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Effect of Thermal Aging on the Self-Healing Properties of High-Content Crumb Rubber–SBS Composite Modified Asphalt

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Abstract

High-content crumb rubber composite modified asphalt (HRCMA), which combines crumb rubber with a styrene–butadiene–styrene (SBS) modifier, offers excellent mechanical performance and environmental benefits, yet its high viscosity requires elevated production and construction temperatures that may accelerate thermo-oxidative aging and impair its durability and self-healing capability. In this study, the self-healing behavior of HRCMA under virgin, Thin Film Oven Test (TFOT)-aged, and Pressure Aging Vessel (PAV)-aged conditions was systematically investigated and compared with that of conventional SBS modified asphalt (SBSMA) using fatigue–healing–fatigue tests. The effects of damage degree, healing interval time, and healing temperature were evaluated, and a comprehensive self-healing index (HI) integrating the recovery of initial mechanical properties and the recovery of damage evolution characteristics was proposed. The results show that increasing the damage degree significantly reduces the self-healing capability of both binders, and thermal aging further intensifies this deterioration; two-way analysis of variance (ANOVA) confirmed that these effects are statistically significant. Extending the healing interval time and raising the healing temperature improve the self-healing performance, although the enhancement weakens after severe aging owing to the reduction in molecular mobility. Compared with SBSMA, HRCMA exhibits higher self-healing capability under all aging conditions, indicating stronger resistance to aging-induced damage. The proposed HI provides a more comprehensive evaluation of asphalt self-healing behavior, and the findings provide theoretical support for the durability assessment and engineering application of highly modified asphalt materials.

Keywords: high-content crumb rubber composite modified asphalt (HRCMA); self-healing properties; aging; initial state; damage rate; sustainable development

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1. Introduction

Asphalt pavement is one of the most widely used pavement structures for high-grade highways due to its excellent riding comfort, convenient maintenance, and economic advantages. However, during long-term service, asphalt materials are inevitably subjected to complex environmental and mechanical conditions, including repeated traffic loading, temperature variation, and oxidative aging. These factors continuously alter the chemical composition, rheological properties, and internal structure of asphalt binders, resulting in the initiation and accumulation of microdamage. With the development of irreversible damage, microcracks gradually propagate and eventually cause macrocracking, which is one of the main factors limiting pavement service life [1]. Therefore, improving the durability and damage resistance of asphalt materials has become an important research objective in pavement engineering.

Among various modified asphalt binders, high-content crumb rubber composite modified asphalt (HRCMA) has attracted increasing attention because it combines the advantages of crumb rubber modified asphalt and styrene–butadiene–styrene (SBS) modified asphalt, as systematically reviewed for recycled tire rubber modified bitumens [2]. The incorporation of high-content crumb rubber improves the elasticity, rutting resistance, and fatigue resistance of asphalt binders through rubber swelling and the interaction between rubber particles and asphalt components, while simultaneously promoting the high-value utilization of waste tires [3]. Ji et al. [4] systematically evaluated the chemical and rheological aging behavior of high-content crumb rubber asphalt binders, and Guo et al. [5] demonstrated that high-content rubber powder-modified asphalt mixtures retain satisfactory low-temperature crack resistance under freeze–thaw cycles. To balance the modification effects and the processing performance, an SBS modifier is often incorporated together with crumb rubber to form composite modified asphalt, in which the SBS polymer network improves the storage stability and structural uniformity of the binder [6,7]. Previous studies on HRCMA have mainly focused on preparation technology, rheological properties, aging resistance, and pavement performance [3–5]. However, although thermal aging has been confirmed to significantly alter the chemical composition and rheological characteristics of asphalt binders and thereby influence their fatigue performance [4], its influence on the self-healing behavior of HRCMA remains insufficiently understood. In addition, the thermo-oxidative aging behavior of high-viscosity modified asphalt binders has been characterized by infrared spectroscopy [8] and under coupled heat–light–water conditions [9], further confirming that the oxidation-induced evolution of the chemical structure and rheological properties largely governs the long-term performance of highly modified binders.

Fatigue damage is one of the primary deterioration mechanisms affecting the long-term performance of asphalt pavements. Under repeated traffic loading, asphalt binders experience continuous energy dissipation, molecular rearrangement, and structural degradation, resulting in damage accumulation and crack initiation. Unlike conventional damage accumulation processes, asphalt binders possess an intrinsic self-healing capability, allowing partial recovery of fatigue-induced damage. Little and Bhasin [10] systematically discussed the healing mechanisms of asphalt materials and attributed the healing process to the re-adhesion of crack faces and the diffusion of asphalt molecules across the crack interface. Molecular dynamics simulations further revealed that the self-healing potential of asphalt binders is closely related to the molecular diffusion rate and the intermolecular interactions of asphalt components [11]. García [12] demonstrated that capillary flow driven by the pressure difference at crack tips contributes to the closure of open cracks in asphalt mastic. This inherent healing behavior can delay crack propagation, improve fatigue resistance, and extend pavement service life [13]. Therefore, understanding the self-healing mechanism of asphalt binders is important for revealing the evolution process of fatigue damage and developing more durable pavement materials.

