

Article

Regenerative and Innovative Utilization of Coffee Residue and Waste Cooking Oil: Improving Rheological Properties of Recycled Asphalt

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Abstract: Currently, there is limited research on the utilization of spent coffee grounds (SCG) in asphalt pavement. This study explores using SCG as a novel rejuvenator together with waste cooking oil (WCO) to enhance the performance of aged asphalt (AA). The high-temperature performance of recycled asphalt was preserved using SCG containing oily components. However, the low-temperature performance of long-term aged asphalt could not be completely restored to the level of virgin asphalt. Therefore, various dosages of SCG and WCO were utilized to optimize the recovery of low-temperature properties while maintaining high-temperature performance. The recycled asphalt (RA) was analyzed through conventional indexes, microscopic characteristics, and rheological properties using penetration and softening point tests, Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and a dynamic shear rheometer (DSR). The results showed that the G^* of W_7S_{12} increased by 90% relative to virgin asphalt. Additionally, at strain levels of 2.5% and 5%, the fatigue life of W_8S_{18} was approximately 3.39 times and 2.34 times greater, respectively, than that of the virgin asphalt. The addition of a rejuvenator can enhance the low-temperature cracking resistance of aged asphalt. Moreover, the FTIR results indicated that the regeneration mechanism of recycled asphalt consisted of physical blending. In summary, W_7S_{12} exhibited the highest high-temperature performance, while W_8S_{18} demonstrated superior fatigue life. This study may promote the sustainable development of asphalt pavements by utilizing organic waste as a rejuvenator through resource recovery.

Keywords: aged asphalt; rejuvenator; spent coffee grounds; waste cooking oil; recycled asphalt; rheological properties



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1. Introduction

As China's economy experiences rapid growth, ecological and environmental concerns have increasingly gained prominence. In response to the global push for energy conservation and emission reduction, the development and utilization of ecological materials may offer significant solutions to current environmental issues. Biomass consists of a renewable energy source that can decrease carbon dioxide emissions. The utilization of biomass energy plays an important role in achieving carbon-neutral and peak carbon goals. Biomass, which contains lignocellulose, can be used as a carbon-neutral raw material due to its large storage capacity, environmental compatibility, and sustainability advantages [1–3].

Coffee, ranked among the top three beverages worldwide, has the potential to be utilized as a carbon-neutral raw material [4,5], utilizing coffee residue as a natural biomass raw material [6]. Therefore, spent coffee grounds (SCG) can function effectively as a natural biomass feedstock.

The International Coffee Organization (ICO) projects that by 2024, the global coffee market will generate approximately 18.91 million tons of coffee. Additionally, global coffee production is anticipated to grow by 5.8% year-on-year, reaching 178 million 60 kg bags between 2023 and 2024 [7]. In addition, more than 90% of the initial dry mass of coffee is produced as SCG after use [8]. At present, SCG is typically treated by incineration or landfill, producing bacteria and mold, and polluting the environment [9,10]. The issue of pollution may be solved by recycling SCG to treat domestic and even global SCG. Research found that SCG contains cellulose, hemicellulose, lignin, protein, oil, and small amounts of other substances [11,12]. Mariana et al. prepared activated carbon from SCG [13,14], while Lin et al. successfully isolated lignin–polysaccharide complexes from SCG, which had good antioxidant properties and could shield against ultraviolet rays [15]. Because SCG has a high oil content, studies found that the percentage of oil content in different types of SCG was 11–21%, according to relevant experiments [16]. The extraction of oil from SCG was more efficient when non-polar solvents were used. The oil extracted from SCG demonstrated better antioxidant properties and stability than other sources [17]. Manjunah et al. investigated the partial replacement of cement with 15% coffee cherry waste and found that this substitution increased the compressive strength by 23% at 28 days. Additionally, the cement strength index reached its lowest point, and the amount of cement required to achieve a compressive strength of 1 MPa was reduced by 31% [18]. Ji et al. reported that adding SCG biochar significantly improved the complex shear modulus (G^*) and the rutting factor ($G^*/\sin \delta$) of modified asphalt. Additionally, it caused a reduction in the phase angle (δ), which contributed to the enhancement of the asphalt's stability under high temperatures [19]. Research has demonstrated that incorporating SCG biochar and bio-oil as rejuvenators greatly improves the properties of aged asphalt. Rheological analysis at high temperatures indicated that the cumulative strain of asphalt rejuvenated with 8% SCG biochar decreased by 489.2% compared to unmodified virgin asphalt. Additionally, the $G^*/\sin \delta$, derived from the G^* and δ , increased by 43.9%, indicating significant improvements in high-temperature performance [20]. Xie et al. enhanced virgin asphalt by incorporating SCG and found that this modification reduced the non-recoverable creep compliance, J_{nr} (0.1) and J_{nr} (3.2), by 22.7% and 22.5%, respectively, compared to unmodified asphalt. Moreover, the recovery rate, R (3.2), increased by 140%, demonstrating that adding SCG greatly enhances the rutting resistance of asphalt under high-temperature conditions [21]. Adding SCGs to geopolymer combined with various mineral additives can greatly improve its stability and mechanical properties, making it an effective filler for road bases or structural components [22]. Meanwhile, asphalt is widely used in the pavement industry in China, due to its numerous advantages. These include providing a more comfortable driving experience, ease of maintenance, reduced noise pollution, and improved skid resistance. Consequently, asphalt pavement has emerged as the most widely utilized type of pavement across the globe [23]. Nevertheless, different environmental conditions, including the influence of sunlight and moisture, can cause cracks and gaps in the asphalt, loosening, and other problems, thus shortening the service life of asphalt pavement. Recycled asphalt pavement (RAP) technology has been utilized in pavement construction to protect the environment and reduce costs [24]. RAP has become an asphalt pavement research hotspot [25], even though incorporating RAP in asphalt mixtures can lead to premature failure due to inadequate performance, meaning the aged asphalt needs to be rejuvenated.

According to statistics, studies have been conducted on rejuvenator types, including waste vegetable oil, waste motor oil, and paraffin oil; despite this, insufficient effects were observed [26]. At present, waste cooking oil (WCO) has demonstrated good potential as a potential rejuvenator. Wang et al. enhanced the fatigue resistance of asphalt by incorporating waste cooking oil and polyphosphoric acid into virgin asphalt [27]. Organic montmorillonite as a rejuvenator for aged asphalt improved the high-temperature performance of recycled asphalt. However, excessive use of WCO negatively impacted the high-temperature performance of the recycled material [28]. Wang et al. pretreated waste cooking oil (WCO) through glycerol esterification before adding it to aged asphalt. The recycled asphalt produced from deeply esterified WCO demonstrated outstanding high-temperature fatigue resistance [29]. Feng et al. explored the surface activity of waste cooking oil (WCO) in combination with crumb rubber to enhance the properties of crumb rubber-modified asphalt. This method significantly boosted the material's high-temperature performance [30]. Additionally, WCO was examined as a rejuvenator for aged asphalt because of the low volatility of its lightweight components [31]. But still, some components in WCO may influence asphalt performance [32]. Ensuring that WCO, as a rejuvenator, retains its high-temperature performance remains a challenge that requires further resolution [33].

Inspired by the above studies, it can be found that all the components in SCG can be extracted and applied by relevant methods, but are hardly ever studied in aged asphalt. Jalkh, R. et al. showed that WCO and SCG oil could be used as asphalt binders in the future by evaluating their physicochemical properties [34], taking into account the porous structure of coffee grounds. It is expected that adding SCG to aged asphalt can improve its performance. However, the process of extracting oily substances from SCG is complicated. This research provides a new recycling method with less pollution and a higher utilization rate while taking into account the objective content of oil components in SCG. This study focused on evaluating the feasibility of using SCG and WCO to regenerate aged asphalt for pavement maintenance and rehabilitation. It assessed the basic physical properties of the recycled asphalt. The effect of dosages of SCG and WCO on regeneration was assessed by dynamic shear rheology (DSR), multiple stress creep recovery (MSCR), and linear amplitude sweep (LAS) analysis. The rheological properties of recycled asphalt were analyzed as well. This analysis aims to enhance the future application of these materials in recycled asphalt pavements.

2. Materials and Methods

2.1. Materials

2.1.1. Virgin Asphalt and Aged Asphalt

In this study, 70# virgin asphalt was provided by a company in Linyi. Long-term aged asphalt is obtained by subjecting the virgin asphalt to a pressure aging vessel (PAV) per ASTM D2872 [35] at a temperature of 100 °C and a pressure of 2.1 MPa for 20 h. Table 1 shows the technical indices of the asphalt.

Table 1. Technical indices of the asphalt.

