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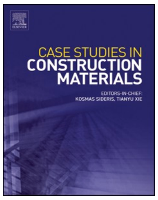

Performance evaluation of surface treatment waste glass as aggregate in asphalt mixture

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ABSTRACT

Surface modification is a crucial strategy for enhancing the pavement performance of waste glass hot mix asphalt (HMA) mixtures. This study conducts a comparative analysis of various surface modification techniques to pinpoint the key factors that influence the interfacial interaction between waste glass and asphalt. To assess the performance of asphalt mixtures containing surface-treated waste glass, the study employed methods such as Energy Dispersive X-ray Spectroscopy (EDS), boiling water tests, and contact angle measurements, which helped to quantify surface morphology and the effectiveness of different treatments. The evaluation also included assessments of water stability, dynamic stability, small beam bending, and anti-skid properties. Findings revealed that applying a silane coupling agent to the waste glass significantly enhanced adhesion with asphalt, resulting in markedly improved pavement performance. The study identified that incorporating surface-treated waste glass at an optimal proportion of 6 % within the asphalt mixture led to a 42.3 % increase in dynamic stability and a 38.6 % enhancement in anti-skid performance compared to standard mixtures. This research offers a comprehensive evaluation framework for surface modification techniques of waste glass, underscoring its potential to considerably improve road performance.

1. Introduction

Asphalt pavements, renowned for its excellent driving comfort and safety, are extensively utilized in the construction of various high-grade road surfaces [1–5]. However, the substantial use of road aggregates, a primary component of asphalt pavements, has led to the excessive depletion of natural stone materials, thereby causing a series of issues such as the exhaustion of natural resources and environmental degradation [6–9]. To mitigate these challenges, there is an urgent need to explore effective alternative materials to alleviate the rapid consumption of resources. Glass artifacts, which occupy a significant market globally, unfortunately, have a high scrap rate of 79 % in the recycling process. Traditional disposal methods for waste glass products, such as landfilling and incineration, do not favor the recycling of energy and resources [10–14]. Existing studies have demonstrated that the incorporation of waste glass into asphalt mixtures for reuse can significantly reduce the consumption of natural stone materials while simultaneously alleviating

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environmental impacts [15–20].

Numerous researchers have conducted research on the recycling and reuse of solid waste in asphalt mixtures [21]. Since the 1960s, developed countries such as Europe and America have pioneered studies on asphalt mixtures incorporating waste glass [22,23]. Du et al. [24] emphasized that, as an alternative to limestone filler, waste glass microspheres can significantly reduce the thermal conductivity of asphalt mixtures. He concluded that waste glass is considered a potential enhancer for the permanent anti-deformation properties of road materials. Simone et al. [25] confirmed through the MSCR test that waste glass power as filler can improve the deformation resistance of asphalt pavements. However, waste glass surfaces are usually smooth and lack sufficient roughness. Additionally, glass has weak adhesive strength with asphalt at its interface, which is detrimental to the durability of the mixture in road applications. To overcome the inert surface properties of waste glass and enhance its adhesion to asphalt, lots of researchers have made numerous efforts. For instance, Sanij et al. [26] found that ZycothermTM improved the adhesion between asphalt and waste glass, reducing the moisture sensitivity of the asphalt mixture. Behbahani et al. [27] evaluated the potential of nanotechnology zycosoil as an antistripping agent to enhance the creep recovery and mechanical properties of waste glass asphalt mixtures. Min et al. [28] discovered that, compared to matrix asphalt, glass aggregates in epoxy asphalt mixtures exhibit better adhesive performance even without the use of antistripping agents. Compared to research on optimizing such antistripping agents and high-viscosity, high-elasticity asphalt formulations, there has been less focus on modifying the surface of waste glass. Zhou et al. [29] improved the viscosity between waste glass aggregate and asphalt by treating the surface of waste glass with a Nano Fe_2O_3 suspension [29]. The above analysis indicates that the inert nature of waste glass surfaces is influenced by factors such as functional group composition and microstructure.

Existing research indicates that crushed waste glass possesses characteristics such as abrasion resistance, anti-slip properties, reflectivity, and strong permeability [30–32]. When glass aggregates replace part of the aggregate in the asphalt mixtures for surface layers, it can improve the road's resistance to wear, slippage, water penetration, and rutting to a certain extent [33–37]. The weak interfacial bonding strength between glass aggregates and asphalt prevents the formation of a robust framework in asphalt mixtures containing glass aggregates. In other words, the asphalt film coating the surface of the aggregate is prone to peeling due to moisture, leading to potholes, loosening, and other water-related damage [3,9,38–40]. To address the issue of insufficient adhesion between waste glass aggregate and asphalt, current solutions primarily rely on using antistripping agents or asphalt with better binding properties. Sanij et al. [41] incorporated Zycotherm™ as an antistripping agent into warm mix asphalt, finding it effectively improved the adhesion between asphalt and glass surfaces. H. Behbahani et al. [21] used zycosoil as an antistripping agent to modify asphalt, and found that when the glass asphalt mixture contained 10 % glass and 4 % zycosoil, the mechanical properties of the mixture were effectively improved. Min et al. [33] replaced traditional asphalt with epoxy asphalt and discovered that even without an antistripping agent, the glass aggregate in the epoxy asphalt mixture exhibited good adhesive performance. Zhou et al. [3] tested the road performance of waste glass as part of the aggregate in porous asphalt mixtures, finding that it exhibits good technical performance in terms of high-temperature stability, low-temperature stability, and water stability. Concurrently, the use of glass significantly enhances road surface brightness, contributing to improved visibility in road environments.

These factors are also influenced by physical methods and chemical modifications. Furthermore, previous studies have limited focus on the differentiated analysis of physical methods and chemical modifications. Therefore, it is necessary to evaluate the different surface treatment processes on the adhesive performance between waste glass and asphalt and select the better surface treatment process and then evaluate surface treated waste glass asphalt mixture performance. An optical imaging system and Energy Dispersive X-ray Spectrometer (EDS) tests were utilized to observe the microstructure of untreated and treated waste glass, followed by employing boiling water methods and contact angle tests to characterize the adhesive properties of untreated and treated waste glass. This enabled us to quantitatively compare the effects of abrasion and silane coupling agent coating on the inertness of the waste glass surface, and to establish an optimal evaluation mechanism for the surface modification processes of waste glass. Finally, based on water stability tests, dynamic stability tests, small beam bending tests, and anti-skid tests, the optimal dosage of surface treated waste glass in the asphalt mixture was recommended.

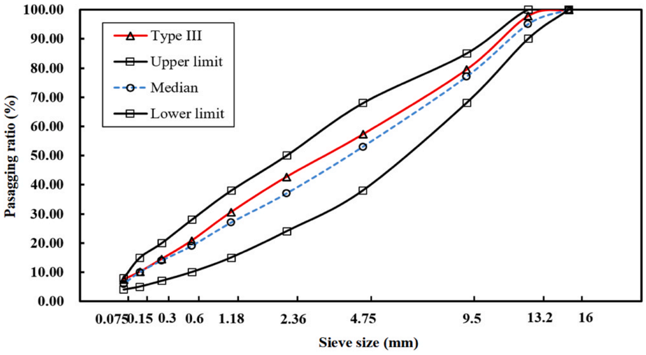


Fig. 1. Aggregate gradation curve used in this study.