To better understand the self-healing mechanism of asphalt binders, extensive studies have investigated the influencing factors and evaluation methods of asphalt self-healing behavior. Daniel and Kim [14] conducted laboratory fatigue–rest–fatigue tests on asphalt mixtures and quantified the recovery of dissipated energy during rest periods, while Carpenter and Shen [15] evaluated healing in hot-mix asphalt using a dissipated energy approach and demonstrated that the healing effect strongly depends on the strain level and the rest period length. At the binder level, Shen et al. [16] characterized the fatigue and healing behavior of asphalt binders using repeated loading tests, and Qiu et al. [17] evaluated the self-healing capability of bituminous mastics with a modified direct tension test. Qiu et al. [18] further divided the healing process into crack closure and strength recovery stages, providing a multi-scale understanding of asphalt healing behavior. With respect to evaluation indices, Luo et al. [19] improved the conventional healing index by considering the influence of loading history, Tang et al. [20] established a healing evaluation method based on energy dissipation theory, Xu and Wang [21] investigated the influence of the healing interval on fatigue recovery, and Miglietta et al. [22] introduced a time–temperature superposition approach to evaluate the self-healing behavior of bituminous binders. In addition, Chen et al. [23] comparatively analyzed the self-healing evaluation indices of asphalt binders based on dynamic shear rheometer (DSR) tests. Previous studies have demonstrated that asphalt self-healing behavior is affected by multiple factors, including damage degree, healing interval time, healing temperature, loading history, aging condition, and binder composition [13,23]. At the microscale, nanoscale characterization has confirmed that molecular diffusion and the free volume fraction play important roles in the self-healing process at different temperatures [24].

In addition to the intrinsic healing capability of asphalt materials, various engineering strategies have been developed to actively promote the self-healing process. Induction heating has been applied to electrically conductive asphalt mastic and mixtures to accelerate crack closure by elevating the binder temperature [25], and nanomaterials such as pretreated graphene have been used to improve the self-healing behavior of asphalt mixtures [26]. Microcapsules containing rejuvenator have been designed to release healing agents upon crack rupture, thereby restoring the performance of fresh and aged bitumen [27,28]. Thermally activated healing tests have further demonstrated that elevating the temperature can substantially recover the fatigue damage of asphalt binders, and that the required healing temperature is closely related to the softening point of the binder [29]. Moreover, studies on the induced heating–healing of asphalt mixtures have shown that the healing efficiency decreases significantly with increasing aging level [30]. These studies confirm that the self-healing capability of asphalt materials is highly temperature- and aging-dependent. However, most of the above strategies require additional materials or external energy input, whereas improving the intrinsic healing capacity and aging resistance of the binder itself remains the most economical route to durable asphalt pavements. Therefore, clarifying how thermal aging affects the intrinsic self-healing behavior of highly modified asphalt binders is of fundamental importance for the design of durable and self-healing asphalt materials, which is the focus of this study.

Although these studies have significantly advanced the understanding of asphalt self-healing behavior, existing evaluation indices still exhibit certain limitations. Most commonly used indices mainly focus on the recovery of mechanical properties, such as modulus or stiffness recovery [19,20], while neglecting the influence of loading history and damage evolution characteristics. Conversely, indices considering loading history effects can better reflect the damage accumulation and recovery process but may not fully describe the restoration of the initial mechanical state [21,22]. Furthermore, conventional single-parameter indices cannot simultaneously characterize the two recovery mechanisms, which becomes particularly problematic under complex aging conditions where material degradation and damage recovery occur simultaneously [23]. A comprehensive evaluation method integrating both mechanical property recovery and loading history characteristics is therefore required to provide a more accurate assessment of asphalt self-healing capability.

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Accordingly, this study systematically investigates the effects of thermal aging on the self-healing behavior of HRCMA by comparing it with conventional SBS modified asphalt under virgin, Thin Film Oven Test (TFOT)-aged, and Pressure Aging Vessel (PAV)-aged conditions. Fatigue–healing–fatigue tests were conducted using a dynamic shear rheometer under different damage degrees, healing interval times, and healing temperatures. Furthermore, a comprehensive self-healing index integrating both the recovery of the initial mechanical state and the recovery of loading history characteristics was proposed to overcome the limitations of existing evaluation indices, thereby providing a more comprehensive assessment of asphalt self-healing behavior under thermal aging conditions. This study is expected to deepen the understanding of the thermal aging-induced healing mechanism of HRCMA and provide a theoretical basis for the durability evaluation and engineering application of high-content crumb rubber composite modified asphalt.

