 FTIR analysis

Figure 10 FTIR characterization of four corn straw fibers.

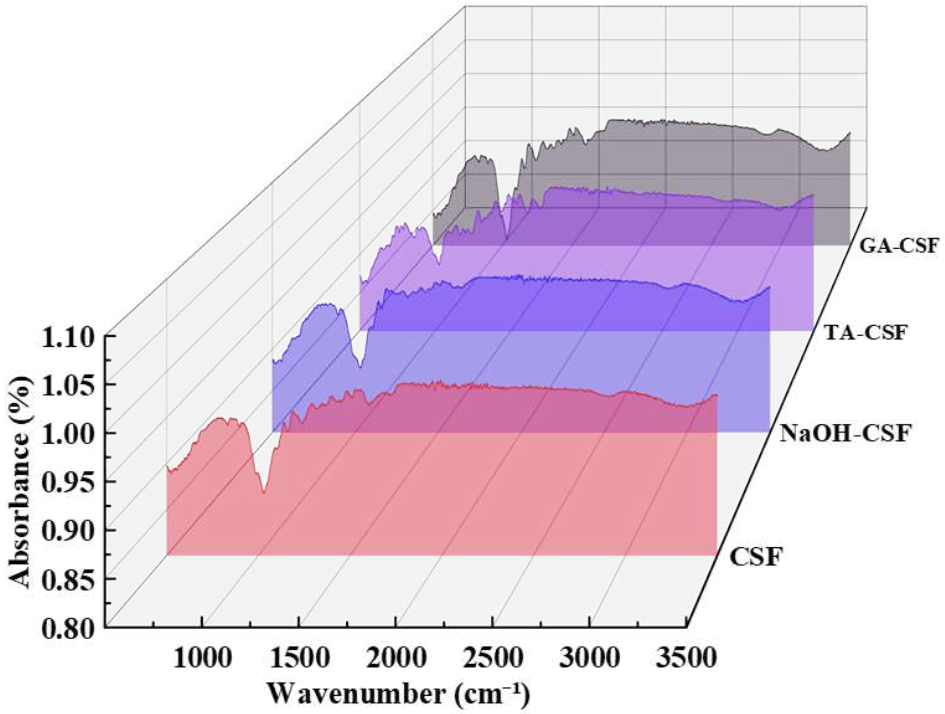


Fig. 10. FTIR spectra of four types of corn stover fibers

As shown in Figure 10, different modification methods significantly affected the surface functional groups of corn stover fibers. CSF exhibited an –OH stretching vibration peak near 3336 cm^{-1}, a C–H stretching vibration peak near 2912 cm^{-1}, and characteristic absorption peaks corresponding to C=O, aromatic skeleton, C–O–C, and C–O near 1720, 1603, 1159, and 1029 cm^{-1}, respectively, indicating that cellulose, hemicellulose, and lignin structures were retained in

the unmodified fibers. In NaOH-CSF, the C=O peak near 1720 cm^{-1} and the aromatic structure peak near 1600 cm^{-1} were significantly weakened, with peak areas decreasing to 0.2507 and 0.3225, respectively, suggesting the removal of part of the hemicellulose, lignin, and surface impurities by alkali treatment. TA-CSF showed enhanced absorption at 1607 cm^{-1} and 1167 cm^{-1}, with peak areas of 0.5515 and 0.6013, respectively, indicating the successful introduction of polyphenolic aromatic structures and C–O bonds from tannic acid onto the fiber surface. GA-CSF exhibited significantly enhanced –OH, aromatic, and C–O peaks, with the –OH peak area reaching 8.9038 at 3338 cm^{-1} and the C–O peak area reaching 3.3941 at 1031 cm^{-1}. This indicates that the laccase–gallic acid system formed a functionalized layer rich in hydroxyl and aromatic structures on the fiber surface. Overall, NaOH treatment mainly resulted in component removal, whereas TA-CSF and GA-CSF reflected the introduction of polyphenolic functional groups. Among them, GA-CSF showed the most pronounced enhancement in functional groups, providing a chemical basis for its improved interfacial bonding potential and toughening behavior.

4.2.3 XPS analysis

In this study, a Thermo Scientific K-Alpha+ X-ray photoelectron spectrometer was used to analyze the surface elemental composition and chemical bonding states of CSF, NaOH-CSF, TA-CSF, and GA-CSF. The differences in surface elemental composition among the four types of fibers are shown in Figure 11.