2. Materials and methods

2.1. Materials

The waste glass (size: 2.36–4.75 m) used in this study was provided by the Chongqing Sanitation Center. The Styrene-Butadiene-Styrene (SBS)-modified asphalt, coarse aggregate (> 4.75 mm) consisting of basalt, and fine aggregate (< 4.75 mm) of limestone, all conform to the JTG E20–2011 standards for highway engineering asphalt and asphalt mixtures [42]. The Marshall test was used to determine the optimum asphalt content (OAC). The OAC for asphalt mixture with 0 % waste glass, asphalt mixture with 6 % waste glass, asphalt mixture with 8 % waste glass, and asphalt mixture with 10 % waste glass are 4.8 %, 4.9 %, 4.9 %, 5.1 %, respectively. The dense aggregate gradation curve used in this study is displayed in Fig. 1.

The detailed physical properties of raw materials are shown in Table 1. The 3-aminopropyltriethoxysilane (APTES also called silane coupling agents) coupling agent was purchased from Macklin Reagents Co., Ltd. The chemical reaction schematic of the silane coupling agent is shown in Fig. 2 below. The silane coupling agent is an organosilicon compound. Through chemical reactions, silane coupling agents can form connections between different materials, enhancing the affinity between inorganic materials and organic substances, or between various organic composite systems. Therefore, to address the issue of weak bonding between acidic aggregates and asphalt, which relies on physical adsorption, the surface of acidic aggregates can be modified using coupling agents. This is because the silane coupling agent interacts with inorganic acidic aggregates. The -Si-OH groups formed by the hydrolysis of the silane coupling agent bond with the inorganic material to form -Si-O-Si- bonds, exhibiting excellent adhesive properties and effectively modifying the surface of acidic aggregates. In this process, the coupling agent acts as a bridge, connecting inorganic acidic aggregates with organic asphalt, significantly improving the overall performance of asphalt mixtures containing acidic aggregates.

2.2. Research schedule

The research schedule is presented in Fig. 3. Initially, two surface treatment methods for waste glass (abrasion method and APETS treatment) were applied. Subsequently, microstructure tests, X-ray energy spectrum analysis, boiling tests on glass aggregate and asphalt, and contact angle tests were conducted to compare the performance of the two treated glass aggregates. Finally, the mix design of APETS-treated glass modified asphalt mixture was created, followed by conducting the trabecular bending test, high-temperature performance test, and anti-skid performance test to evaluate the effectiveness of APETS treatment.

2.3. Surface treated of waste glass

2.3.1. Abrasion surface of waste glass

The adhesive interaction between asphalt and aggregate at their interface largely depends on mutual mechanical interlocking and anchoring effects, and the surface texture and roughness of the aggregate also significantly impact adhesion [43]. Therefore, this study employs the Los Angeles abrasion surface treatment process aimed at increasing the surface roughness of waste glass aggregates to treat the surface of the used glass. The pretreatment of waste glass involved cleaning the surface, followed by drying. Weighing 5 kg of

Table 1
Properties of raw materials.

Materials	Project type	Conventional value	Experimental value	Reference Standard
SBS modified asphalt	Penetration (25℃, 0.1 mm)	40–60	57	ASTM D5
	Penetration Index	≥ 0	0.39	ASTM D5
	Softening point (℃)	≥70	80	ASTM D36
	Ductility (5℃, cm)	≥25	28	ASTM D113
	Dynamic viscosity (135℃, Pa·s)	≤3	2.1	ASTM D4402
	Segregation (48 h, ℃)	≤2.5	1.5	ASTM D7173
	Elastic recovery (25℃, %)	≥85	94	ASTM D6084
Change after TFOF	Change in mass (%)	−1–1	−0.085	ASTM D6
	Penetration ratio (%)	≥65	75	ASTM D5
	Ductility (5℃, cm)	≥15	16.1	ASTM D113
Coarse aggregate	Crushing value (%)		10.7	ASTM C131
	Los Angeles wear value (%)		6.0	ASTM C131
	Needle-like content (%)		9.6	ASTM C295
	Polishing value (BPN)			ASTM C779
	Density (g/cm^3)	9.5–13.2 mm	2.802	ASTM C29
		4.75–9.5 mm	2.823	
		2.36–4.75 mm	2.690	
Fine aggregate	Sand equivalent (%)		2	ASTM D2419
	Methylene blue value (g/kg)		3	ASTM C1777
Waste glass	Density (g/cm^3)	1.18–2.36 mm	2.718	ASTM C29/ ASTM C693
	Water absorption (%)		0.23	ASTM C373

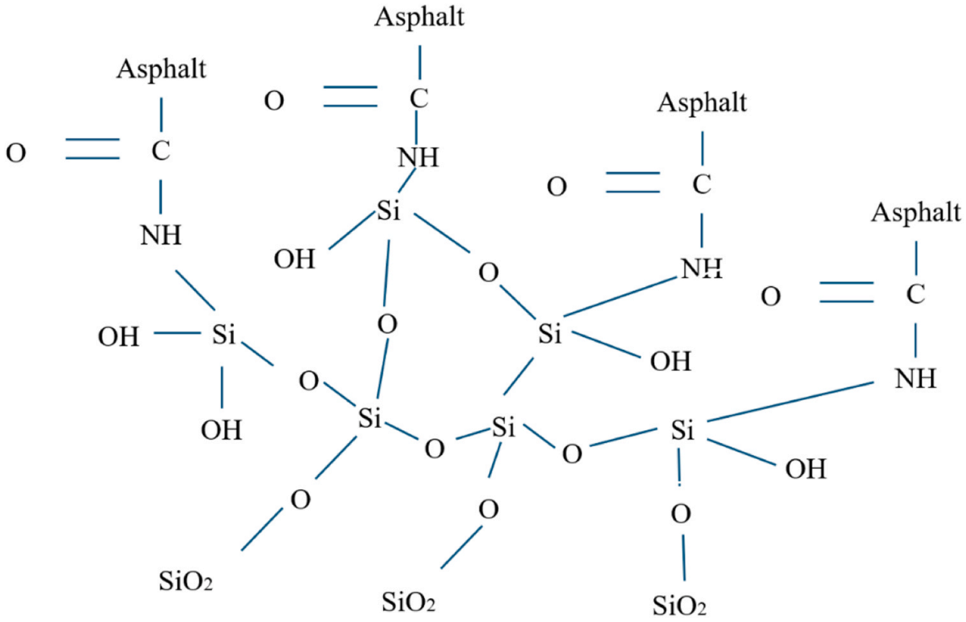


Fig. 2. Schematic diagram of chemical reaction of silane coupling agent. (adopt from [17]).