2. Materials and Methods

2.1. Raw Materials

High-content crumb rubber composite modified asphalt (HRCMA) and styrene–butadiene–styrene modified asphalt (SBSMA) were selected as representative modified asphalt binders to investigate the influence of thermal aging on asphalt self-healing performance. HRCMA was prepared by incorporating high-content crumb rubber and an SBS modifier into the asphalt binder, consisting of 25% crumb rubber and 4% SBS modifier by mass. SBSMA containing 5% SBS modifier was selected as the reference binder for comparison. The base asphalt, crumb rubber, and SBS modifier used in this study were commercial products obtained from local suppliers in Nanjing, China, and were used as received.

The selection of HRCMA and SBSMA was based on their different modification mechanisms and engineering applications. SBS modification mainly improves elasticity, deformation resistance, and recovery capability of asphalt binders through the formation of a polymer network structure. In comparison, HRCMA combines the reinforcing effect of SBS with the elastic recovery and energy dissipation characteristics of crumb rubber. The swelling of crumb rubber particles and their interaction with asphalt components can promote the formation of a flexible rubber–asphalt network, while SBS contributes to structural stability through polymer cross-linking. Therefore, the synergistic modification effect of crumb rubber and SBS is expected to enhance the fatigue resistance, damage recovery capability, and durability of asphalt binders [6,7]. The selected HRCMA formulation (25% crumb rubber and 4% SBS) was determined based on previous studies and engineering considerations [3,4]. A crumb rubber content of 25% by mass was adopted because previous studies have demonstrated that medium-/high-content crumb rubber asphalt exhibits enhanced high-temperature stability, elastic recovery, and aging resistance compared with conventional rubberized asphalt [3], while the SBS dosage of 4% was selected to ensure the formation of a continuous polymer network and adequate storage stability of the composite binder [6,7]. SBSMA containing 5% SBS modifier was selected as the control binder because this dosage represents a widely used polymer-modified asphalt formulation in pavement engineering. It should be noted that HRCMA and SBSMA differ in both crumb rubber content (25% versus 0%) and SBS content (4% versus 5%), which reflects their typical engineering formulations. Therefore, the comparison between the two binders represents the combined effect of crumb rubber incorporation and SBS dosage variation under practical conditions, rather than the isolated contribution of crumb rubber alone; this point is further discussed in Section 4.1. Nevertheless, the comparison provides an opportunity to clarify the improvement in the self-healing behavior of modified asphalt binders achieved by high-content crumb rubber incorporation under thermal aging conditions.

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The basic physical properties of the two asphalt binders were evaluated according to the relevant specifications, and the test results are presented in Table 1. As shown in Table 1, HRCMA exhibits a higher softening point and elastic recovery compared with SBSMA, indicating enhanced high-temperature stability and elastic recovery capability. These characteristics provide a basis for further investigating the influence of thermal aging on the self-healing performance of HRCMA.

Table 1. Basic properties test results of asphalt.

Technical Indicators	Test Results		Test Method [31]
	SBSMA	HRCMA	
Penetration (25 °C, 100g, 5 s) (0.1 mm)	46.8	51.6	T0604
Ductility (5 °C, 5 cm/min) (cm)	33.5	36.2	T0605
Softening point (°C)	83.5	89.0	T0606
Viscosity (Pa·s)	2.8 (135 °C)	2.2 (175 °C)	T0625
Elastic Recovery Rate (%)	89.0	99.0	T0662

2.2. Preparation of HRCMA Binder

HRCMA binder was prepared through a composite modification process using crumb rubber and an SBS modifier. The base asphalt was first heated to achieve sufficient fluidity, followed by the incorporation of the SBS modifier to promote the formation of a polymer-modified asphalt structure. Subsequently, high-content crumb rubber was gradually added to the asphalt binder under continuous mixing conditions. The modified binder was further subjected to sufficient shearing and swelling treatment to facilitate the interaction between crumb rubber particles and asphalt components. Through the synergistic effect of crumb rubber swelling and SBS polymer networking, a rubber–SBS composite modified asphalt binder was obtained [7]. The prepared HRCMA binder was then used for subsequent thermal aging treatment and self-healing performance evaluation.