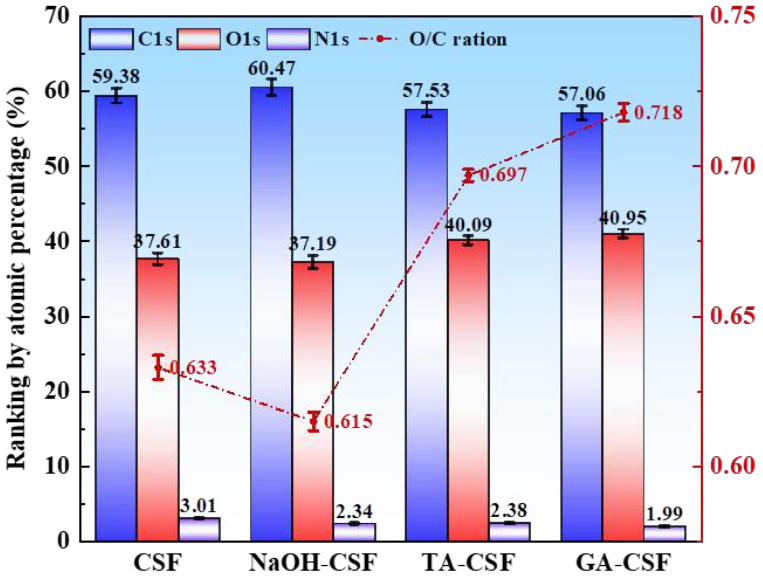


Fig. 11. Differences in surface elemental composition among the four fibers

As shown in Figure 11, the surface elemental composition of corn straw fibers is significantly influenced by the modification method. The C1s content follows the order NaOH-CSF > CSF > TA-CSF > GA-CSF, with NaOH-CSF exhibiting the highest C1s content at 60.47%, while the O/C ratio decreases to 0.615. This suggests that NaOH treatment removes part of the hemicellulose, pectin, and oxygen-containing functional groups, leading to a relative enrichment of carbon on the fiber surface. The associated surface etching and structural damage are consistent with the SEM observations. Conversely, the C1s content of TA-CSF and GA-CSF decreases to 57.53% and 57.06%, respectively, while the O1s content increases to 40.09% and 40.95%, indicating that polyphenol modification introduces additional oxygen-containing functional groups on the fiber surface. Overall, GA-CSF possesses the highest proportion of surface oxygen-containing groups and polar characteristics, which is beneficial for enhancing interfacial interactions with the polar components of asphalt.

4.3 Rheological analysis of asphalt binders

4.3.1 Temperature scanning

Figure 12 shows the temperature sweep results of the four asphalt binders.

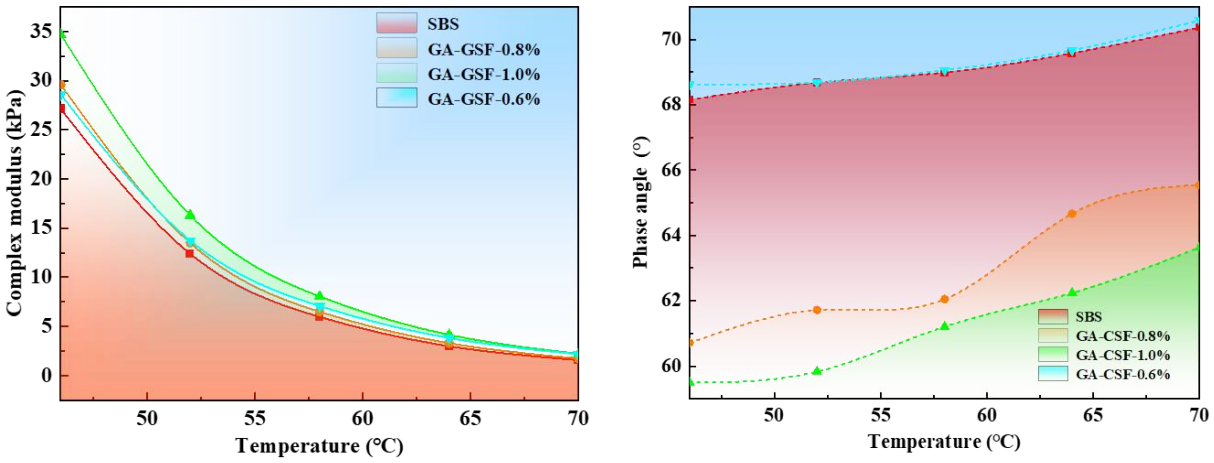


Fig. 12. Temperature sweep curves of the four asphalt binders

As shown in Figure 12, the complex shear modulus of all asphalt samples decreases markedly as the temperature rises from 46°C to 70°C, indicating a reduction in shear deformation resistance at elevated temperatures. Compared with SBS asphalt, GA-CSF-modified binders generally exhibit higher complex shear modulus, with GA-CSF-1.0% showing the most pronounced improvement, increasing by 27.90%, 31.57%, 34.90%, 38.92%, and 40.81% at 46, 52, 58, 64, and 70°C, respectively. This demonstrates that higher GA-CSF content effectively enhances the high-temperature stiffness of asphalt. Phase angle analysis reveals that GA-CSF significantly influences the viscoelastic balance of the binder. While the phase angle of SBS increases from 68.16° to 70.37° with rising temperature, indicating dominant viscous behavior, GA-CSF-0.6% shows a phase angle comparable to SBS. In contrast, GA-CSF-0.8% and GA-CSF-1.0% exhibit considerably lower phase angles, with average reductions of 6.22° and 7.87°, respectively. GA-CSF-1.0% consistently shows the lowest phase angles across all temperatures, reflecting the highest elastic contribution and the strongest restriction on viscous flow. However, excessive fiber

content may increase mixing viscosity and hinder dispersion; therefore, optimal fiber content should be determined considering practical mixing and storage stability in future studies.