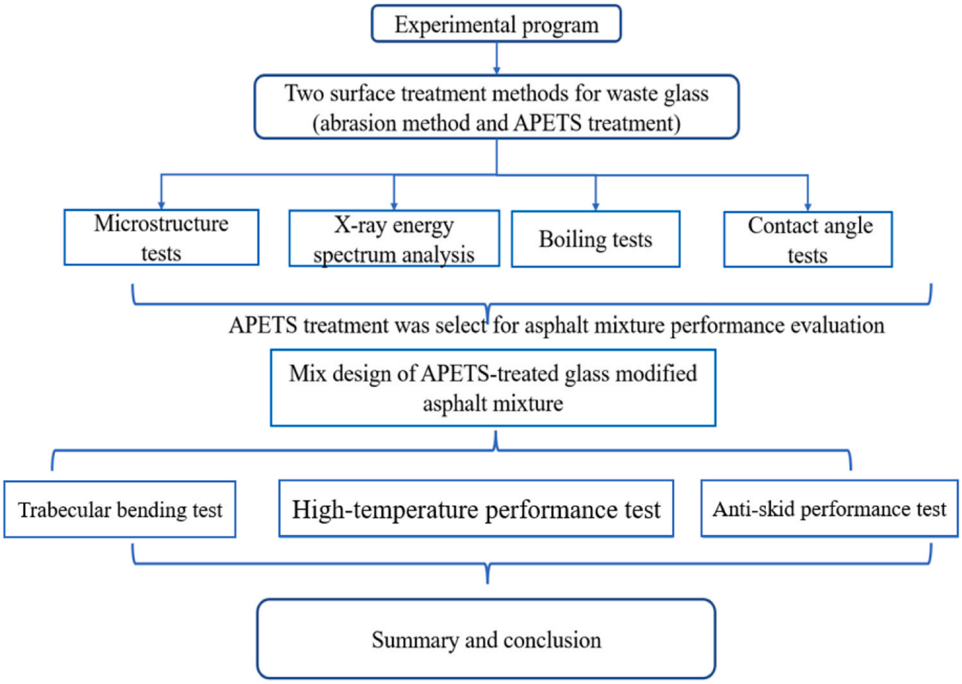


Fig. 3. Schematic diagram of experimental program in this study.

waste glass, it was subjected to three cycles of abrasion treatment using a Los Angeles abrasion machine, at 500 revolutions (A2), 1000 revolutions (A3), and 1500 revolutions (A4), compared to the untreated control materials (A1). It should be mentioned that no steel balls were used during the entire abrasion process to avoid altering the particle size of the waste glass.

2.3.2. APTES treated

Weigh an appropriate number of APTES (also named silane coupling agents), water, and anhydrous ethanol to prepare the APTES mixture solution (mass ratio of 5:45:50) and thoroughly stir for 20 minutes at room temperature. Subsequently, immerse the waste glass in the APTES mixture solution and cure at 25°C, 60°C, 120°C, and 160°C for 2 hours to obtain samples B1, B2, B3, and B4 respectively.

2.4. *Glass surface microstructure test*

The Leica microscope is equipped with research-grade high-resolution objectives, including 2.5x, 5x, 10x, 20x, 50x, and 100x six research-grade objectives. Based on the principles of geometric optical imaging, it allows observations down to the micrometer scale and can perform single-field 3D imaging and 3D measurements. The main technical parameters include a wide-field trinocular tube, beam splitting of 0–50 %-100 %, a field of view diameter of 25 mm, and a magnification range of 25–1000 times. During the experiment, glass aggregates that meet the microscope sample size requirements were selected from the glass samples after abrasion treatments of 0r, 500r, 1000r, and 1500r for subsequent observation. The prepared glass samples were placed on the microscope stage for surface observation. To more clearly study the microstructure of the glass surface after abrasion treatment, a Leica microscope was used at a magnification of 50 times and a scale of 200μm to observe the apparent morphology of the glass aggregate specimens after abrasion surface treatments of 0r, 500r, 1000r, and 1500r.

2.5. *X-ray energy spectrum analysis test*

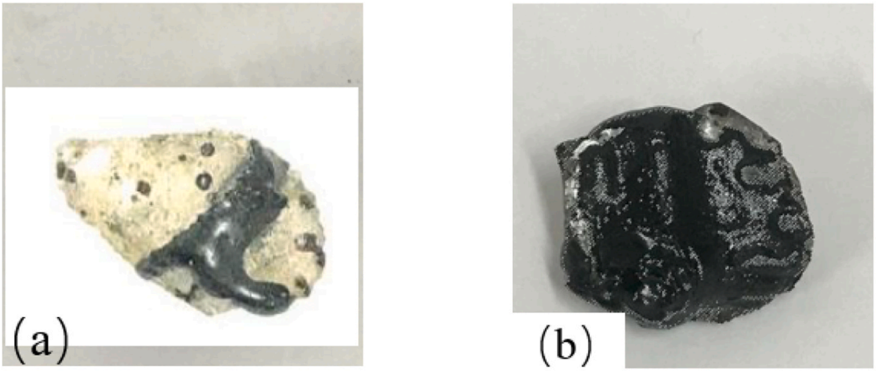
The energy dispersive X-ray spectrometer (EDS) analyzes the composition of samples by spreading spectra based on the characteristic X-ray energy differences. In this experiment, EDS is employed to perform micro-area compositional analysis of glass samples to ascertain the chemical composition changes on the surface of waste glass aggregates before and after treatment with silane coupling agents. To meet the X-ray spectrometer's sample size requirement of 3.5 mm×3.5 mm, it is necessary to select glass aggregates with surfaces that are as parallel and flat as possible from the 2.36–4.75 mm size range. Subsequently, a low-speed gemstone grinding machine is used to cut the glass specimens to conform to the experimental sample size requirements of 3.5 mm×3.5 mm. The cut glass specimens are then cleaned and placed in an oven to dry to a constant weight, followed by surface pretreatment using silane coupling agents. Since glass is a non-conductive material, to ensure precise observation and analysis by the spectrometer later, several glass samples, both untreated and treated with silane coupling agent, must coated with gold before testing.

2.6. *Boiling test glass aggregate and asphalt*

In this experiment, waste glass aggregates closely resembling cubic shapes and sized between 13.2 and 19 mm were selected. After abrasion and silane coupling agent treatments, five aggregate samples were prepared for each type of surface treatment to serve as replicates for parallel experiments. To quantitatively evaluate the adhesion performance between the surface-treated waste glass and asphalt, this study utilized image analysis technology. Based on the image, the areas of the same region and color were distinguished to extract the delaminated surface or asphalt-coated surface of the samples. Pixel area quantities were obtained using histograms. The delamination area pixel ratio or adhesion area pixel ratio, known as the delamination rate or adhesion rate, quantitatively characterizes the adhesion performance of waste glass aggregates with asphalt after, silane coupling agent surface treatment. The typical untreated glass samples and APTES treated samples are displayed in Fig. 4 below.

2.7. *Contact angle test*

The wetting ability of asphalt on the aggregate surface characterizes the strength of the interfacial bonding between them. If the asphalt can completely wet the aggregate, it indicates strong adhesive performance. The process of modified asphalt wetting the



(a). Untreated glass samples and (b). Coupling agent for glass samples.

Fig. 4. The untreated glass sample and APTES treated sample.

aggregate is typically measured using the contact angle (θ).

The Young equation describes the relationship between the surface energies of the solid, liquid, and gas phases at their interfaces:

$$\gamma_{s-l} - \gamma_{s-g} + \gamma_{l-g}\cos\theta = 0 \tag{1}$$

Where :

γ_{s-l} represents the solid/liquid interfacial tension.

γ_{s-g} represents the solid/gas interfacial tension.

γ_{l-g} represents the liquid/gas interfacial tension.