2.3. Asphalt Aging Procedure

Thermal aging is one of the primary factors affecting the chemical composition, rheological properties, and self-healing capability of asphalt binders. During long-term pavement service, asphalt binders undergo oxidation, volatilization of lightweight components, and structural degradation, which subsequently influence their fatigue damage evolution and healing capability. Therefore, both short-term and long-term aging procedures were employed in this study to investigate the evolution of the self-healing behavior of HRCMA and SBSMA under different aging conditions. The unaged, TFOT-aged, and PAV-aged binders were selected to represent different stages of asphalt aging, enabling a comprehensive evaluation of the influence of thermal aging on self-healing performance.

2.3.1. Thin Film Oven Test (TFOT)

The short-term aging procedure was conducted according to JTG E20-2011 (T0609) [31]. TFOT was adopted to simulate the aging of asphalt binders during storage, transportation, mixing, and paving processes, where oxidation and volatilization of lightweight components occur under high-temperature exposure.

Before aging, the asphalt binders were heated until reaching a sufficiently fluid state and then poured into TFOT containers. Approximately 50 g of asphalt binder was placed in each container to form a thin asphalt film. The samples were subsequently aged at 163 °C under continuous rotation conditions to ensure uniform exposure to heat and air.

After completion of the TFOT procedure, the aged binders were collected and stored for subsequent self-healing performance evaluation.

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2.3.2. Pressure Aging Vessel (PAV)

Long-term aging was simulated using the Pressure Aging Vessel (PAV) method. Compared with TFOT, PAV provides a more severe aging environment by applying elevated temperature and pressure, thereby accelerating oxidative aging and reproducing the degradation process of asphalt binders during long-term pavement service.

In this study, the PAV aging procedure was performed at 100 °C ± 0.5 °C under an air pressure of 2.1 ± 0.1 MPa for 20 h according to the corresponding specification [31]. After completion of aging, the pressure was gradually released, and the aged asphalt binders were collected for subsequent self-healing tests.

2.4. Fatigue–Healing–Fatigue Test

During pavement service, asphalt binders experience repeated traffic loading accompanied by intermittent unloading periods. These rest periods provide opportunities for the recovery of fatigue-induced damage through the intrinsic self-healing capability of asphalt materials. Therefore, a fatigue–healing–fatigue (F-H-F) test procedure was adopted in this study to simulate the damage accumulation and recovery process of asphalt binders under intermittent traffic loading conditions.

Fatigue–healing–fatigue tests were conducted using a dynamic shear rheometer (DSR) under a stress-controlled mode. The DSR equipment used in this study was a modular compact rheometer (MCR-102, Anton Paar, Graz, Austria). The test procedure consisted of three stages: initial fatigue loading, healing process, and secondary fatigue loading. During the initial fatigue stage, cyclic loading was applied to induce damage in asphalt binder specimens until the predetermined damage level was reached. Subsequently, the loading was interrupted, and the damaged specimens were allowed to recover under controlled conditions during the healing stage. After the designated healing period, secondary fatigue loading was performed under identical conditions to evaluate the recovery capability of asphalt binders [31].

All fatigue–healing–fatigue tests were conducted using an 8 mm diameter parallel-plate geometry with a 2 mm gap, which is recommended for asphalt binders at intermediate test temperatures. The test temperature was 25 °C, and the loading frequency was 10 Hz, corresponding to the loading frequency applied to pavement under vehicle speeds of 60–65 km/h. Before testing, the specimen was placed on the lower plate, pressed to the target gap, trimmed, and then conditioned at 25 °C for 20 min to eliminate the influence of temperature fluctuation on the test results. The linear viscoelastic (LVE) range of each binder was determined through preliminary strain sweep tests. Suitable loading stress levels for each binder and aging condition were determined through preliminary time scan tests; the applied stress levels adopted in this study (100–150 kPa for the virgin and TFOT-aged binders and 220 kPa for the PAV-aged binders) were verified to ensure that the initial strain response of each binder remained within its LVE range at the beginning of loading.

The self-healing performance was evaluated based on the recovery of dynamic shear modulus and the variation in fatigue resistance before and after the healing process. To improve the reliability and repeatability of the test results, preliminary investigations were performed to determine the key testing parameters, including the number of healing intervals, loading mode, healing interval time, and loading stress level. The determination procedures for these parameters are described in the following sections.

Three replicate specimens were prepared for each testing condition to ensure the reliability of the experimental results. The average values were used for analysis, and the standard deviations were calculated to represent the experimental variability. Error bars were added to the corresponding figures to illustrate the dispersion of the measured results.

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2.4.1. Determination of Healing Interval Number

In practical pavement conditions, asphalt materials may experience multiple loading and unloading cycles. However, excessive intermittent loading cycles may introduce additional damage accumulation and complicate the evaluation of intrinsic self-healing behavior. Therefore, the influence of different numbers of healing intervals on the modulus evolution of asphalt binders was investigated.