Index	Virgin Asphalt	Aged Asphalt	Test Methods [36]
Penetration (25 °C)/0.1 mm	63.2	22.6	T 0604-2011
Softening point (°C)	48.7	68	T 0606-2011
Ductility (15 °C)/cm	>100	brittle fracture	T 0605-2011

2.1.2. Spent Coffee Grounds

SCG was purchased from a coffee shop supplied by a coffee bean plantation in Yunnan Province. The coffee beans are large and contain moisture after brewing, so they need to be ground into 120-mesh powder to better disperse and fully dissolve in the asphalt, thus better utilizing their porous structure characteristics. The majority of SCG had similar compositions, with cellulose and hemicellulose accounting for 50%, lignin accounting for 20% [37,38], and oil accounting for 15–20% [39]. Furthermore, SCG contains various phenolic compounds, with antioxidant effects [40]. The surface of the dried SCG was treated with a gold coating. In Scanning Electron Microscopy (SEM) mode, the surface morphology of the SCG was first examined at a low magnification, and specific regions of interest were selected for Energy Dispersive Spectroscopy (EDS) analysis and the collection of spectral data. The primary chemical elements of SCG were identified based on characteristic peaks in the spectrum. The SEM images of SCG were processed using specialized software for analysis. The resulting data was then imported into OriginPro 2021 software to create a particle size distribution diagram. This was done to determine whether the particle size distribution conforms to a normal distribution. Figure 1b shows that the average particle size of SCG is 31.7 μm . Figure 2 illustrates the microstructure of the SCG surface. Figure 2a presents an SEM image of coffee grounds at 100 $\times$ magnification, revealing an irregular shape. Figure 2b and 2c show images of location A at magnifications of 1000 $\times$ and 3000 $\times$, respectively, while Figure 2d,e depict location B at the same magnifications. It can be seen from the red circles in the figure that SCG has a porous structure. By examining the images at different magnifications of two distinct locations, it becomes apparent that the surface of the SCG is characterized by protruding particles and a rough, porous texture. This porous structure increases the specific surface area of the SCG, thereby enhancing its adsorption capacity and improving its binding effectiveness with asphalt. Moreover, the irregular shape of the SCG facilitates the formation of a skeletal network structure within the asphalt. Table 2 presents the analysis of chemical elements in SCG [41].

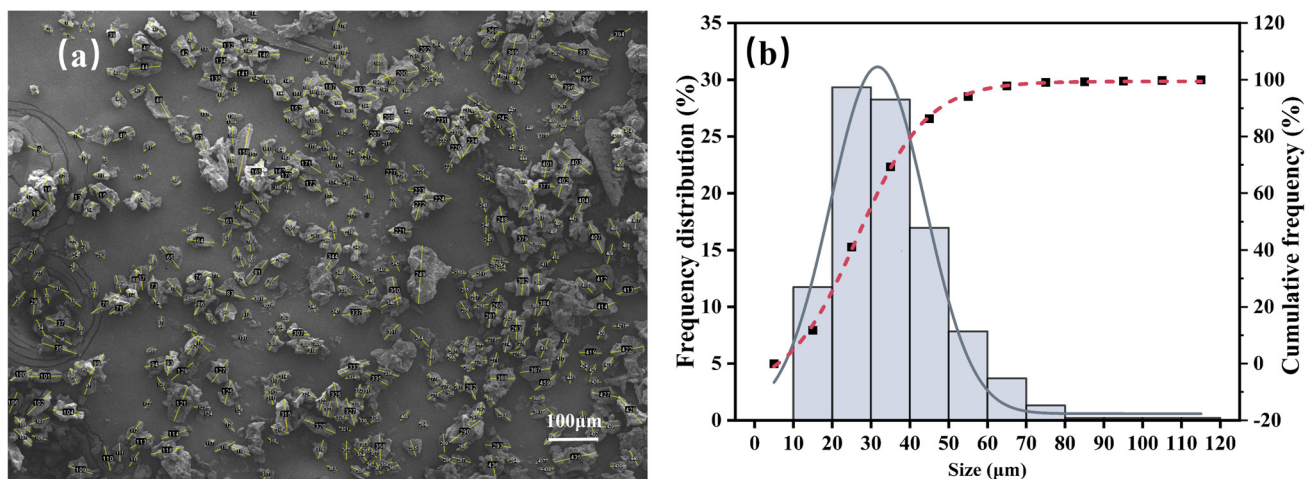


Figure 1. (a) Particle size measurement markers; (b) the particle size distribution of coffee grounds.

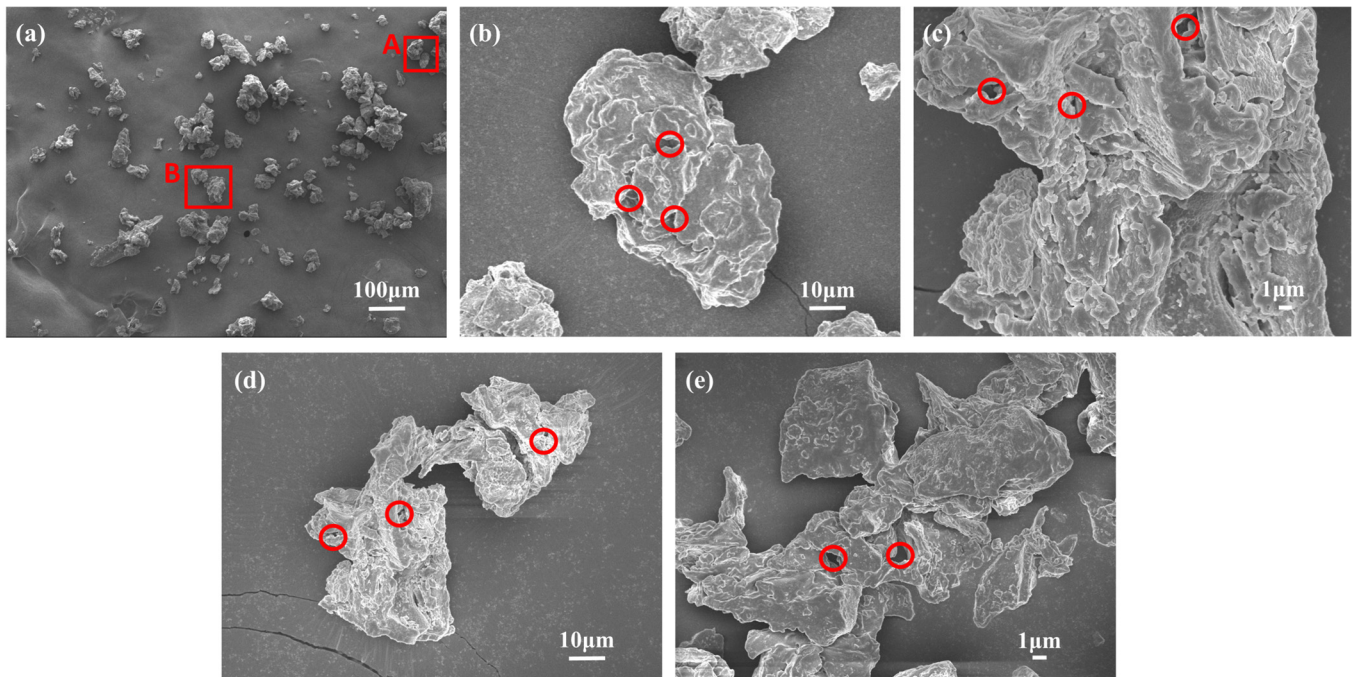


Figure 2. SEM images of coffee grounds: (a) 100×; (b) 1000×; (c) 3000×; (d) 1000×; (e) 3000×.

Table 2. SCG elemental analysis.

C (%)	H (%)	O (%)	N (%)	S (%)
56.10	7.20	34.00	2.40	0.14

2.1.3. Waste Cooking Oil

WCO was obtained by processing post-cooking oil obtained from a catering company in Hubei province. Table 3 shows the physical and chemical properties of WCO.

Table 3. Physical and chemical properties of WCO.

Property	Technical Indicators	Test Methods
Acid value (mg KOH/g)	0.85	ASTM D974 [42]
Flashpoint (°C)	232	ASTM D3794 [43]
Water content (%)	0.09	ASTM D4006 [44]
Mass loss (%)	0.47	ASTM E2402 [45]
Density (g/cm ³)	0.91	ASTM D1298 [46]

2.2. Determination of the Test Dosage

The initial amount of rejuvenator was determined through pre-experiments including penetration tests and softening point tests. Figure 3 illustrates how the penetration and softening point indices of short-term and long-term aged asphalt change with the incorporation of SCG. Figure 3 demonstrates that incorporating SCG into short-term aged asphalt can effectively restore the penetration index to a level comparable to virgin asphalt while exerting minimal influence on the softening point. The addition of SCG has been shown to preserve the high-temperature stability of recycled asphalt. This effect is attributed to the porous structure of SCG, which absorbs oily components and reduces the light fractions in the asphalt. When the SCG content was at 20%, the penetration and softening point exhibited a gradual change trend. Consequently, the SCG contents were subsequently determined as 12%, 15%, and 18%. Figure 3 shows the inability to restore long-term aged asphalt to its virgin state; therefore, a suitable amount of WCO was added

for regeneration. Nonetheless, due to the unstable regenerative effect of WCO on aged asphalt [47], SCG exhibited antioxidant capabilities [48], resulting in a blend of SCG and WCO for recycled asphalt.