Surface tension can also be divided into components of various intermolecular forces, including the non-polar force component γ^d and the polar force componentγ^p. The adhesion work between glass aggregate and asphalt can be calculated from the modified asphalt's surface energy and the contact angle values between asphalt and glass:

$$W_a = \gamma_l(1 + \cos\theta) \tag{2}$$

Where :

W_a represents the work of adhesion (mJ/m^2)

γ_l represents the surface energy of asphalt (mJ/m^2)

θ represents the contact angle between asphalt/glass aggregate (°)

Adhesion work characterizes the adhesive performance between asphalt and glass aggregate in a dry state; the higher the value, the stronger the adhesion between the glass aggregate and asphalt, which indicates better resistance to water damage. Distilled water, glycerol, and formamide are three liquids with significantly different surface energy values, none of which are miscible with asphalt. Their surface energies are shown in Table 2 and Table 3. Therefore, this experiment uses these three liquids to conduct the modified asphalt surface energy test, using the sessile drop method to measure the contact angle values of SBS-modified asphalt with distilled water, glycerol, and formamide, thus determining the surface free energy of SBS-modified asphalt. Based on this, several relatively flat surface waste glass aggregates are selected, and glass samples treated with abrasion and silane coupling agents are prepared, followed by rapid data collection of the contact angle between the asphalt and glass aggregate. By inserting the calculated asphalt surface free energy and the measured contact angles on different glass sample surfaces into adhesion work formula 2, the adhesion work between the waste glass aggregate and asphalt after various surface treatment processes can be obtained. The contact angle goniometer is shown in Fig. 5.

2.8. *Trabecular bending test*

Under low-temperature conditions, asphalt mixtures are susceptible to temperature stresses causing shrinkage deformation. When the thermal shrinkage stress exceeds the allowable tensile stress of the mixture, shrinkage cracking occurs. The low-temperature crack resistance of asphalt mixtures containing surface treated waste glass is studied using the small beam bending test. The surface treated waste glass content is 0 %, 6 %, 8 %, and 10 %, with a particle size range of 2.36–4.75 mm. The dimensions of the small beam specimens are 250 mm×30 mm×35 mm. The testing equipment is an Materials Test Systems (MTS), with a loading speed of 50 mm/min, a span of 200 mm, and a test temperature of −10°C. Each set of specimens undergoes four parallel tests. The cross-section of the small beam specimens and the failure interface of the small beam are shown in Fig. 6.

2.9. *High temperature performance test*

Regarding channelized traffic asphalt pavements, the repetitive action of vehicular loads gradually accumulates permanent deformations, leading to rutting on the road surface. This excessive deformation weakens the strength of the surface layer and the overall pavement structure, thereby facilitating the onset of other defects. The dynamic stability (DS) in terms of cycles per millimeter is commonly used to characterize the high-temperature performance of asphalt mixtures. Therefore, a rutting tester is used to evaluate the high-temperature stability differences between conventional asphalt mixtures and those containing waste glass aggregates treated with silane coupling agents, with sizes of 2.36–4.75 mm and contents of 0 %, 6 %, 8 %, and 10 %. The test temperature is set at 60°C.

2.10. *Anti-skid performance test*

The anti-skid performance are evaluated based on tire-road dynamic friction testing system [44,45]. the friction coefficients were measured for asphalt mixture specimens that incorporated waste glass aggregates sized 2.36–4.75 mm with contents of 0 %, 6 %, 8 %,

Table 2
Surface free-energy parameters.

liquid	γ	γ^d	γ^p
glycerin	64.0	34.0	30.0
distilled water	72.8	21.8	51.0
formamide	58.0	39.0	19.0

Table 3
Contact angle test results between test liquid and SBS modified asphalt.

Materials	*Distilled water*	Glycerin	Formamide
SBS modified asphalt	101.48	90.3	81.78

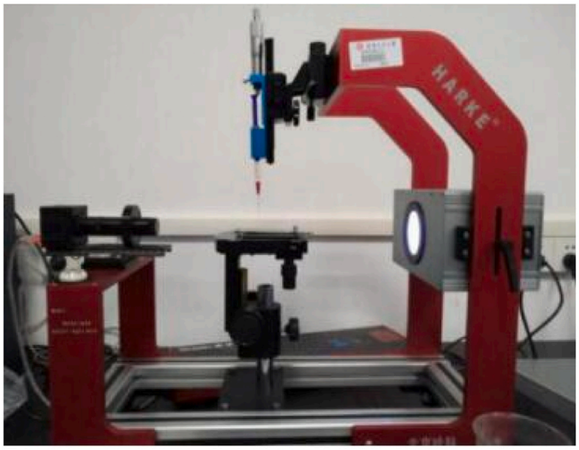


Fig. 5. Contact Angle Detector.

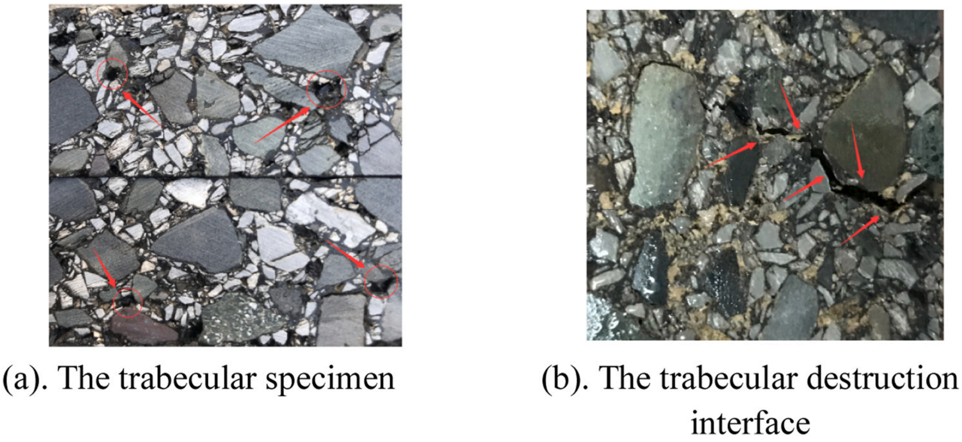


Fig. 6. Trabecular bending test samples.

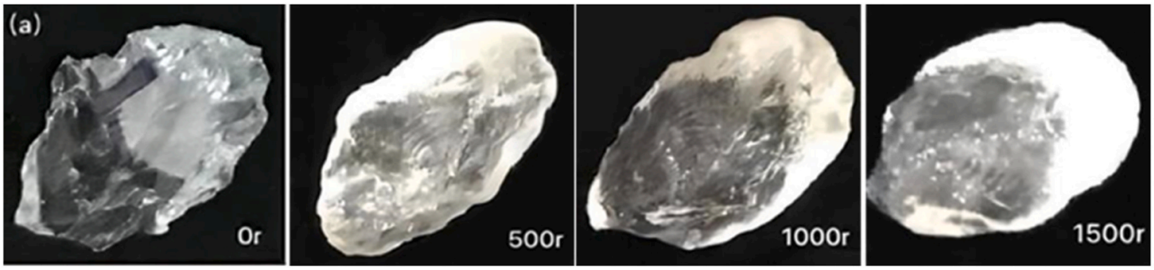


Fig. 7. Surface morphology of waste glass under different abrasion cycles.

and 10 %. Tire-road dynamic friction testing system primarily consists of a power system, hydraulic system, sensor system, and testing software “Friction Instrument Center for Analysis and Control”.

3. Results and discussion

3.1. Evolution of surface morphology of waste glass

The morphological characteristics of the waste glass surface were obtained through photography. As the number of abrasion cycles increased from 0 r to 1500 r (with each 500 r as a gradient), the smooth surfaces and edges of the waste glass gradually became rougher. As shown in Fig. 7. At the same time, the number of indentations and grooves continued to increase, leading to an increasingly complex texture on the surface of the waste glass. This is attributed to the continuous friction between the waste glass and the Los Angeles abrasion machine, as well as the mutual abrasion caused by collisions between pieces of waste glass.