As shown in Figure 1, the dynamic shear modulus curve after the first healing interval exhibited a significant difference compared with that of the initial loading stage, indicating obvious damage recovery after the first unloading period. In contrast, the modulus evolution curves after subsequent intervals showed relatively limited changes. This result suggests that the first healing interval contributes the majority of the measurable recovery effect, whereas additional intervals provide limited improvement. Therefore, a single healing interval was selected in this study to balance experimental efficiency and the accurate characterization of asphalt self-healing behavior.

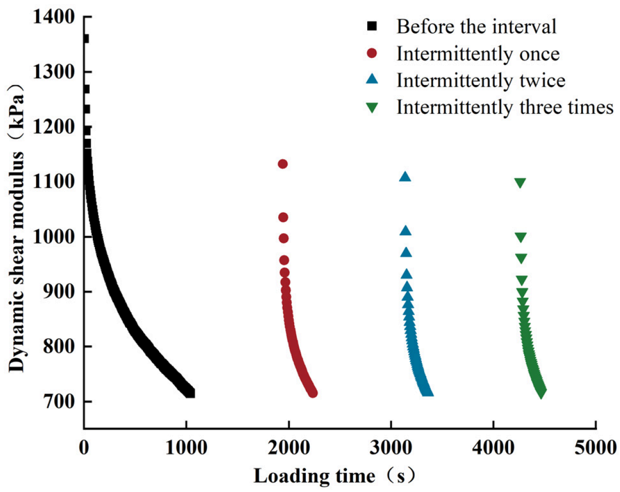


Figure 1. Schematic diagram of multiple intermittent tests.

2.4.2. Loading Mode

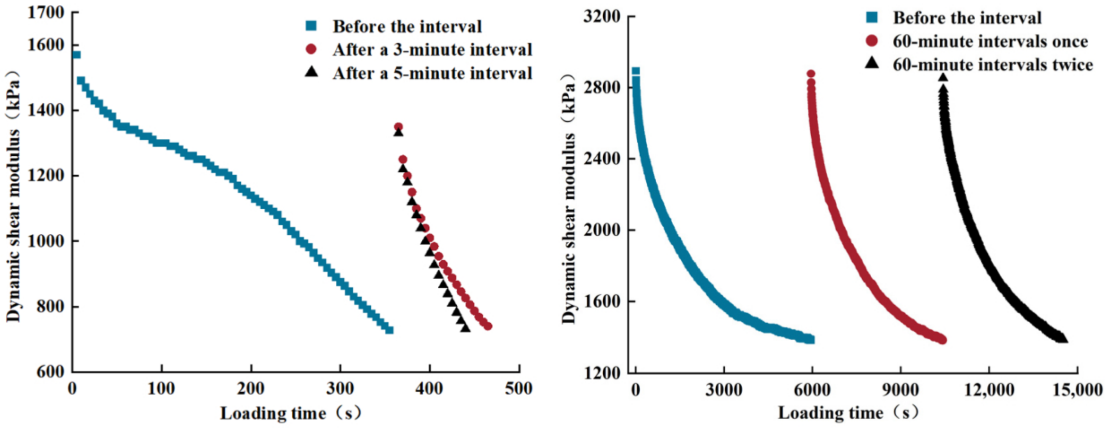
Time scan fatigue tests of asphalt binders can generally be conducted under stress-controlled or strain-controlled loading modes. Under stress-controlled conditions, the applied stress remains constant while the strain increases continuously with fatigue damage development. Under strain-controlled conditions, the applied strain remains constant while the stress decreases as the material deteriorates.

Preliminary comparisons showed that strain-controlled loading within the linear viscoelastic range resulted in excessively long testing durations because the reduction in stress was relatively slow during the early fatigue stage. In comparison, the stress-controlled mode provided a more efficient damage development process while maintaining the ability to capture modulus evolution during fatigue and healing. Therefore, the stress-controlled mode was selected for the self-healing tests in this study.

2.4.3. Selection of Healing Interval Time

The healing interval time is an important factor affecting the evaluation of asphalt self-healing capability. To determine suitable healing durations, preliminary tests were conducted using five interval times: 3 min, 5 min, 15 min, 30 min, and 60 min.

The variation in dynamic shear modulus recovery and fatigue performance after different healing intervals is shown in Figure 2. The results indicate that the recovery behavior after 3 min was similar to that after 5 min, suggesting that extending the healing duration from 3 min to 5 min produced limited additional recovery. Therefore, 3 min was selected as the shortest healing interval to capture the early-stage self-healing behavior.



(**a**) 3 min interval and 5 min interval (**b**) 60 min interval

Figure 2. Schematic diagram of self-healing test results after different interval times.