4.3.2 Frequency sweep analysis

To evaluate the effects of different fiber contents on the viscoelastic properties of asphalt binders, frequency sweep tests were conducted over the same frequency range at different temperatures to construct master curves of complex shear modulus and phase angle. The master curves of complex shear modulus and phase angle for different modified asphalt binders are shown in Figure 13.

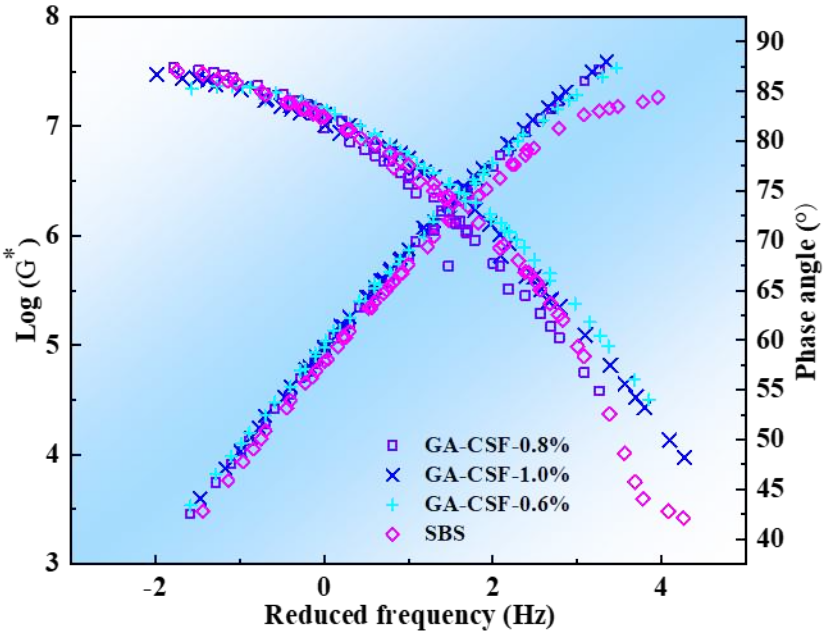


Fig. 13. Master curves of complex shear modulus and phase angle for four asphalt binders

As shown in Figure 13, all four asphalt binders exhibited clear frequency dependence. The complex shear modulus increased with reduced frequency, indicating that high-frequency loading restricts asphalt molecular motion and shifts the system from viscous-dominated to elastic-dominated behavior, enhancing shear resistance. Compared to SBS asphalt, GA-CSF-modified asphalt showed higher complex shear modulus across the frequency range, suggesting improved

stiffness through fiber network support, interfacial adhesion, and spatial constraint. In the medium- and high-frequency regions, GA-CSF-modified asphalt generally had lower phase angles than SBS, reflecting enhanced elasticity and reduced irreversible deformation. Notably, GA-CSF-0.8% and GA-CSF-1.0% showed the greatest phase angle reduction at high frequencies, indicating stronger molecular motion restriction and improved high-temperature stability and rutting resistance.

4.3.3 Multiple stress creep recovery (MSCR) test

Figure 14 shows the non-recoverable creep compliance and creep recovery of the four asphalt binders.