The surface morphology of the abraded waste glass was further examined under a Leica microscope to observe its microscale evolution. As shown in the Fig. 8, dark phases appear on the surface of the abraded waste glass, which are the results of pits and indentations at the microscopic level. With an increase in the number of abrasion cycles, the area of the dark phase expands, indicating that abrasion imparts a complex texture to the waste glass. According to surface structure theory, a complex texture and rough surface are beneficial for the bonding between aggregate and asphalt, thereby increasing the thickness of the asphalt film.

3.2. Morphological characteristics of glass aggregate after chemical treatment

3.2.1. Morphology characteristics of glass aggregate after APTES treated

The comparison between the glass specimens before and after the silane coupling agent surface treatment is shown in Fig. 9(a) and (b). It can be observed that the surface color of the glass specimens has changed from the original transparent texture to a white film-like surface. This is due to the formation of silanol hydroxyl groups in the silane coupling agent mixture after being left to sit, which, when sprayed onto the glass surface, undergo a chemical reaction forming hydrogen bonds, followed by a dehydration reaction that creates covalent bonds, resulting in a layer of white mist-like coating on the glass surface. Subsequently, the silane component in the coupling agent undergoes hydrolysis, its silanol molecules condense with each other, and undergo polymerization, also creating a network-structured film on the glass specimen surface. This film covers the surface of the inorganic waste glass aggregate, ensuring that the asphalt film on the glass aggregate surface is not easily detached and has better adhesiveness. After treatment with APTES, a white-like coating forms on the surface of the waste glass. Macroscopic surface morphology results indicate that APTES has successfully coated the surface of the waste glass. Additionally, the APTES coating increases the surface roughness of the waste glass, which is beneficial for the bonding between the waste glass and asphalt. The protonated amine of APTES covalently condenses with

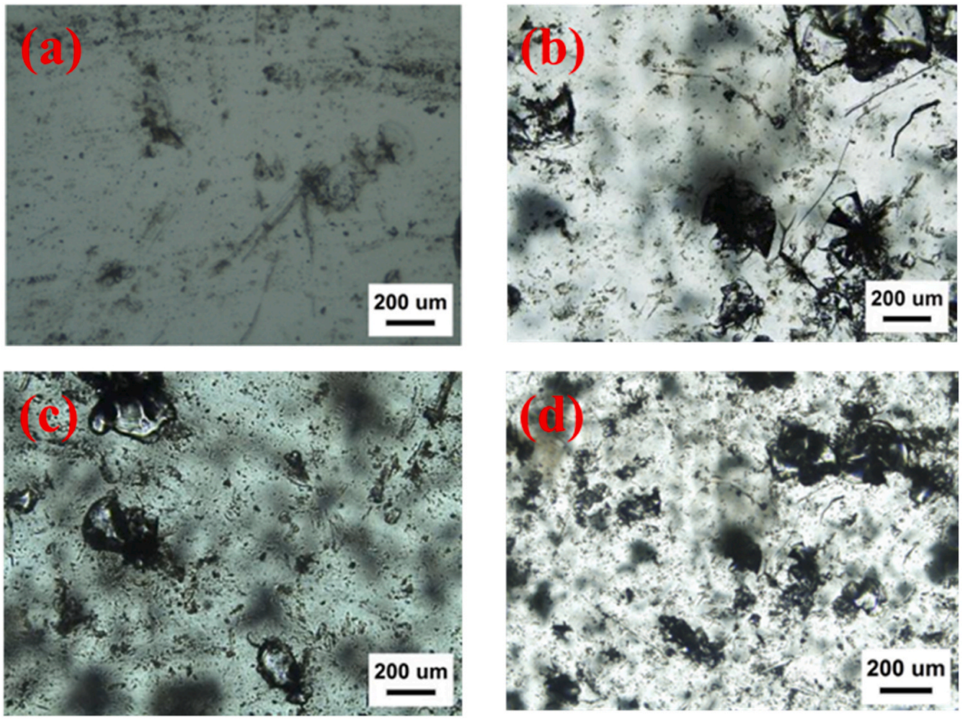


Fig. 8. Surface morphology of the abraded waste glass. (a: 0 r, b: 500 r, c: 1000 r, d: 1500 r).

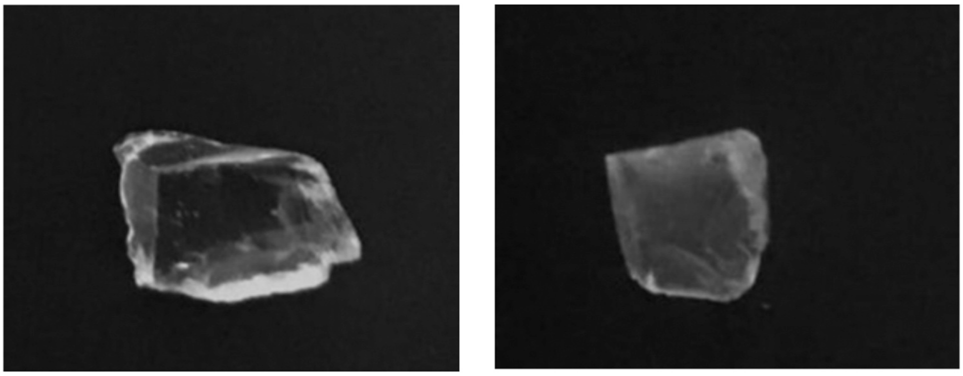

(a) Untreated (b) Surface treated

Fig. 9. Glass samples before and after coupling treatment.

silicon dioxide, thereby forming a strong bond with the surface of the waste glass.

3.2.2. EDS results of APTES treated glass particle

The EDS spot analysis was used to measure the elemental mass fraction content of the untreated glass samples and those treated with silane coupling agents and cured at 160°C. The study explored the changes in elemental composition between the two. The comparison of elemental mass fraction contents of the samples before and after surface treatment is shown in Fig. 10. It is evident that after surface treatment, there have been significant changes in the content of various chemical elements in the waste glass aggregate. This is due to the effective components in the silane coupling agent modifier, such as organic solvents and resins, which chemically adsorb on the surface of the waste glass and form a polysiloxane coupling layer, creating a gel film on its surface. From the results of elemental mass fractions, it is found that after surface treatment with the silane coupling agent, the C element from the modifier's organic long chains is introduced to the surface of the aggregate, and the content of C increased from 0 % to 35.38 %. The mass fraction of acidic group elements, such as Na, Mg, Si, etc., decreased to varying degrees. Specifically, the Na element decreased from 9.48 % to 3.5 %, and Si decreased from 37.83 % to 26.55 %. This reduction in the content of acidic oxides reduces the inert reaction state originally between the waste glass and asphalt, transforming their reactive state and enhancing the adhesiveness between the glass aggregate and asphalt.