Furthermore, when the healing interval reached 60 min, the dynamic shear modulus approached a relatively stable recovery level, and a further extension of the healing time resulted in a negligible improvement. This indicates that approximately 60 min represents the effective upper limit of the healing process under the selected experimental conditions. Considering both experimental efficiency and the ability to distinguish different healing behaviors, healing intervals of 3 min, 15 min, and 30 min were selected for subsequent tests.

2.4.4. Loading Stress

The selection of an appropriate loading stress is critical for obtaining representative fatigue damage evolution and self-healing behavior. An excessively low stress level may result in insufficient damage accumulation and significantly prolong the testing duration. Conversely, excessive stress may accelerate damage development and lead to failure behavior inconsistent with the actual fatigue process of asphalt binders.

Therefore, preliminary tests were performed to determine suitable loading stresses for different asphalt binders and aging conditions. As shown in Figure 3, when unaged HRCMA was tested under a stress level of 100 kPa, the dynamic shear modulus decreased rapidly during the initial loading stage. However, the modulus reduction rate gradually decreased in the later stage, and the slope of the modulus–time curve approached a relatively stable state. This indicates that under 100 kPa, the damage evolution rate became insufficient in the later loading stage, resulting in a reduced distinction between fatigue damage accumulation and self-healing recovery. Therefore, a higher stress level was required for HRCMA, and 150 kPa was selected as the appropriate loading stress.

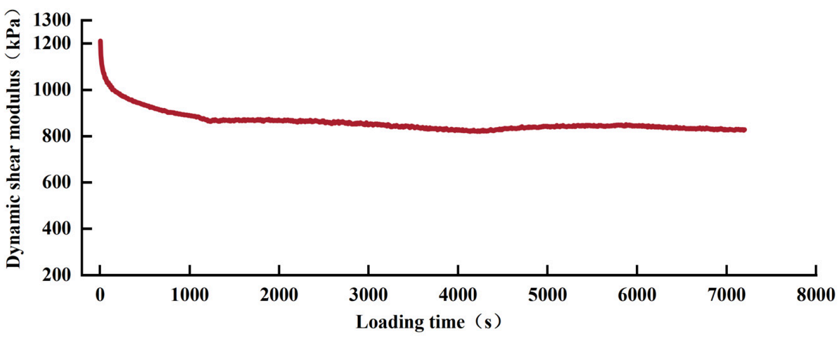


Figure 3. Dynamic shear modulus-loading time diagram of HRCMA (100 kPa).

For TFOT-aged samples, the same stress levels as those applied to the corresponding unaged binders were adopted to ensure comparability when evaluating the influence of aging on self-healing performance. For PAV-aged samples, preliminary results showed that the modulus degradation rate remained relatively low under these stress conditions. Therefore, a loading stress of 220 kPa was selected for both SBSMA and HRCMA after PAV aging. This higher stress level was selected to compensate for the increased stiffness of PAV-aged binders and to ensure comparable damage evolution among different aging conditions.

2.4.5. Statistical Analysis

To evaluate the statistical significance of the observed differences in the self-healing indices among different aging conditions and binder types, a two-way analysis of variance (ANOVA) with aging condition and damage degree as factors was performed separately for the measured HI_E, HI_t, and HI values (three replicates per condition), followed by Tukey's post hoc test at a significance level of $\alpha = 0.05$. Independent samples t-tests were used to compare the two binders under specific aging and damage conditions.

3. Evaluation Index

During the fatigue–healing–fatigue test, the dynamic shear modulus of asphalt binders decreases continuously during fatigue loading due to damage accumulation. After an intermittent healing period, partial recovery of the modulus occurs, followed by further degradation during the second loading stage, as illustrated in Figure 4. Therefore, the self-healing performance of asphalt binders can be evaluated by considering both the recovery of mechanical properties and the recovery of fatigue damage evolution characteristics.

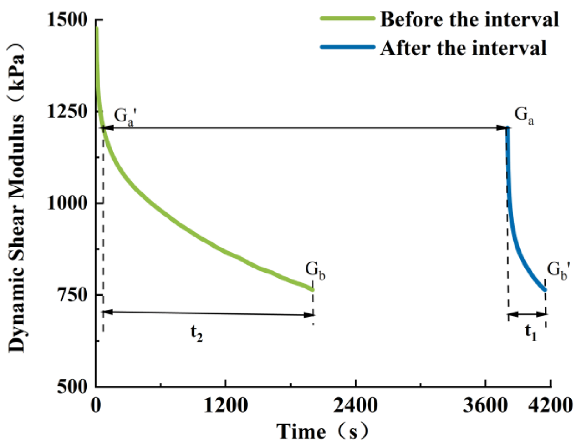


Figure 4. Modulus change graph of asphalt self-healing test.