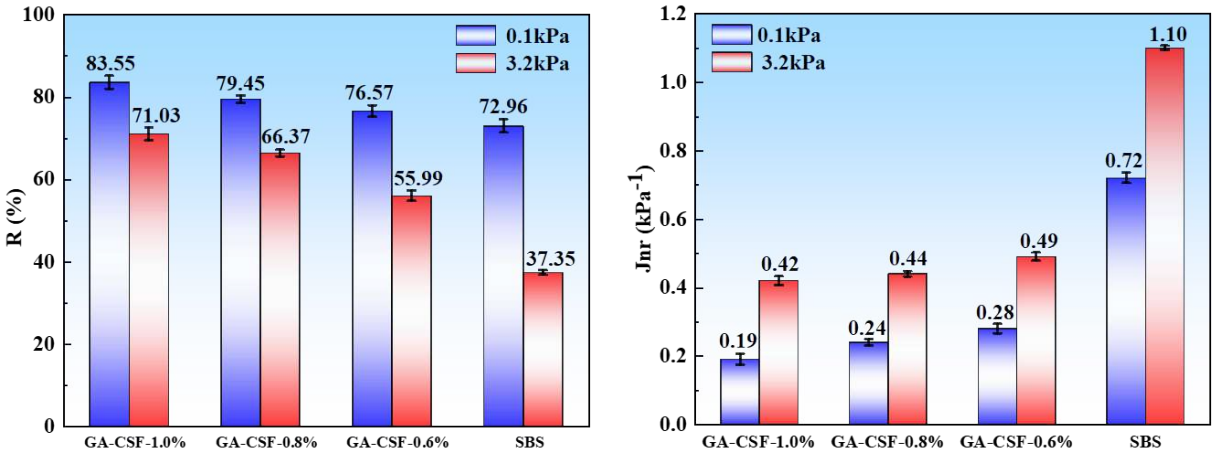


Fig. 14. Non-recoverable creep compliance and creep recovery of the four asphalt binders

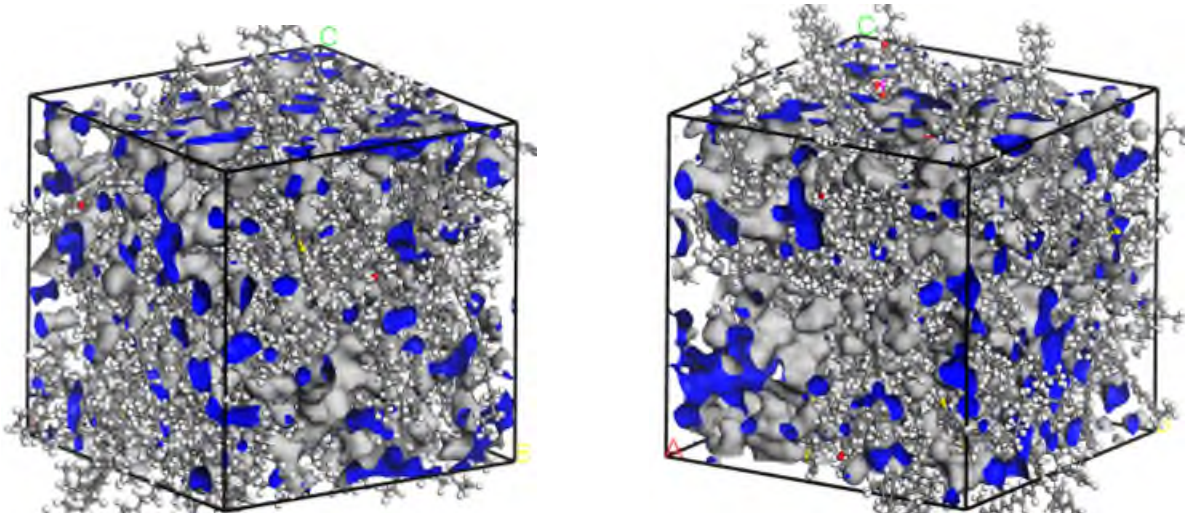
As can be seen from Figure 14, when the stress level increases from 0.1 kPa to 3.2 kPa, the creep recovery rate (R) of the four asphalts decreases, while the unrecoverable creep compliance increases. This indicates that under high stress conditions, the elastic recovery ability of asphalt binder weakens, and the unrecoverable deformation increases. At 0.1 kPa, the creep recovery rate of SBS modified asphalt is 72.96%, and GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% show improvements of 4.95%, 8.90%, and 14.52% respectively compared to SBS. At 3.2 kPa, the

modifying effect of GA-CSF is more pronounced, with GA-CSF-1.0% exhibiting the highest recovery ability. The results further show that GA-CSF can significantly reduce the unrecoverable deformation of asphalt. At 0.1 kPa, the unrecoverable creep compliance of GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% decreases by 61.11%, 66.67%, and 73.61% respectively compared to SBS, and at 3.2 kPa, it decreases by 55.45%, 60.00%, and 61.82% respectively. Overall, the addition of GA-CSF improves the elastic recovery ability of asphalt binder and reduces the sensitivity to permanent deformation, which is mainly attributed to the constraining effect of GA-CSF on the molecular movement of asphalt and the inhibitory effect of the fiber network on shear deformation.