Further discussion on the evolution of the surface morphology of waste glass after silane coupling agents silane coupling agents silane coupling agents silane coupling agents coating is based on EDS test results. The composition of the waste glass surface mainly consists of Si and O elements, along with a small amount of Na, Mg, Ca, etc., which correspond to SiO2, silicates. After APTES coating, the content of C on the waste glass surface significantly increased, which is a result of the introduction of the C chain from APTES. The protonated amine of APTES covalently condenses with silicon dioxide or self-condenses with unreacted APTES molecules. These

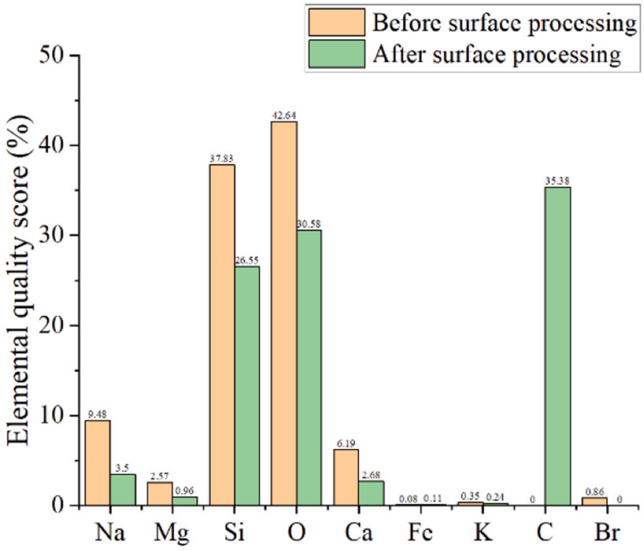


Fig. 10. Element quality fraction before and after surface treatment.

reactions promote partial polymerization of APTES, resulting in high loading of APTES on the waste glass surface. Moreover, the relative reduction in silicon dioxide content helps isolate the asphalt from acidic oxides, which in turn enhances the adhesiveness between the waste glass and asphalt.

3.3. Waste glass/asphalt interface adhesion properties

3.3.1. Analysis of waste glass/asphalt interface adhesion

The glass samples treated with boiling and silane coupling agent, designated as A (1−4) and B (1−4), are shown in Fig. 11. It is evident that compared to the untreated waste glass samples, the extent of asphalt film coverage on the surface-treated waste glass samples increased to varying degrees after boiling for 3 minutes. Additionally, the adhesive capacity between the chemically surface-treated waste glass samples and asphalt is superior to that of physically surface-treated ones. Also, based on the Los Angeles abrasion surface treatment, as the number of abrasions increased from 0r to 1500r, the area covered by the asphalt film on the waste glass samples increased, and the adhesion grade improved from level 1 to level 3. Moreover, once the abrasion exceeded 1000r, the adhesion grade tended to stabilize, enhancing the bonding between the glass and asphalt to some extent. The glass samples treated with the silane coupling agent were firmly enveloped by the asphalt film, showing no significant detachment. Samples cured at temperatures of 25°C, 60°C, 120°C, and 160°C were tightly wrapped by asphalt, reaching the highest adhesion level of 5, effectively improving the adhesion between the waste glass and asphalt.

The results of the boiling test are shown in Fig. 11. Compared to untreated waste glass, both physical abrasion and APTES coating enhanced the bonding performance between waste glass and asphalt. With increasing abrasion cycles, the adhesion level of the waste glass increased from 1 to 3. Furthermore, when the abrasion exceeded 1000r, the adhesion level of the waste glass tended to stabilize, possibly due to physical abrasion maximizing the improvement of surface roughness and micro-texture of the waste glass. However, the APTES coating showed a significantly better effect than physical abrasion. After the boiling test, compared to physical abrasion, the four APTES-coated waste glass surfaces showed a higher degree of asphalt film coverage. The adhesion rate, quantified using pixel analysis method. The adhesion rate of the asphalt film after physical abrasion increased sequentially to 43.4 % (500 r), 68.5 % (1000 r), and 76.7 % (1500 r). The number of physical abrasion cycles greatly influenced the adhesion properties of waste glass, attributed to the surface roughness and texture being susceptible to physical abrasion. Meanwhile, the adhesion rates of all four APTES-coated waste glass were higher than those of the waste glass at the maximum abrasion cycles, and the adhesion rates showed gentle fluctuations. These results indicate that the dual functionality of APTES bridges the waste glass and asphalt, creating strong chemical cross-links, and the improved adhesion effect is significantly better than physical abrasion.

The adhesion rates of glass samples treated with various surface processes are shown in Fig. 12. This Figure reveals that the interface adhesion rate between chemically treated glass and asphalt is higher than that of physically treated surfaces. In other words, chemical surface treatment of waste glass aggregate is superior to physical surface treatment processes. The glass aggregate treated with silane coupling agent shows the highest adhesion rate with asphalt, with adhesion rate values exceeding 90 % across all sample curing temperatures and showing little fluctuation. This indicates that the asphalt film can well coat the surface of the aggregate without significant detachment. Therefore, the chemical adsorption between waste glass aggregate and asphalt is superior to physical adsorption, and it can more effectively enhance the adhesion.

3.3.2. Contact angle and adhesion work analysis

The contact angle and adhesion work test results between waste glass aggregates treated with different surface processes and

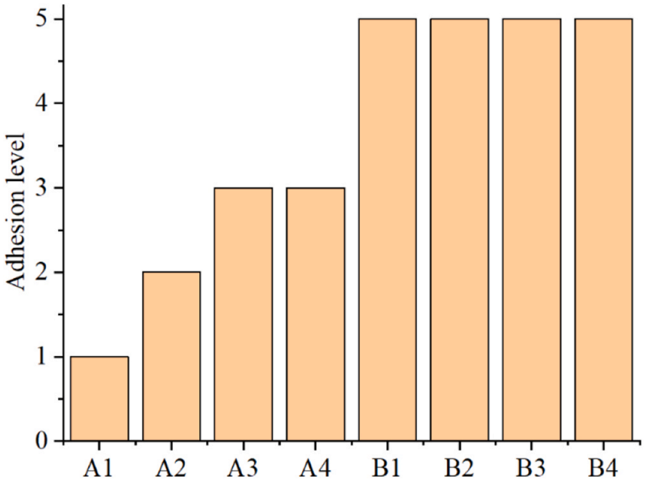


Fig. 11. Glass aggregates boiling results. Note: Los Angeles abrasion machine, at untreated control materials (A1), 500 revolutions (A2), 1000 revolutions (A3), and 1500 revolutions (A4). Immerse the waste glass in the silane coupling agent treatment and cure at 25°C, 60°C, 120°C, and 160°C for 2 hours to obtain samples B1, B2, B3, and B4 respectively.

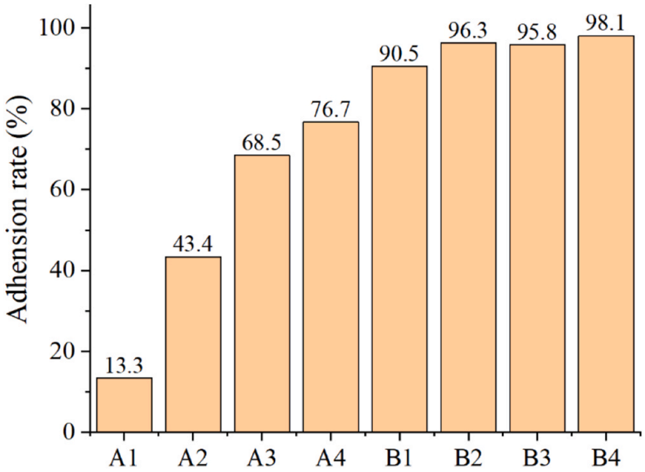


Fig. 12. Results of the adhesion rate test by the pixel analysis method. Note: Los Angeles abrasion machine, at untreated control materials (A1), 500 revolutions (A2), 1000 revolutions (A3), and 1500 revolutions (A4). Immerse the waste glass in the silane coupling agent treatment and cure at 25°C, 60°C, 120°C, and 160°C for 2 hours to obtain samples B1, B2, B3, and B4 respectively.