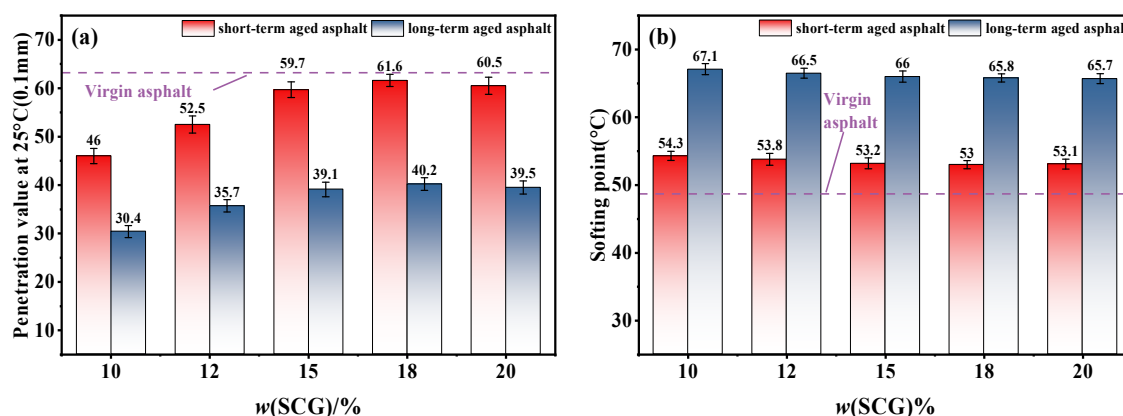


Figure 3. Physical indicators of the short-term aged and long-term aged asphalt: (a) penetration; (b) softening points.

Figure 4 shows the variation trends in penetration and softening points of SCG recycled asphalt as different amounts of WCO are added. It can be seen from the figure that WCO contains chemical components similar to the saturation and aromatic classification of asphalt components, which could supply the light fraction lacking in aged asphalt, improve its colloidal structure, soften aging asphalt, increase its penetration and reduce its softening point. Figure 4a shows that at a WCO content of 6%, the penetration of recycled asphalt remains lower than that of virgin asphalt. Similarly, Figure 4b indicates that at 9% WCO content, the softening point of recycled asphalt could not be fully restored to align with that of virgin asphalt. However, when the WCO contents were 7% and 8%, the penetration and softening points met the standards set for virgin asphalt. As a result, the optimal WCO contents were set to 7% to 8%.

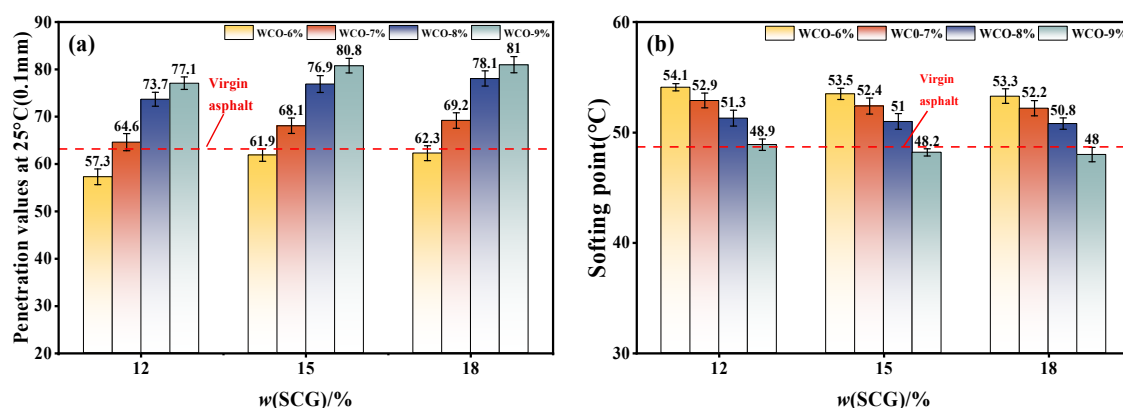


Figure 4. Penetration and softening point of the recycled asphalt with different proportions of WCO and SCG: (a) penetration; (b) softening points.

2.3. Recycled Asphalt Preparation

(1) First, SCG and WCO were placed in an oven set to 105 °C and dried until all moisture was entirely eliminated.

(2) The aged asphalt was heated in an oven at 135 °C until achieving a fluid consistency. Subsequently, various proportions of WCO (by mass of RA) were incorporated into the asphalt to ensure full integration, and then placed in a shearer at 150 °C for 30 min at 3000 r/min.

(3) Next, SCG (by mass of RA) was added to the prepared asphalt, heated at 150 °C for 10 min, and stirred at a speed of 1000 r/min. Afterward, the mixture was agitated at 3000 r/min for 30 min to achieve complete expansion.

(4) Finally, the material was placed in an oven kept at a constant 135 °C for 30 min, ensuring the production of homogeneous and well-dispersed samples.

Finally, six groups consisting of SCG and varying levels of WCO were designed to be mixed into aged asphalt to evaluate their regenerative effect and determine the optimal dosage. The details of each dosage are shown in Table 4.

Table 4. Test dosage of recycled asphalts.

Samples	Ingredient	Samples	Ingredient
W ₀ S ₀	VA + 0%WCO + 0%SCG (by mass of VA)		
W ₇ S ₁₂	RA + 7%WCO + 12%SCG	W ₈ S ₁₂	RA + 8%WCO + 12%SCG
W ₇ S ₁₅	RA + 7%WCO + 15%SCG	W ₈ S ₁₅	RA + 8%WCO + 15%SCG
W ₇ S ₁₈	RA + 7%WCO + 18%SCG	W ₈ S ₁₈	RA + 8%WCO + 18%SCG

2.4. Experimental Methods

2.4.1. Basic Index Experiment

The basic index experiment was performed following the JTG E20-2011 [36] test method. The physical properties of both virgin and recycled asphalt were evaluated by measuring their penetration at 25 °C and softening point. For the penetration test, the average value for each dosage was determined from three sets of data, while the softening point was calculated using the average of two sets of data for each dosage.

2.4.2. Temperature Frequency Experiment

The high-temperature performance of recycled asphalt was examined through a temperature–frequency sweep test. Temperature–frequency sweep tests were performed in 6 °C increments, covering a range from 46 °C to 82 °C. The stress level remained constant at 5%, while the angular frequency for the tests was fixed at 10 rad/s. In addition, the angular frequency for the frequency scanning experiments varied between 0.1 and 100 rad/s. During the tests, the complex shear modulus (G^*), phase angle (δ), and rutting factor ($G^*/\sin \delta$) were measured, and three samples of each asphalt were taken for repeated testing. A master curve was developed using the Williams–Landel–Ferry (WLF) equation, with a reference temperature of 64 °C, derived from the results of temperature–frequency sweeping (Equation (1)). Additionally, fitting the master curves for each blend of recycled asphalt over a wider range of conditions was achieved through the CAM model Equation (2):

$$\log a_T = -\frac{C_1(T - T_r)}{C_2 + (T - T_r)} \quad (1)$$

where a_T is the displacement factor, T is the test temperature (°C), T_r denotes the reference temperature (°C), and C_1 and C_2 are the equation coefficients.

$$|G^*| = \frac{G_g^*}{\left[1 + \left(\frac{\omega_c}{\omega_r}\right)^v\right]^{\frac{w}{v}}}, \quad (2)$$

where G_g^* is glass modulus (Pa), ω_c is the crossover angular frequency, ω_r is the curtailment angular frequency, $\omega_r = \omega \times a_T$, and w and v serve as the main curve fitting parameters.