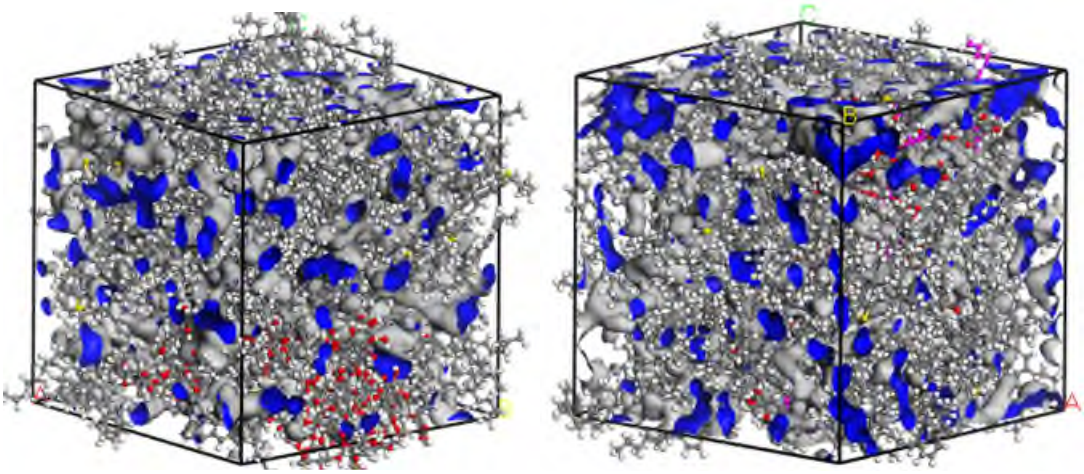
4.4 Molecular simulation analysis

4.4.1 FFV

In this study, the Connolly surface method was used to calculate the free volume of different asphalt models. The three-dimensional Connolly surface views of the four asphalt systems are shown in Figure 15.



(a) SBS (b) GA-CSF-0.6%



(c) GA-CSF-0.8% (d) GA-CSF-1.0%

Fig. 15. Three dimensional Connolly views of four types of asphalt

Figure 15 shows the three-dimensional Connolly surface distributions of the four asphalt systems, where the gray regions represent the van der Waals surface and the blue regions correspond to the Connolly surface of the free volume. Compared with the SBS system, the continuity and distribution range of the blue regions decreased after GA-CSF incorporation, indicating that GA-CSF reduced the internal free space of the asphalt system and promoted more compact molecular packing. With increasing GA-CSF content, the free volume in the system was further compressed, suggesting that the cellulose backbone and gallic acid-functionalized groups enhanced intermolecular interactions in asphalt and restricted molecular segment motion.

4.4.2 Cohesive energy density

Figure 17 shows the cohesive energy density and solubility parameter of the four asphalt binders.

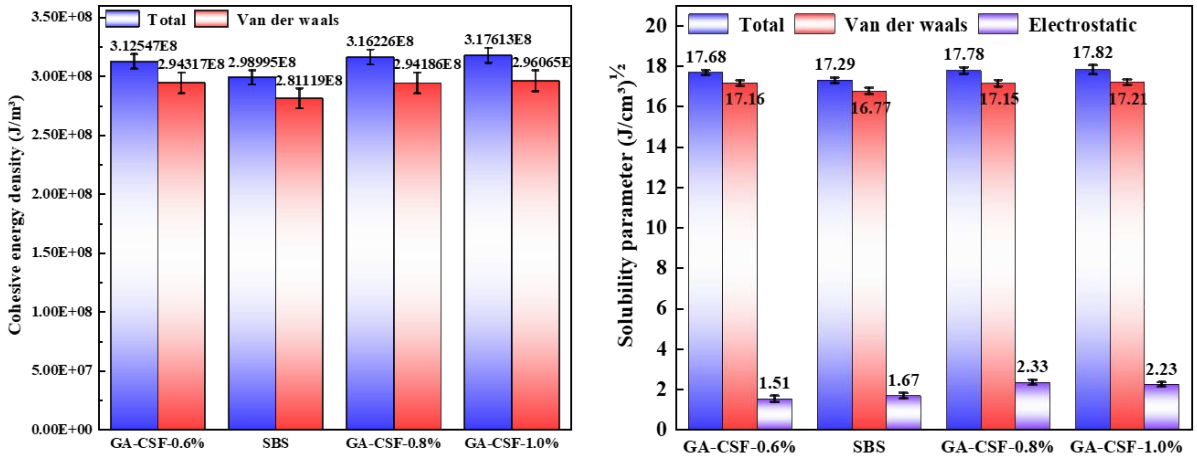


Fig. 16. Cohesive energy density and solubility parameter of the four asphalt binders

Figure 16 shows the cohesive energy density and solubility parameter of the four asphalt systems. The incorporation of GA-CSF increased the total cohesive energy density of the asphalt system. Compared with SBS-modified asphalt, the total cohesive energy density of GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% increased by 4.68%, 5.69%, and 6.35%, respectively. This indicates that GA-CSF strengthened the intermolecular attraction within the asphalt system, thereby stabilizing the molecular aggregation structure. Among the different interaction components, van der Waals interactions made the dominant contribution to cohesive energy density. The van der Waals cohesive energy densities of GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% were 4.63%, 4.63%, and 5.34% higher than that of SBS, respectively, suggesting that GA-CSF primarily improved the intermolecular binding state of asphalt by enhancing non-bonded interactions. The solubility parameter results showed a similar trend. The electrostatic solubility parameters of asphalt systems with higher GA-CSF contents increased markedly, with GA-CSF-0.8% and GA-CSF-1.0% showing increases of 39.52% and 33.53%, respectively, compared with SBS. This may be attributed to the polar groups in GA-CSF, such as hydroxyl, carboxyl, and ester groups, which enhanced the interaction between GA-CSF and the polar components of asphalt.