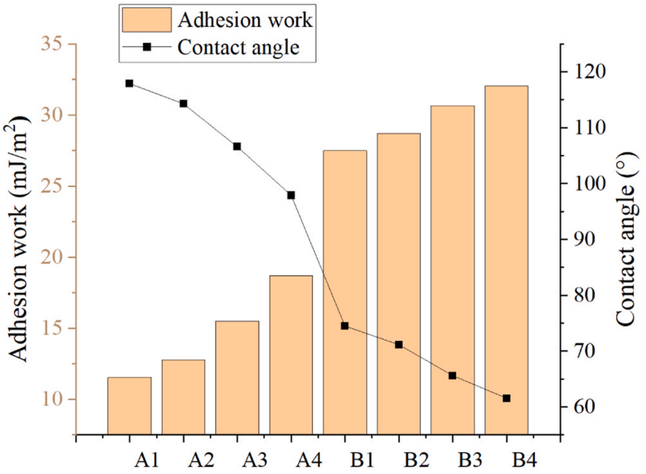


Fig. 13. Contact Angle and adhesion work test results. Note: Los Angeles abrasion machine, at untreated control materials (A1), 500 revolutions (A2), 1000 revolutions (A3), and 1500 revolutions (A4). Immerse the waste glass in the APTES mixture solution and cure at 25°C, 60°C, 120°C, and 160°C for 2 hours to obtain samples B1, B2, B3, and B4 respectively.

asphalt are shown in Fig. 13. The figure indicates that the contact angles between asphalt and waste glass aggregates treated with various surface processes display different trends. According to the theory of contact angle and adhesion work, compared to untreated surfaces, the contact angles between glass aggregates treated with physical and chemical surface processes and asphalt tend to decrease, while the adhesion work values show the opposite trend, indicating a continuous improvement in bonding ability. Moreover, the contact angles after chemical surface treatment are consistently lower than those after physical surface treatment. In other words, the bonding interaction between the two is effectively enhanced after chemical surface treatment.

For glass samples treated with silane coupling agents, as the curing temperature increases from room temperature to 160°C, the contact angle values show a decreasing trend, while the adhesion work values gradually increase. This is because the effective components in the silane coupling agent fully exert their bonding effect, strengthening the bond between the glass aggregate and asphalt. In terms of adhesion work, the order of adhesion work values between waste glass aggregates treated with different surface processes and asphalt is: silane coupling agent surface treatment > abrasion surface treatment > untreated surface. Hence, it is known that waste glass aggregates cured at 160°C based on silane coupling agent surface treatment, namely surface treatment process type B4, have the highest adhesion work value at this point, achieving the best adhesiveness between the waste glass and asphalt. Additionally, in practical engineering applications, the adhesion ability between waste glass and asphalt is effectively enhanced after silane coupling agent surface treatment.

3.4. Low temperature performance

The results of the low-temperature crack resistance tests on asphalt mixture specimens containing surface treated waste glass are shown in Figs. 14, 15, and 16. From these figures, it can be observed that after surface treatment with silane coupling agents, the flexural tensile strength and other indicators of the small beam specimens change under different surface treated waste glass content levels. Among them, the small beam specimens with a 6 % surface treated waste glass content reach the highest low-temperature performance indicators, at which point the surface treated waste glass asphalt mixture achieves optimal low-temperature crack resistance. Surface treated waste glass aggregate is weaker than natural minerals and more prone to brittle failure under low temperatures. As the content increases, it can cause insufficient overall resistance of the small beam specimens, leading to a decline in low-temperature crack resistance indicators. However, incorporating an appropriate amount of surface treated waste glass treated with silane coupling agents can effectively enhance its low-temperature crack resistance.

3.5. High temperature performance

As shown in Fig. 17, based on the surface treatment with silane coupling agents, the dynamic stability values of surface treated waste glass asphalt mixture specimens with different replacement percentages R are higher than those of conventional asphalt mixture specimens. For conventional asphalt mixture specimens, namely specimen A, the dynamic stability is 4158 cycles/mm. Compared to specimen A, the dynamic stability increases by 42.3 % for the specimen with 6 % replacement, 54.2 % for the specimen with 8 % replacement, and 3.1 % for the specimen with 10 % replacement. In other words, as the surface treated waste glass aggregate content increases from 6 % to 10 %, the dynamic stability (DS) of the asphalt mixture specimens shows a trend of initially increasing and then decreasing. Specifically, the DS increases from 6 % to 8 %, but from 8 % to 10 %, the DS decreases. When 8 % of surface treated waste glass aggregates sized 2.36–4.75 mm are used, the dynamic stability (DS) of the mixture specimen reaches the highest value of 6412 cycles/mm, indicating the best high-temperature stability performance. The reason for this is that after the silane coupling agent's treatment of the glass aggregate surface, it enhances the asphalt film thickness at the interface between glass and asphalt, forming a new tri-phase structure. This newly formed structure, compared to the original, has stronger adhesive bonds, inhibiting internal interactions within the mixture specimen, thereby improving its shear strength and enhancing high-temperature stability performance. Additionally, the angularity of the surface treated waste glass plays a role in promoting a more compact interlocking structure within the mixture.

3.6. Anti-skid performance

As shown in Fig. 18, the inclusion of glass aggregates treated with silane coupling agents has altered the friction coefficients of the mixture specimens, with consistent change trends and similar fluctuation amplitudes. The pendulum value and friction coefficient of the mixture specimens without glass aggregate is 0.44. Compared to the 0 % replacement specimens, the skid resistance friction coefficient increased by 38.6 % for the 6 % replacement specimens; it increased by 29.5 % for the 8 % replacement specimens, and by 18.2 % for the 10 % replacement specimens. In other words, as the surface treated waste glass content increased from 6 % to 10 %, the pendulum values and friction coefficients of the mixture specimens showed a gradual decreasing trend, with the friction coefficient value decreasing from 0.61 to 0.52. Particularly, when 6 % surface treated waste glass sized 2.36–4.75 mm is included, the skid resistance of the mixture specimens is optimal, with a friction coefficient of 0.61, which is about a 10 % improvement compared to

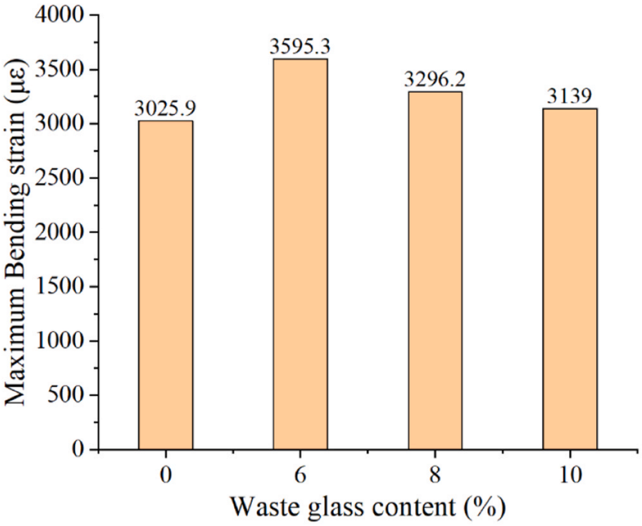


Fig. 14. Maximum bending tensile strain of asphalt mixture mixed with surface treated waste glass.

treated waste glass.