3.1. Conventional Self-Healing Indices

The self-healing index based on dynamic shear modulus recovery (HI_1) is commonly used to evaluate the recovery of asphalt binder properties after a healing period. Equation (1) [23] calculates HI_1:

$$HI_1 = \frac{G_a - G_b}{G_0 - G_b} \quad (1)$$

where G_0 is the initial dynamic shear modulus of asphalt before the healing interval; G_a is the dynamic shear modulus after the healing interval; and G_b is the reference dynamic shear modulus corresponding to a specific damage state, which was defined as $0.5G_0$ in this study.

In previous studies, G_b was generally defined as $0.5G_0$ to provide a unified reference damage state for comparing the self-healing performance of different asphalt binders. This assumption did not represent a universal failure criterion but provided a consistent basis for evaluating the relative recovery of modulus. However, HI_1 mainly reflects the recovery of the initial mechanical state and does not consider the influence of loading history on modulus evolution.

Considering that the variation in dynamic shear modulus during fatigue loading is also related to loading duration, Luo [19] proposed another self-healing index (HI_2), which incorporates the effect of loading time. Equation (2) calculates HI_2:

$$HI_2 = \frac{G_a - G_b}{G_0 - G_b} \times \frac{t_1}{t_2} \quad (2)$$

where t_1 is the time required for G_a to decrease to $G_b{}'$ after the loading interval; t_2 is the time required for $G_a{}'$ to decrease to G_b before the loading interval, as illustrated in Figure 4.

Although HI_2 considers both modulus recovery and loading duration, the comparison between t_1 and t_2 may be affected by the different initial states before and after healing. Therefore, the time-related parameter in HI_2 may not fully represent the recovery of the material's load-dependent characteristics. A more reasonable evaluation method should compare the modulus evolution under an equivalent damage state before and after healing.

3.2. Development of the Comprehensive Self-Healing Index

The self-healing process of asphalt binders involves two aspects: the recovery of initial mechanical properties and the recovery of resistance to subsequent loading. The former can be characterized by modulus recovery, while the latter is related to the recovery of the modulus degradation process under the same damage condition.

Therefore, in this study, the asphalt binder before and after the healing interval was regarded as two different damage states of the same material. A comprehensive self-healing index was developed by combining the modulus recovery ratio and the recovery of loading time required to reach the same damage degree.

First, the damage degree of the asphalt sample after the second loading stage was calculated by Equation (3).

$$D_2 = \frac{G_a - G_b}{G_a} \times 100\% \quad (3)$$

where D_2 is the damage degree (%) accumulated during the second loading stage.

Then, based on this damage index, the modulus G_d that the asphalt sample reached this damage index during the first loading was calculated using the following formula:

$$G_d = G_0 \times (1 - D_2) = G_0 \times \frac{G_b}{G_a} \quad (4)$$

Furthermore, the loading time t_3 when the modulus reached G_d during the first loading was obtained. As shown in Figure 5, the healing performance characterizing the material's loading response behavior is represented by t_1/t_3.

Based on the above analysis, the proposed comprehensive self-healing index (HI) considers both modulus recovery and loading response recovery. Equation (5) expresses the HI:

$$HI = HI_E \times HI_t = \frac{G_a - G_b}{G_0 - G_b} \times \frac{t_1}{t_3} \quad (5)$$

where HI_E represents the recovery of initial mechanical properties based on modulus recovery, and HI_t represents the recovery of loading-dependent characteristics based on the time required to reach an equivalent damage degree. By integrating these two components, the proposed HI simultaneously considers the restoration of initial properties and the recovery of fatigue damage evolution behavior, providing a more comprehensive evaluation of asphalt self-healing performance.

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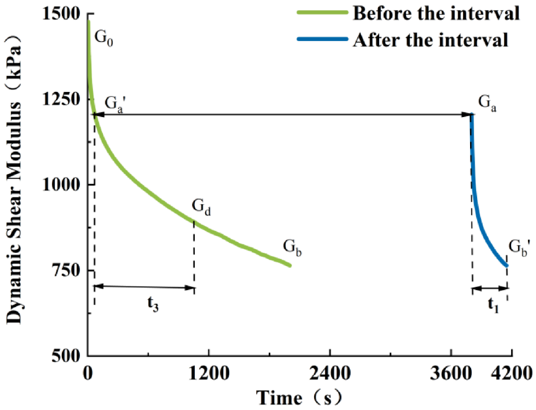


Figure 5. Dynamic shear modulus and time relationship under the same damage degree.

4. Results and Discussion

The self-healing capability of asphalt can be characterized by the degree of performance recovery during the interval period. During the self-repair process, various external conditions influence the healing effect. In this study, damage degree, interval time, and healing temperature were considered.