Overall, the addition of GA-CSF improved the molecular cohesive strength and thermodynamic compatibility of the asphalt system, facilitating the formation of a more stable modified asphalt structure.

4.4.3 Elastic properties

In this study, the Mechanical Properties function of the Forcite module was employed to calculate the elastic properties of four equilibrated asphalt molecular models, as shown in Figure 17. The results indicate that GA-CSF significantly enhances the Young's modulus, bulk modulus, and shear modulus of the asphalt system, demonstrating its ability to improve the overall stiffness and deformation resistance. Compared to SBS asphalt, the Young's modulus of GA-CSF-0.6%, GA-CSF-0.8%, and GA-CSF-1.0% increased from 5.72 GPa to 5.93, 6.27, and 6.39 GPa, corresponding to increases of 3.67%, 9.62%, and 11.71%, respectively, suggesting a restraining effect on tensile deformation. The bulk modulus rose from 4.18 GPa to 4.63, 4.87, and 4.91 GPa, representing increases of 10.77%, 16.51%, and 17.46%, indicating enhanced resistance to volumetric compression. The shear modulus increased from 2.25 GPa to 2.31, 2.49, and 2.57 GPa, corresponding to increases of 2.67%, 10.67%, and 14.22%, suggesting improved shear stability by restricting relative slippage between asphalt molecules. Overall, all three moduli exhibit an upward trend with increasing GA-CSF content. Beyond 0.8%, the growth in Young's and bulk moduli slows, implying that the system's rigidity approaches a plateau at higher fiber concentrations. These findings indicate that GA-CSF reinforces asphalt at the molecular scale through its rigid fiber skeleton and surface polar groups, enhancing the binder's mechanical response.

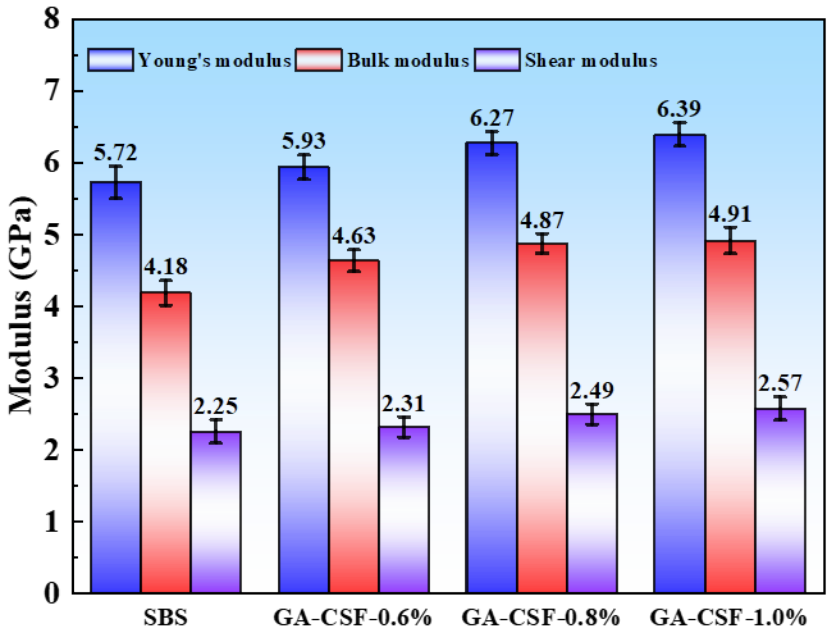


Fig. 17. Simulated mechanical properties of the four asphalt binders

5. Conclusion

This study investigates the high-value utilization of corn straw fibers in asphalt binders, employing NaOH, tannic acid, and gallic acid–laccase for fiber modification. By integrating fiber performance evaluation, microscopic characterization, rheological testing, and molecular dynamics simulations, the effects of different modification methods were systematically compared. The results underscore the superior toughening effect of gallic acid–laccase synergistic modification on corn straw fibers and reveal the mechanisms by which the modified fibers enhance the high-temperature viscoelastic response and molecular structural stability of asphalt. The key conclusions are summarized as follows.

1) Compared to NaOH and tannic acid modifications, the gallic acid–laccase synergistic treatment (GA-CSF) exhibits the most pronounced improvement in the mechanical properties of corn straw fibers. The tensile strength, elastic modulus, and elongation at break increase by

127.51%, 154.00%, and 46.18%, respectively, relative to unmodified fibers, demonstrating that this modification simultaneously enhances fiber strength, stiffness, and toughness.