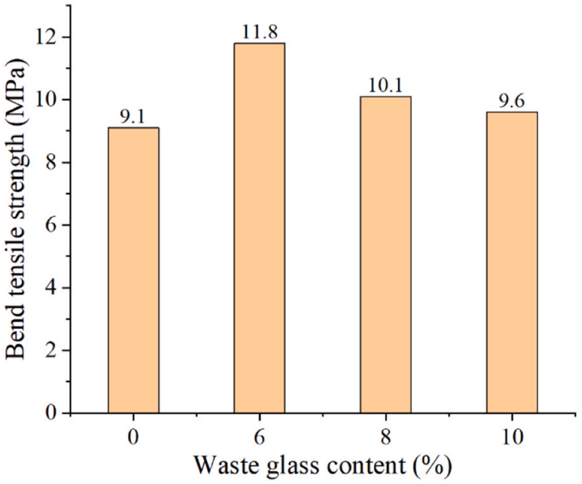


Fig. 15. Flexural tensile strength of asphalt mixture mixed with surface treated waste glass.

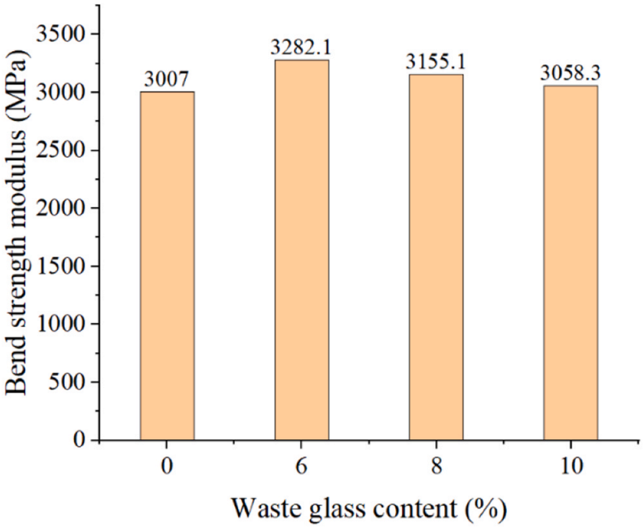


Fig. 16. Bending stiffness modulus of surface treated waste glass asphalt mixture.

conventional asphalt mixture specimens. The reason for this is that the dense surface of the surface treated waste glass aggregate increases the frictional contact area, positively contributing to friction. Additionally, the glass aggregate treated with silane coupling agent and asphalt exhibits higher cohesive forces, which also contribute to the skid resistance of the mixture. Furthermore, with the increase in content, the trend in changes in the pendulum values and friction coefficients of the mixture specimens is stable, showing only a slight decrease. It is evident that when the surface treated waste glass content ranges from 6 % to 10 %, it has a minor impact on the skid resistance performance of the mixture specimens.

4. Conclusion

This study investigates the mechanisms by which various surface modification techniques, such as physical abrasion and chemical coating, influence the adhesive performance between waste glass aggregate and asphalt. Through an integrated analysis of both micro and macro interface characterizations, the research thoroughly evaluates the debonding resistance, high-temperature stability, low-temperature performance, and skid resistance of surface-treated waste glass mixtures. The study led to the following conclusions:

(1). Surface modification significantly reduces the surface inertness of waste glass. Physical abrasion increases the surface roughness of the waste glass, imparting surface texture, which is positively correlated with the number of abrasions. Due to the effective bridging action of the dual-functional characteristics, 3-aminopropyltriethoxysilane coupling agent enhances the adhesion at

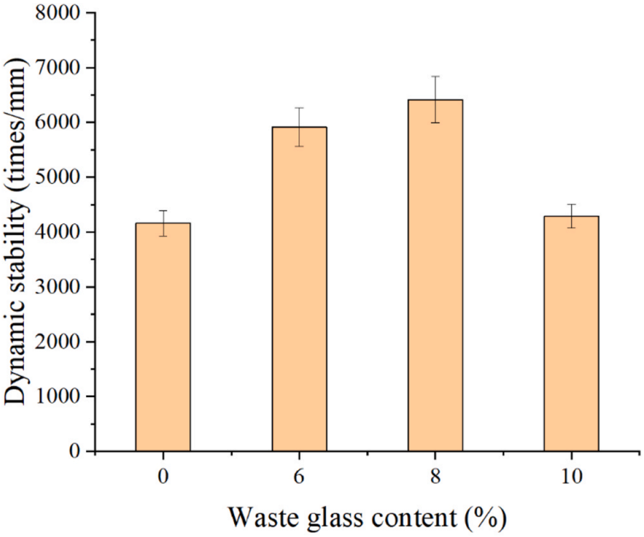


Fig. 17. High temperature test results.

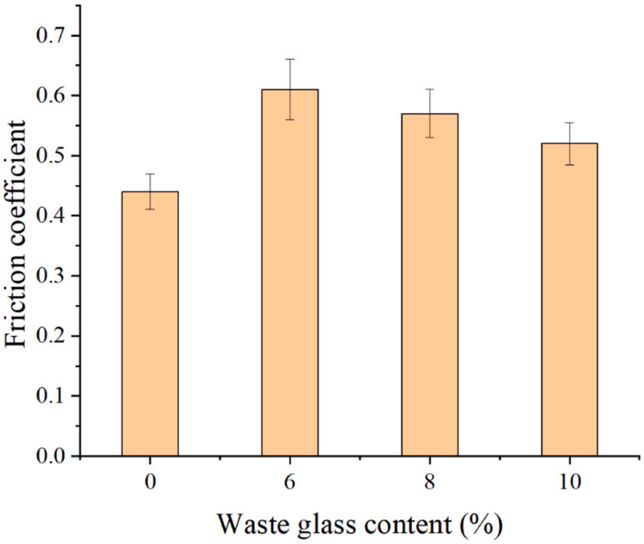


Fig. 18. Anti-skid test results of the asphalt mixture with different surface treated waste glass content.

the interface between waste glass aggregate and asphalt. Furthermore, the optimal curing temperature for APTES is found to be 160°C.

(2). Surface modification effectively improves the debonding resistance of waste glass asphalt mixtures. Based on the contact angle tests, the adhesion work of three types of waste glass asphalt mixtures is ranked as follows: silane coupling agent treatment > abrasion treatment > untreated. The adhesion work results confirm that the chemical cross-linking of silane coupling agents is superior to physical abrasion.

(3). Incorporating waste glass appropriately treated with silane coupling agents enhances the low-temperature crack resistance and high temperature rutting resistance of asphalt mixtures. The 6 % surface treated waste glass in asphalt mixture can lead to a 42.3 % increase in dynamic stability compared to conventional mixtures.

(4). The surface treated waste glass with silane coupling agents treated increased the skid resistance of the mixture. It is recommended that the surface modification process for waste glass is APTES coating and recommend content of waste glass is 6 % by total aggregate weight.

This study explores the surface modification of waste glasses and their impacts on the debonding and high-temperature, low-temperature, anti-skidding performance of waste glass asphalt mixtures, providing process and theoretical references for the practical application of waste glass asphalt roads.

Future work

The long-term performance of the recycled glass-modified asphalt mixture still requires verification. Additionally, further research should focus on optimizing surface modification techniques and material types (such as KH540, KH560, and KH570) to improve durability and evaluating the environmental impact of incorporating treated waste glass into asphalt applications. In addition to performance testing, a detailed cost analysis will be conducted including materials cost, process cost and maintenance costs.

CRediT authorship contribution statement

Zhanping You: Methodology, Conceptualization. **Yalong Li:** Methodology, Conceptualization. **Dongzhao Jin:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Investigation, Conceptualization. **Yu Liu:** Resources, Methodology, Funding acquisition. **Miao Yu:** Writing – original draft, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of Competing Interest

We declare that this manuscript is original, has not been published before and is not currently being considered for publication elsewhere.

Data availability

Data will be made available on request.

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