The resistance of recycled asphalt to low-temperature cracking was evaluated through temperature–frequency sweep tests. The asphalt materials' complex shear modulus (G^*)

and phase angle (δ) were measured at temperatures of 5 °C, 15 °C, 25 °C, and 35 °C. Subsequently, the G–R constants were calculated according to Equation (3), and black space diagrams were plotted for visualization:

$$G - R = \frac{G^* \times (\cos \delta)^2}{\sin \delta} \quad (3)$$

2.4.3. Multiple Stress Creep Recovery Experiment

The multiple stress creep recovery (MSCR) experiment was used to evaluate the asphalt's resistance to permanent deformation under high-temperature conditions [49]. At a test temperature of 64 °C, stress cycles of 0.1 kPa and 3.2 kPa were applied through creep testing. Each stress level consisted of 10 cycles, where each cycle included a 1 s loading (creep) phase followed by a 9 s unloading (recovery) phase. The high-temperature performance of the asphalt was evaluated through two key indicators: the elastic recovery rate (R%) and the non-recoverable creep compliance (J_{nr}). Three samples of each asphalt were taken for repeated testing. The elastic recoverable rate (R%) was calculated by Equation (4) and the non-recoverable creep (J_{nr}) was calculated by Equation (5):

$$R = \frac{\varepsilon_p - \varepsilon_u}{\varepsilon_p} \times 100\% \quad (4)$$

where ε_p is the peak strain, and ε_u is the unrecovered strain.

$$J_{nr} = \frac{\varepsilon_u}{\sigma} \times 100\% \quad (5)$$

where σ is the shear stress.

2.4.4. Linear Amplitude Sweeping Experiment

The linear amplitude sweep (LAS) test, following the guidelines of AASHTO TP101 [50], was employed to evaluate the fatigue resistance of asphalt by accelerating its level of damage. The LAS test was divided into two distinct stages. In the first stage, a frequency sweep was performed at a loading strain of 0.1%, with the frequency range spanning from 0.1 to 30 Hz. In the second stage, amplitude sweeping was carried out, where the loading strain increased linearly from 0.1% to 30% under a fixed frequency of 10 Hz and a total loading duration of 300 s. Three samples of each asphalt were taken for repeated testing. The LAS test temperatures were all 25 °C. The fatigue prediction equation was constructed as shown in Equation (6):

$$N_f = A \times (\gamma_{\max})^{(-B)} \quad (6)$$

where N_f is the predicted asphalt fatigue life, $\gamma_{\max}$ is the predicted maximum strain of the pavement structure, and A and B signify fatigue equation parameters.

2.4.5. Scanning Electron Microscopy

The microstructure of SCG and recycled asphalt was analyzed using the Hitachi-Regulus8230 scanning electron microscope from Hitachi, Tokyo, Japan. SCG required a conductive adhesive to fix a small amount of powder sample onto the specimen stage for testing. The asphalt samples used in this study were prepared following the method described in Section 2.3; the prepared asphalt was taken from its natural surface to ensure that it was uncontaminated, with no further tests performed on it. Since asphalt is non-conductive, gold sputtering was applied prior to characterization to enhance its

conductivity and stabilize image quality. The magnification range for microstructural characterization was between 100 and 3000 times.

2.4.6. Fourier Transform Infrared Spectroscopy Analysis

To investigate the microscopic mechanism of the effects of WCO and SCG on the properties of aged asphalt, the asphalt and WCO were dissolved in a tetrahydrofuran solvent, and then the solution was dropped on a potassium bromide chip. Meanwhile, the SCG was combined with anhydrous potassium bromide (KBr) powder. The changes in functional groups were investigated by using the Bruker Vertex 70 spectrometer from Germany, with the spectra recorded in the range of 4000 to 400 cm^{-1} , scanning 32 times, with scanning resolution of 4 cm^{-1} . The asphalt samples used in this study were prepared following the method described in Section 2.3. Three samples were taken for repeated experiments in each test.

3. Results and Discussion

3.1. Penetration and Softening Point

In practical engineering, the penetration and softening points serve as important and common parameters for evaluating asphalt which can indirectly evaluate the deformation resistance, high-temperature stability, and other related indexes [51]. Figure 5 presents the basic parameters of virgin asphalt, aged asphalt (AA), and varying dosages of recycled asphalt. The results indicate that long-term aging of virgin asphalt led to a reduction in penetration and an increase in the softening point. This phenomenon can be explained by the transformation of light components in asphalt into asphaltenes during thermal oxygen aging, which results in an overall increase in the consistency of the asphalt [52]. Adding only SCG resulted in an increase in penetration and a gradual decrease in the softening point as the dosage rose. Notably, when the SCG dosage reached 18%, the penetration of virgin asphalt increased by 33.4%, while the softening point decreased by 5.5%.

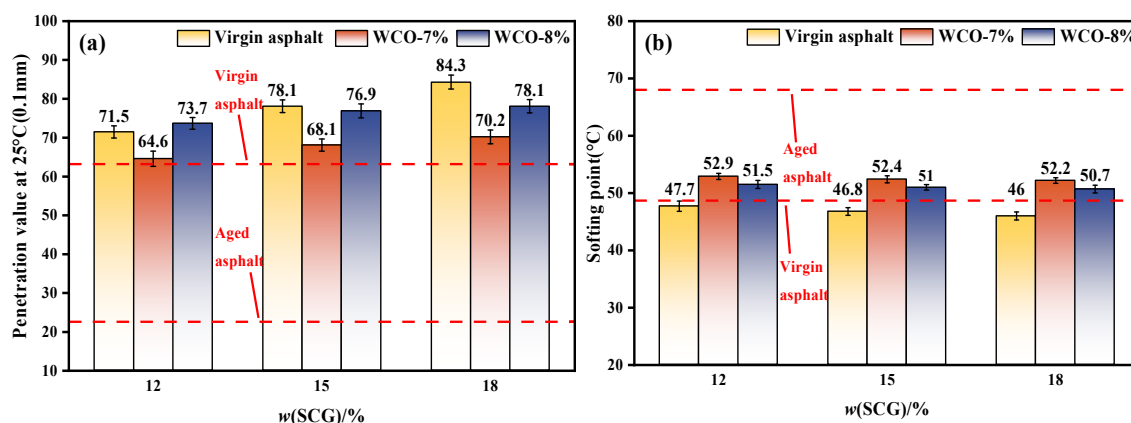


Figure 5. Conventional properties of asphalt: (a) penetration; (b) softening point.

The simultaneous addition of both SCG and WCO resulted in the recovery of the penetration of aged asphalt, surpassing that of virgin asphalt. When the WCO dosage was kept constant, increasing the SCG dosage resulted in a corresponding rise in penetration, which was attributed to the oily components in SCG [53], which further softened the asphalt. At the W_8S_{18} dosage level, the maximum penetration value was observed. This dosage led to a recovery rate of 230.9% relative to the aged asphalt and demonstrated a 23.6% improvement when compared to the virgin asphalt. Although the softening point of the aged asphalt showed a declining trend, it remained above that of the virgin asphalt. When comparing the softening point with the aged asphalt, the W_7S_{12} dosage showed

a reduction of 21.4%, while an improvement of 11.6% was observed when compared to the virgin asphalt. The results suggested that recycling with SCG and WCO enhanced the high-temperature performance of the asphalt.

3.2. Temperature Sweeping Analysis

Figure 6 presents the complex modulus (G^*) and phase angle (δ) of the recycled asphalt under varying temperatures and content levels. These parameters play an important role in describing the viscoelastic behavior of asphalt materials.

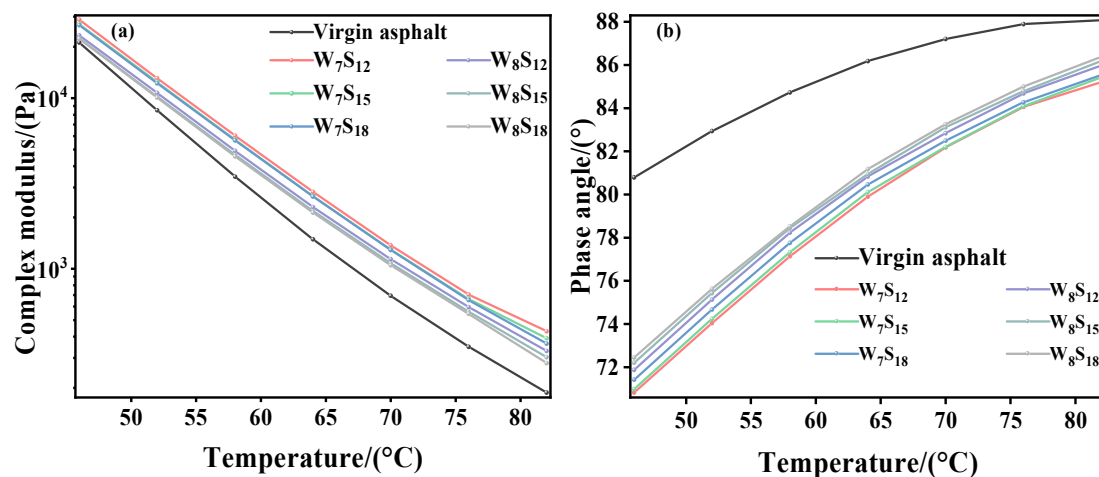


Figure 6. Temperature sweep test results: (a) complex modulus variations with temperature; (b) phase angle variations with temperature.