2) GA-CSF shows superior resistance to mechanical damage, with an attrition loss rate of only 4.25%, representing a 56.85% reduction compared to unmodified fibers. In contrast, NaOH treatment increases fiber attrition, suggesting that excessive alkali etching may disrupt the continuity of the lignocellulose skeleton, compromising structural stability.

3) SEM, FTIR, and XPS analyses indicate that gallic acid–laccase treatment forms a polyphenol-functionalized layer rich in oxygen-containing groups on the fiber surface. The O/C ratio of GA-CSF is 13.43% higher than that of unmodified fibers, reflecting enhanced surface polarity and chemical activity, which favor improved interaction with the polar components of asphalt.

4) Rheological testing shows that GA-CSF significantly enhances the high-temperature viscoelastic properties and resistance to permanent deformation of SBS-modified asphalt. Across 46–70 °C, the complex shear modulus of GA-CSF-1.0% increases by 27.90%–40.81% relative to SBS asphalt. At a stress level of 3.2 kPa, the creep recovery rate increases by 90.17%, while the unrecoverable creep compliance decreases by 61.82%.

5) Molecular dynamics simulations reveal that incorporating GA-CSF increases the cohesive energy density and elastic modulus of the asphalt system and reduces the free volume fraction from 0.21 to 0.16. These results indicate that the modified fibers strengthen intermolecular interactions and restrict molecular segment mobility, providing a molecular-level explanation for the enhanced high-temperature deformation resistance.

Funding Sources

This research was funded by the National Natural Science Foundation of China (52408499).

Data availability

The datasets used and analyzed during the current study are available from the corresponding author on reasonable request.

REFERENCES

[1] R. Zhang, P. Hu, Y. Zhong, L. Wen, J. Ji, L. Liang, Research progress on evaluation and prediction of degradation in service performance for asphalt pavement, J. Traffic Transp. Eng. Engl. Ed. 12 (2025) 1011–1039.

[2] J. Zhang, X.D. Zhang, Review of studies on asphalt pavement fatigue damage theory, J. Highway Transp. Res. Dev. 37 (2020) 1–11.

[3] H. Di Benedetto, F. Olard, C. Sauzéat, B. Delaporte, Linear viscoelastic behaviour of bituminous materials: From binders to mixes, Road Mater. Pavement Des. 5 (2004) 163–202.

[4] S.M. Rangappa, S. Siengchin, J. Parameswaranpillai, M. Jawaid, T. Ozbakkaloglu, Lignocellulosic fiber reinforced composites: Progress, performance, properties, applications, and future perspectives, Polym. Compos. 43 (2022) 645–691.

[5] S.K. Ramamoorthy, M. Skrifvars, A. Persson, A review of natural fibers used in biocomposites: Plant, animal and regenerated cellulose fibers, Polym. Rev. 55 (2015) 107–162.

[6] P.K. Akarsh, S. Marathe, H.K. Sapal, N.A. Krishna, Synergistic effects of natural fibres and agro-waste ash on the engineering and sustainability of stone-matrix asphalt mixes, Int. J. Transp. Sci. Technol. (2025).

[7] J. Zhang, W. Huang, Y. Zhang, Q. Lv, C. Yan, Evaluating four typical fibers used for OGFC mixture modification regarding drainage, raveling, rutting and fatigue resistance, Constr. Build. Mater. 253 (2020) 119131.

[8] Y. Zhang, H. Wang, X. Sun, Y. Wang, Z. Liu, Separation and characterization of biomass components (cellulose, hemicellulose, and lignin) from corn stalk, BioResources 16 (2021) 7205–7219.

[9] C. Lou, Y. Zhou, A. Yan, Y. Liu, Extraction cellulose from corn-stalk taking advantage of pretreatment technology with immobilized enzyme, RSC Adv. 12 (2022) 1208–1215.

[10] T.R. Caixeta Silva, L.A. Silva de Aquino, L.C. Mesquita, M.G. Marques, A.R.G. de Azevedo, M.T. Marvila, Innovative cementitious composites produced with corn straw fiber: Effect of the alkaline treatments, Appl. Sci. 14 (2024) 11117.