Figure 6 illustrates the decrease in G^* in all asphalt binders and the gradual increase in δ as the temperature increased. As shown in Figure 6a, the G^* of W_7S_{12} increased by 90% compared to virgin asphalt at 64°C , indicating its superior high-temperature performance. With the same WCO content, G^* gradually changed with SCG addition. This phenomenon was primarily ascribed to the development of a denser mesh-like spatial structure formed through the interaction at the interface between SCG and the polymer matrix. Moreover, in this process, the shear load exerted on the polymer matrix facilitated the transfer of stress from the low-modulus carrier asphalt to the high-modulus SCG. The efficiency of load transfer depended on both the size of the particles and their total surface area. The strength of recycled asphalt improved as the specific surface area of filler particles increased. This enhancement was attributed to the substantial specific surface area provided by SCG [54]. The recycled asphalt G^* values were higher than the virgin asphalt, indicating greater shear deformation resistance in the asphalt binder. A larger G^* value signified stronger resistance to shear deformation and less susceptibility to road surface rutting.

As illustrated in Figure 6b, the phase angle of the asphalt binder progressively increased as the temperature rose. This was due to the temperature increase causing the asphalt to transition from elastic to viscous. When the temperature approached 90°C , it almost completely lost its elasticity [55]. Across all temperatures, the value of δ remained consistently lower than that of the virgin asphalt. Additionally, it was found that the use of 7% WCO resulted in lower values than using 8% WCO in all cases, which could be attributed to WCO containing a large amount of light fraction providing a viscous fraction to the asphalt. When the WCO content remained constant, increasing the SCG content caused δ to decrease more gradually as the temperature rose. This suggests that adding SCG enhanced the elasticity of the recycled asphalt. This phenomenon was primarily attributed to the SCG powder's larger specific surface area and rough texture. These properties promoted the development of a spatial network structure upon combination with

asphalt, which improved its ability to adsorb additional binder. This process effectively reduced the mobility and adjusted the viscosity of the aged asphalt, thereby improving its high-temperature rheological properties, which aligned with the G^* results.

To comprehensively assess the impact of G^* and δ on asphalt's high-temperature performance, the rutting factor was measured through a temperature sweep test. This approach provided insights into the asphalt's rheological behavior under elevated temperatures, as illustrated in Figure 7. As the temperature rose, the rutting factor of all asphalt binders showed a decline. This behavior was attributed to the asphalt transitioning from a viscoelastic state to a predominantly viscous state at elevated temperatures. Consequently, its fluidity increased while its resistance to deformation diminished [56]. Throughout all temperature ranges, the rutting factor of recycled asphalt was consistently higher than that of virgin asphalt. Specifically, W_7S_{12} exhibited the highest $G^*/\sin \delta$ and optimal high-temperature rutting resistance across all temperature ranges, which was consistent with the trend of G^* . This suggested that asphalt recycled with SCG and WCO demonstrated superior high-temperature rheological performance.

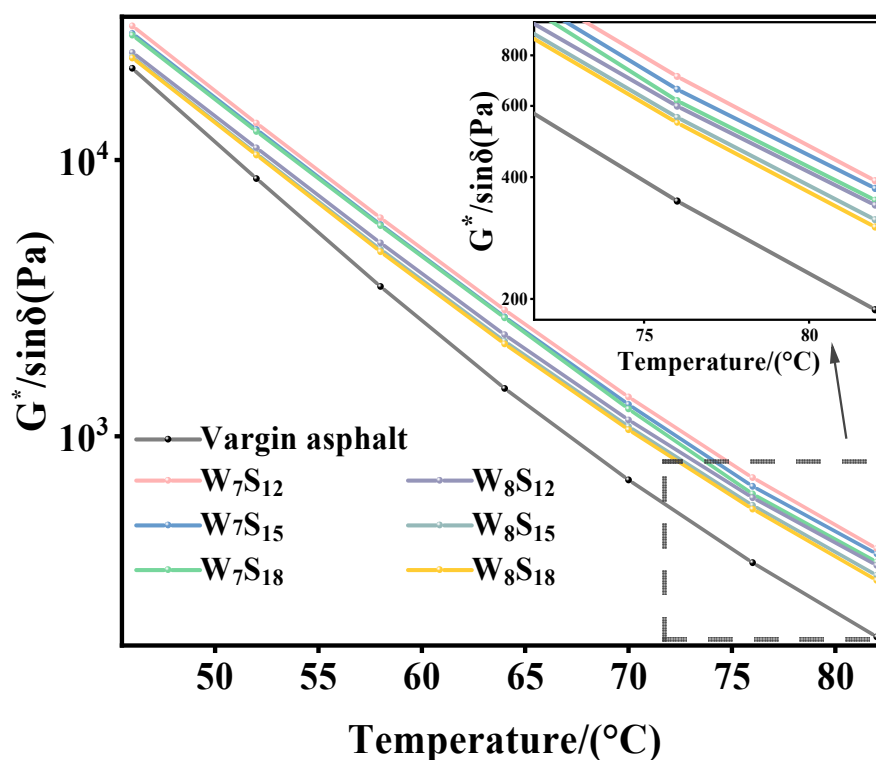


Figure 7. Rutting factor.

3.3. Complex Modulus Master Curves Analysis

Figure 8 presents the master curves of recycled asphalt across a broader frequency range, derived using the time-temperature equivalence principle. Based on this principle, high temperatures were associated with low frequencies, while low temperatures corresponded to high frequencies [57].

As illustrated in Figure 8, the distribution of the frequency curve in the low-frequency region appeared relatively sparse. This suggests a notable enhancement in the high-temperature stability of the recycled asphalt, with W_7S_{12} demonstrating the best performance under elevated temperatures. Nevertheless, with increasing frequency, the curve gradually approached the middle frequency, possibly due to the weak influence of SCG and WCO on the asphalt in the middle-temperature stage. In the high-frequency region, the curves progressively converged, with the G^* of 8% WCO showing greater

alignment. This suggests that the WCO content plays a critical role in influencing the low-temperature performance of asphalt. Based on the time–temperature equivalence principle, the low-frequency range corresponds to high-temperature performance, whereas the high-frequency range is indicative of low-temperature performance. A higher modulus in the high-temperature region indicates improved deformation resistance under such conditions. Conversely, a high modulus in the low-temperature region suggests an increased likelihood of brittle fracture in low-temperature environments. Furthermore, W₈S₁₈ demonstrated the best low-temperature performance, while W₇S₁₂ reached an optimal value at high frequencies, due to the influence of asphalt aging. The main curve continued to increase from low to high frequency, indicating an increase in asphalt deformation resistance with loading frequency. The G^* value in the low-frequency region was smaller than in the high-frequency region. This phenomenon could be attributed to the shorter stress time at a point acting on the asphalt as the loading frequency increased. During this period, the intermolecular forces within the asphalt could withstand the stress, resulting in reduced permanent deformation of the asphalt.

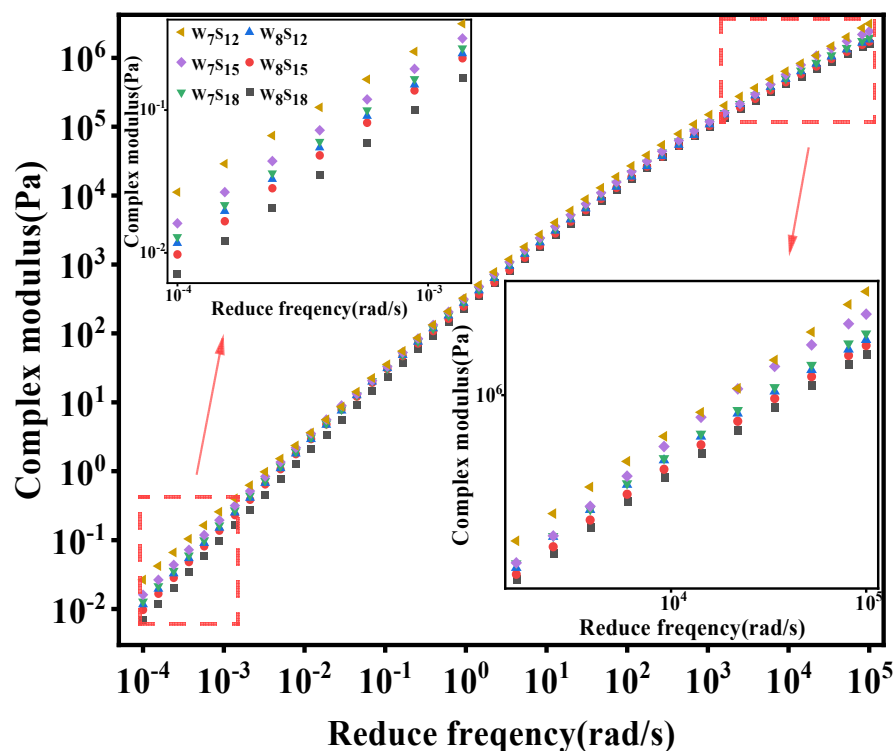


Figure 8. Recycled asphalt master curve.

3.4. Creep and Recovery Behavior

The MSCR test evaluated the rutting resistance of asphalt at elevated temperatures by measuring the elastic recovery rate (R%) and the non-recoverable creep compliance (J_{nr}) [58]. Certain tests showed that a higher R% value was associated with a lower J_{nr}.

As illustrated in Figure 9a, the R% value of recycled asphalt exceeded that of virgin asphalt, indicating that the addition of SCG and WCO has, to some extent, improved the deformation recovery capability of aged asphalt. Furthermore, the R% value for the 0.1 kPa stress level was greater than that of the 3.2 kPa stress level, which indicated that rutting deformation recovery was larger and the resulting rutting deformation became smaller for smaller vehicle loading during pavement travel. As shown in Figure 9b, J_{nr} at the 0.1 and 3.2 stress levels could also be used as an indicator of asphalt stiffness, and the stiffness increased as J_{nr} decreased, which improved rutting resistance. The results clearly show

that the J_{nr} value of recycled asphalt is lower than that of virgin asphalt. As the content of SCG increased, J_{nr} showed a gradual increase with a relatively flat trend, indicating minimal impact on the high-temperature performance with the addition of SCG. The J_{nr} value of the asphalt binder at a stress level of 3.2 kPa was higher than that at 0.1 kPa. This suggests that heavily loaded vehicles cause greater rutting deformation. At a dosage of W_7S_{12} , a maximum $R\%$ (0.1) and minimum J_{nr} (0.1) were observed, representing an increase of 228% and a decrease of 57.26%, respectively, compared to virgin asphalt. This finding aligned with the asphalt's performance under high-temperature conditions. The comprehensive tests demonstrated that SCG and WCO effectively enhanced the elastic properties of asphalt, specifically, with an increase in the elastic ratio and a decrease in the cumulative strain of the previous cycle during cyclic loading, leading to a higher final recovery rate.

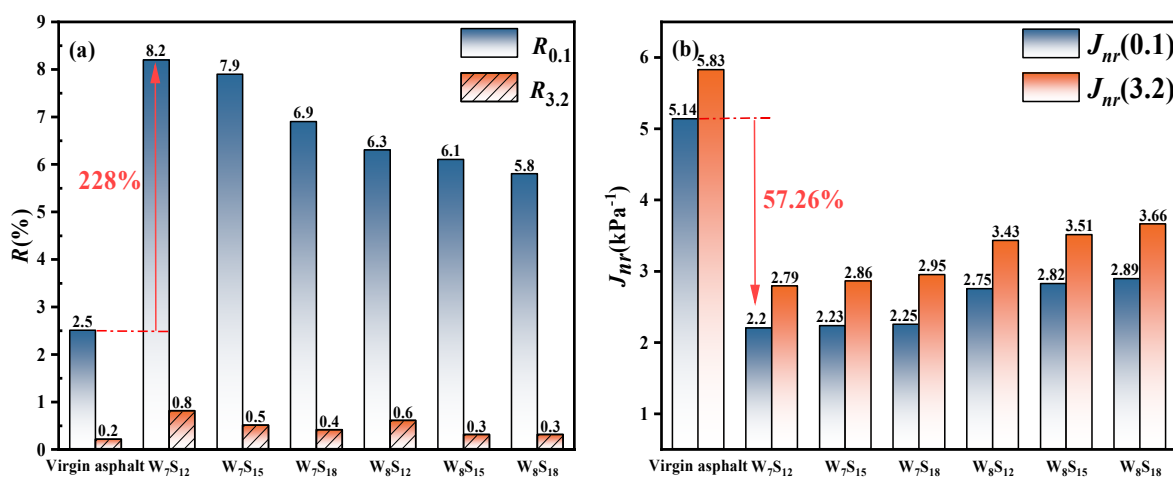


Figure 9. $R\%$ values, J_{nr} values for two stress levels: (a) $R\%$ versus doping; (b) J_{nr} versus doping.

3.5. Fatigue Resistance Performance

Figure 10a presents the projected fatigue life of recycled asphalts at a temperature of 25 °C, comparing strain levels of 2.5% and 5%. The recycled asphalt showed a longer fatigue life than virgin asphalt under both strain levels. This indicates that incorporating SCG and WCO may improve the flexibility of aged asphalt. This occurred because, during aging, asphalt lost its lighter components through volatilization. SCG and WCO supplemented these light components, softening the asphalt and enabling it to endure more load cycles, thereby enhancing its fatigue performance. When the strain level reached 2.5%, the N_f value of W_8S_{18} peaked, exhibiting a fatigue life 3.39 times longer than that of virgin asphalt, which highlights its superior fatigue performance under these conditions. Likewise, when the strain level was 5%, the N_f value of W_8S_{18} reached its peak, resulting in a fatigue life 2.34 times longer than that of virgin asphalt, demonstrating exceptional fatigue performance at this specific strain level. This was attributed to the heating process of SCG and asphalt, which formed a stable network structure. This structure could effectively absorb and disperse stress in the asphalt, thereby improving its deformation resistance. The fatigue life of asphalt significantly decreased when the strain was increased from 2.5% to 5%. Specifically, W_7S_{12} decreased by 85.21%; this phenomenon could be attributed to the susceptibility of the asphalt to damage under heavy loads. Because N_f at a 5% strain level provided a more accurate indication of actual fatigue damage to the pavement, at this level, W_8S_{18} demonstrated the highest N_f , validating its potential for use in pavement applications. Overall, W_8S_{18} demonstrated satisfactory fatigue performance at both strains.

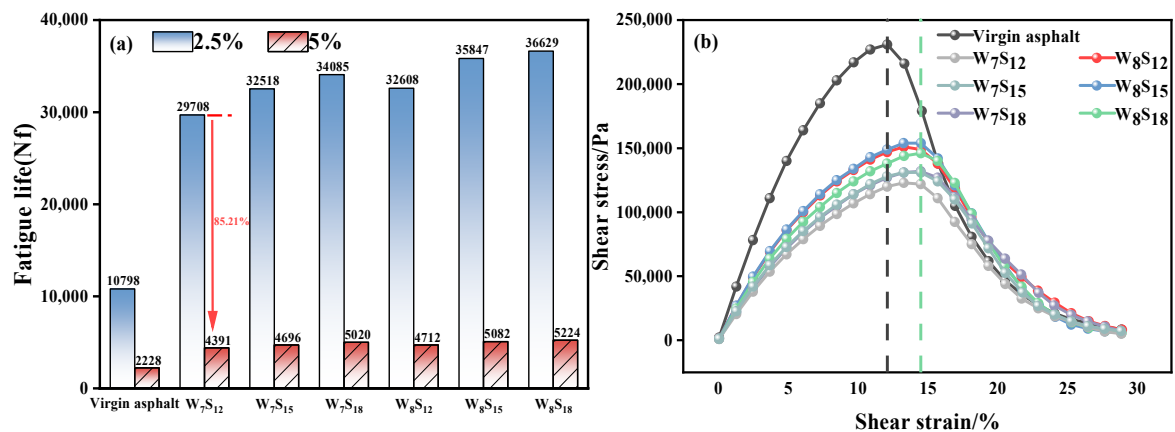


Figure 10. Fatigue life, stress–strain relationship at two strains: (a) fatigue life of recycled asphalt; (b) stress–strain relationship.

Figure 10b illustrates the variation curve depicting the relationship between asphalt shear stress and shear strain at various dosage levels. As the strain increased, the asphalt shear stress gradually rose to its peak, after which it declined and eventually reached the yield point. The recycled asphalt was supplemented with light fractions, resulting in a softer asphalt, and consequently, a decrease in the shear stress. The higher the yield stress of the material, the greater its resistance to deformation; similarly, a higher yield strain indicated a greater ability to resist load and deformation, reflecting the superior elasticity of the material. Virgin asphalt demonstrated the greatest resistance to deformation, along with the lowest resistance to load-induced deformation and minimal resilience. The recycled asphalt exhibited a lower yield stress compared to virgin asphalt, but its yield strain was higher. This indicated increased resistance to load deformation, suggesting superior elastic properties of the material. Under the same dosage of WCO, the yield strain gradually increased with increasing SCG content. The maximum value was reached at a WCO dosage of W₈S₁₈, indicating that this combination resulted in the best elastic properties for recycled asphalt. This indicates that incorporating WCO and SCG improved the resistance to mid-temperature deformation as well as the extent of deformation in recycled asphalt.