[11] S.A. Kavitha, R.K. Priya, K.P. Arunachalam, S. Avudaiappan, E.I. Saavedra Flores, D. Blanco, Experimental investigation on strengthening of Zea mays root fibres for biodegradable composite materials using potassium permanganate treatment, Sci. Rep. 14 (2024) 12754.
[12] Z. Li, K. Li, W. Chen, W. Liu, Y. Yin, P. Cong, Investigation on the characteristics and effect of plant fibers on the properties of asphalt binders, Constr. Build. Mater. 338 (2022) 127652.
[13] C. Hu, J. Feng, N. Zhou, J. Zhu, S. Zhang, Hydrochar from corn stalk used as bio-asphalt modifier: High-temperature performance improvement, Environ. Res. 193 (2021) 110157.
[14] X. Chen, P. Cheng, Y. Li, H. Wang, C. Zheng, R. Ma, T. Nian, M. Wang, Application of corn straw biochar in pavement asphalt: Suppression of hazardous fume emissions and maintenance of rheological properties, Constr. Build. Mater. (2025) 141635.
[15] S. Chang, R. Guo, J. Geng, X. Yin, Y. Yang, F. Tan, From performance enhancement to mechanism elucidation: A comprehensive review of straw fiber-modified asphalt mixtures, Results Eng. (2026) 109290.
[16] T. Nian, P. Li, J. Ge, J. Song, M. Wang, Green environmental protection and sustainable utilization of straw: Investigation for molecular dynamics of highland barley straw fiber to enhancing road performance of asphalt, J. Clean. Prod. 452 (2024) 141940.
[17] Y. Liu, W. Jia, Q. Yang, Y. Ma, J. Tong, A. Shi, S. Siengchin, Physical, mechanical, and environmental properties of corn stalk fiber reinforced braking composites prepared by wet granulation, J. Nat. Fibers 19 (2022) 14515–14524.
[18] S. Devi, P.K. Poonia, V. Kumar, A. Tiwari, R.K. Meena, U. Kumar, M.K. Al-Sadoon, Characterization of natural fiber extracted from corn (Zea mays L.) stalk waste for sustainable development, Sustainability 14 (2022) 16605.
[19] Z.H. Kamaruddin, R. Jumaidin, R.A. Ilyas, M.Z. Selamat, R.H. Alamjuri, F.A.M. Yusof, Influence of alkali treatment on the mechanical, thermal, water absorption, and biodegradation properties of Cymbopogan citratus fiber-reinforced, thermoplastic cassava starch–palm wax composites, Polymers 14 (2022) 2769.
[20] S.S. Shifa, M.M.H. Kanok, M.S. Haque, Exploring water absorption behaviors on mechanical properties of wheat straw-glass fiber hybrid composites for sustainable engineering solutions, Hybrid Adv. 7 (2024) 100297.

[21] T.R. Caixeta Silva, L.A. Silva de Aquino, L.C. Mesquita, M.G. Marques, A.R.G. de Azevedo, M.T. Marvila, Innovative cementitious composites produced with corn straw fiber: Effect of the alkaline treatments, Appl. Sci. 14 (2024) 11117.

[22] M. Stepczyńska, P. Rytlewski, K. Moraczewski, A. Pawłowska, T. Karasiewicz, Novel biocomposite of starch and flax fiber modified with tannic acid with biocidal properties, Polymers 16 (2024) 1108.

[23] Z. Li, C. Yan, W. Xu, Y. Shang, Q. Wu, S. Mehmood, F. Wang, C. Cheng, Q. Liu, Z. Shao, Hydrophobic modification of cellulose nanofibers by gallic acid and the application in pressure sensing, Int. J. Biol. Macromol. 261 (2024) 129770.

[24] W. Guo, G. Cao, K. Gu, Y. Li, Y. Yuan, W. Tian, Oil shale waste as asphalt fine aggregate: Multiscale insights into pore structure, rheology and molecular mechanisms, Int. J. Pavement Eng. 27 (2026) 2681688.

[25] W. Guo, K. Gu, Y. Li, W. Tian, S. Wang, H. Xu, W. Chen, Enhancement of asphalt properties through SBS and CNT synergism: A comprehensive multi-scale analysis, Constr. Build. Mater. 505 (2025) 144747.

[26] M.M. Xiao, L. Fan, Ultraviolet aging mechanism of asphalt molecular based on microscopic simulation, Constr. Build. Mater. 319 (2022) 126157.

[27] D. Hu, X. Gu, Q. Dong, L. Lyu, B. Cui, J. Pei, Investigating the bio-rejuvenator effects on aged asphalt through exploring molecular evolution and chemical transformation of asphalt components during oxidative aging and regeneration, J. Clean. Prod. 329 (2021) 129711.

[28] H. Yao, Q. Dai, Z. You, Molecular dynamics simulation of physicochemical properties of the asphalt model, Fuel 164 (2016) 83–93.

[29] K. Shi, F. Ma, J. Liu, Z. Fu, R. Song, D. Yuan, C. Li, Rejuvenation effect of aged SBS-modified asphalt utilizing molecule analysis, J. Clean. Prod. 405 (2023) 136964.

[30] H. Ding, H. Wang, X. Qu, A. Varveri, J. Gao, Z. You, Towards an understanding of diffusion mechanism of bio-rejuvenators in aged asphalt binder through molecular dynamics simulation, J. Clean. Prod. 299 (2021) 126927.