Table 5 presents the parameters of the fatigue equations for asphalt binders at various dosages, calculated using the VECD model and the fatigue prediction method. Parameter A represents the material's mechanical properties in the online viscoelastic region, which correlates with the energy storage modulus ($G^* \cdot \cos \delta$) of the asphalt binder [59]. This parameter decreased as the energy storage modulus decreased during cyclic loading, and has been commonly used to represent the integrity of the material when subjected to cumulative damage during loading. The parameter B represents the rate of decay of the strength of the material during the loading process. A larger value of B indicated a reduced capacity of the material to resist fatigue damage and sustain structural integrity under cyclic loading conditions. Consequently, as the amplitude of the strain level increased, the fatigue life of the asphalt decreased more rapidly [60]. The gradual rise in A and B with higher dosages of WCO and SCG indicated that the recycled asphalt demonstrated improved material integrity when subjected to cyclic loading. Conversely, the fatigue life diminished at a faster rate with increasing strain level amplitude.

Table 5. Fatigue equation parameters.

Asphalt Binder	Virgin Asphalt	W ₇ S ₁₂	W ₇ S ₁₅	W ₇ S ₁₈	W ₈ S ₁₂	W ₈ S ₁₅	W ₈ S ₁₈
A	87,000	371,936	419,866	420,585	428,748	457,248	498,590
B	2.28	2.76	2.76	2.78	2.79	2.79	2.85

3.6. Glover–Rowe Parameter

Figure 11 shows the black space diagram for recycled asphalt, aged asphalt, and virgin asphalt. Asphalt pavements tend to crack when the G–R parameter reaches or exceeds 180 kPa. However, when this parameter surpasses 450 kPa, severe cracking becomes evident [61]. At this point, necessary measures would need to be implemented for maintenance. The condition of the aged asphalt was between initial cracking and significant cracking; this occurs because light components, including saturated and aromatic phenols, are converted into additional asphaltenes during the asphalt aging process. Consequently, the proportion of hard components in asphalt increases, causing the pavement to become rigid and brittle. The addition of the rejuvenator caused the G–R parameter of the recycled asphalt, as shown in the figure, to decrease progressively from the upper left to the lower right, gradually approaching the properties of virgin asphalt. This suggests that the recycled asphalt exhibited enhanced resistance to low-temperature cracking, thereby decreasing the probability of cracking during this phase. However, this enhancement did not achieve the performance level of virgin asphalt. In recycled asphalt, particularly W_8S_{18} , the addition of SCG and WCO led to the lowest relative G–R parameter and the highest δ value. This demonstrates a complete recovery of the asphalt's low-temperature characteristics and viscous components, resulting in optimal performance at low temperatures. These findings were consistent with the asphalt master curve.

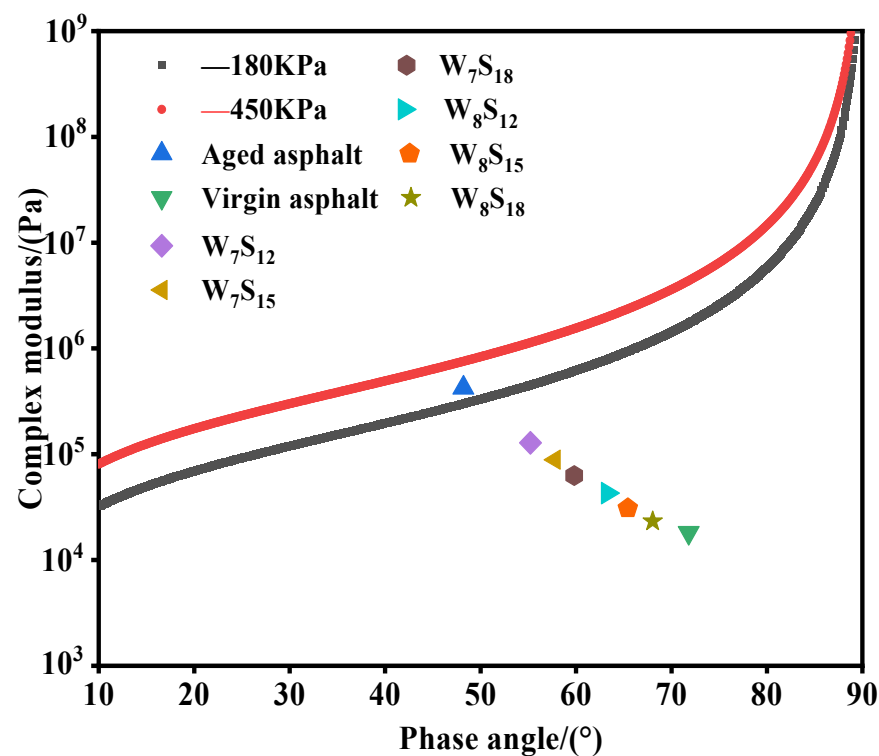


Figure 11. Black space diagram.

3.7. Scanning Electron Microscopy Analysis

Figure 12 presents the SEM images of virgin asphalt, aged asphalt, and recycled asphalt. As shown in Figure 12a, the microstructure of aged asphalt exhibits severe damage. The red circles in the figure illustrated increased wrinkles, prominent cracks, and interface delamination. These features indicate that the uniform phase of the asphalt has been disrupted due to the partial loss of light components, resulting in brittleness and structural deterioration. Figure 12b and 12c depict the microstructures of W_7S_{12} and W_8S_{18} recycled asphalt, respectively. The incorporation of SCG and WCO significantly reduces the cracks

present in aged asphalt. Although the red circles in Figure 12b illustrate W_7S_{12} retains some minor wrinkles, W_8S_{18} exhibits a smoother and more uniform surface, demonstrating that the additives effectively restore the lost light components. Figure 12d highlights the microstructure of W_7S_{12} recycled asphalt at another location, where fewer cracks and slight wrinkles are visible, aligning with observations from Figure 12b. At a $3000\times$ magnification, Figure 12e focuses on point A in Figure 12d, showing the uniform distribution of SCG and WCO within the asphalt. The particles visible of red circles in the microstructure are likely the result of finer SCG particles absorbing oil during the shear blending process. This minor agglomeration does not weaken the integrity of the asphalt system; instead, it enhances overall stability. The irregular, rough, and porous surface of SCG contributes significantly to forming a stable structure when combined with WCO and asphalt. This synergistic interaction improves the asphalt's structural stability, increases adhesiveness, enhances resistance to fatigue cracking, and boosts the self-healing ability of microcracks.

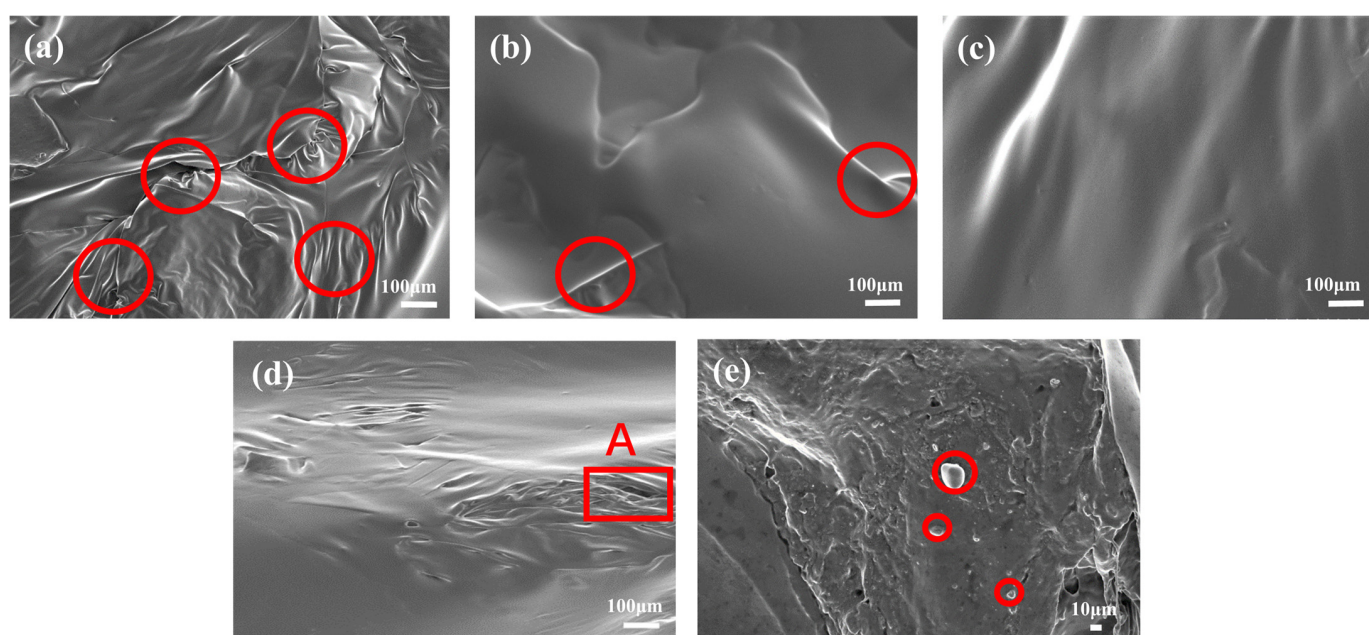


Figure 12. SEM images of various asphalts: (a) aged asphalt ($1000\times$); (b) W_7S_{12} ($1000\times$); (c) W_8S_{18} ($1000\times$); (d) W_7S_{12} ($1000\times$); (e) W_7S_{12} ($3000\times$).

3.8. FTIR Analysis

Figure 13a presents the infrared spectra of SCG and WCO individually. It can be seen that the two have the same chemical bond characteristic peaks and similar wavelengths. For WCO, the bands at 2923 cm^{-1} , 2855 cm^{-1} and 1747 cm^{-1} , which are the characteristic peaks for C–H stretching vibrations, N–H stretching vibrations, and C=O stretching vibrations, respectively, are derived from the carbonyl group in the sample. The bands at 1652 cm^{-1} and 1458 cm^{-1} correspond to a C=C stretching vibration and C–OH stretching vibration, respectively [62]; this came from the benzene ring and the sample single bond skeleton. The bands at 1372 cm^{-1} and 1165 cm^{-1} were associated with C–C asymmetric stretching and C–H in-plane deformation vibrations. These features originate from the carboxyl groups present in the sample. The spectra of SCG exhibited similarities to those of WCO. A distinct peak at 1749 cm^{-1} , corresponding to C=O stretching vibrations, was identified in SCG, suggesting the presence of oily components and unsaturated aliphatic hydrocarbons.

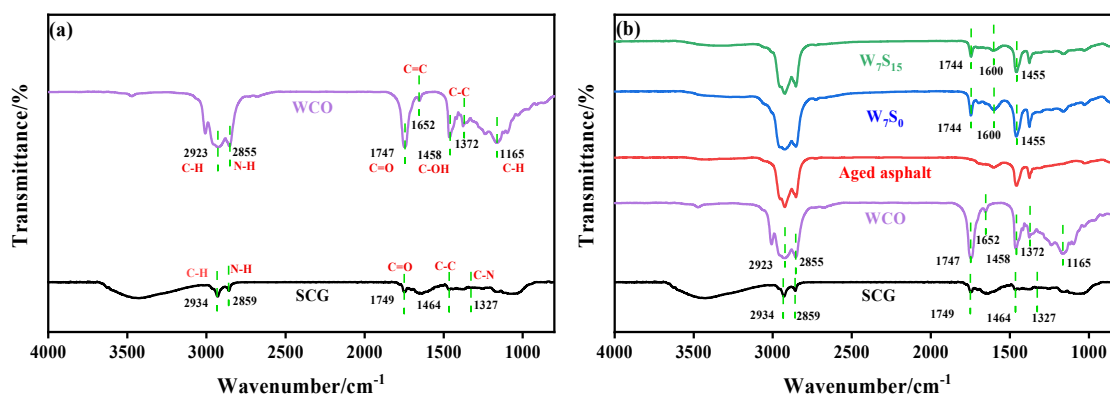


Figure 13. FTIR analysis: (a) SCG and WCO; (b) recycled asphalt.

The FTIR spectrum of SCG, WCO, aged asphalt, W₇S₀ recycled asphalt, and W₇S₁₅ recycled asphalt are shown in Figure 13b. In the case of W₇S₀ recycled asphalt, the incorporation of WCO into aged asphalt resulted in the enhancement and blue-shifting of characteristic peaks. The C=O, C=C, and C-OH stretching vibrations, originally observed at 1747 cm⁻¹, 1652 cm⁻¹, and 1458 cm⁻¹, shifted to 1744 cm⁻¹, 1600 cm⁻¹, and 1455 cm⁻¹, respectively. This phenomenon may result from the binding of these groups to the hydroxyl group, where the bonding energy is increased due to the interaction between WCO and aged asphalt under heating conditions. Consequently, the thermodynamic properties of the aged asphalt are improved. Additionally, a reduction in the intensity of peaks within the range of 1400 cm⁻¹ to 1000 cm⁻¹ was observed following the addition of WCO. This suggests that WCO exhibits volatility and instability under heating conditions. A comparison of the spectra for WCO, aged asphalt, and W₇S₀ recycled asphalt revealed that the absorption peaks of the recycled asphalt were essentially a superposition of those observed in the spectra of WCO and aged asphalt. This observation indicates that the regeneration process involving WCO and aged asphalt was primarily a physical blending. For W₇S₁₅ recycled asphalt, the addition of WCO and SCG enhanced the characteristic peaks at 1747 cm⁻¹, 1652 cm⁻¹, and 1458 cm⁻¹, corresponding to C=O, C=C, and C-OH stretching vibrations, respectively. These peaks were blue-shifted to 1744 cm⁻¹, 1600 cm⁻¹, and 1455 cm⁻¹, respectively. This shift suggests that SCG may, like WCO, increase the bonding energy of the hydroxyl group, thereby strengthening the molecular interactions between asphalt, WCO, and SCG. A comparison of the spectra of SCG, W₇S₀ recycled asphalt, and W₇S₁₅ recycled asphalt showed no difference in the adsorption peaks and no new adsorption peak generation. The spectrum of W₇S₀ recycled asphalt has been demonstrated to be essentially a superposition of the spectra of WCO and aged asphalt. This finding further confirms that the blending process of WCO and SCG in recycled asphalt is primarily a physical modification.

4. Conclusions

Recycled asphalt was produced by incorporating SCG and WCO into aged asphalt. To evaluate the asphalt's performance, conventional index tests, rheological analyses, infrared spectroscopy, and scanning electron microscopy were performed. The results were analyzed, leading to the following conclusions.

- (1) With the addition of SCG and WCO, the penetration of aged asphalt increases and the softening point decreases, which has the same effect as the addition of virgin asphalt, and restores the aged asphalt to the level of virgin asphalt at all dosages.
- (2) The rheological experiment results show that the complex shear modulus, rutting factor and elastic recoverable rate of recycled asphalt with each content are higher than

those of virgin asphalt, indicating that the porous structure of SCG and WCO combined with aged asphalt can adsorb oily substances, which makes the recycled asphalt have better high-temperature performance. W₇S₁₂ had the best high-temperature performance, improved by 90% compared to virgin asphalt.

- (3) The recycled asphalt was compared and analyzed by linear amplitude sweeping test. With the addition of SCG and WCO, which can provide light fractions, soften asphalt, and improve the flexibility of aged asphalt, the results show that SCG and WCO help improve the fatigue life of aged asphalt. At a strain level of 2.5%, the fatigue life of W₈S₁₈ recycled asphalt is 3.39 times higher compared to virgin asphalt.
- (4) According to the results of the G–R parameter analysis, the low-temperature cracking resistance of recycled asphalt can be recovered with the incorporation of SCG and WCO, approaching the level of virgin asphalt.
- (5) SEM analysis demonstrated that incorporating SCG and WCO into aged asphalt enhances its properties, resulting in a smoother surface. The irregular, rough, and porous surface characteristics of SCG contribute to forming a more stable structure between SCG, WCO, and the asphalt. FTIR analysis revealed that the infrared spectra of SCG, WCO, and aged asphalt were all a simple superposition of the two, indicating that the recycled asphalt experienced physical modification.

In this study, SCG and WCO were used as innovative rejuvenators to investigate the properties of aged asphalt and its rejuvenation mechanism. However, this study still needs to be improved. The performance of recycled asphalt may vary depending on the sources of SCG and WCO. Additionally, future studies should consider the storage conditions of SCG and WCO. This paper did not examine the application effects or pavement performance of SCG and WCO recycled asphalt mixtures. Future research could investigate how SCG and WCO originating from various production regions affect the performance of recycled asphalt and its mixtures. Additionally, it was possible to investigate anti-aging performance using the antioxidant components found in SCG.

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