4.1. Effect of Damage Degree

During pavement service, asphalt binders gradually accumulate fatigue damage under repeated loading, leading to the initiation and propagation of microcracks. The damage evolution process can be quantified by the damage degree, which reflects the reduction in dynamic shear modulus during loading. In this study, the damage degree accumulated during the first loading stage (D_1) was calculated by Equation (6).

$$D_1 = \frac{G_0 - G_b}{G_0} \times 100\% \tag{6}$$

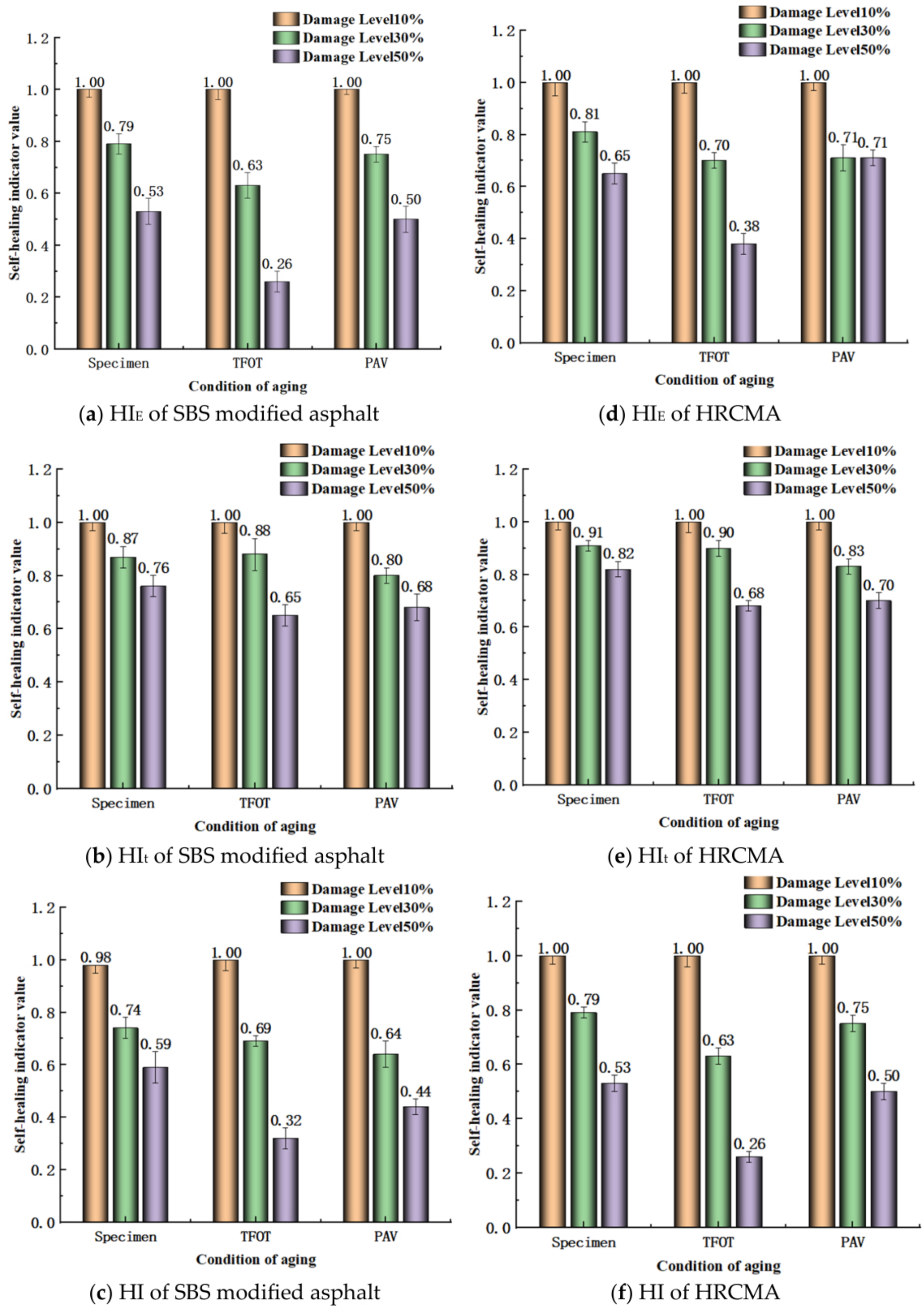
where D_1 is the damage degree (%), G_0 is the initial dynamic shear modulus (MPa), and G_b is the dynamic shear modulus after fatigue loading (MPa).

It should be noted that D_1 defined in Equation (6) and D_2 defined in Equation (3) follow the same fractional reduction form of the dynamic shear modulus, but they characterize the damage degrees accumulated during the first (initial) loading stage and the second loading stage, respectively. D_1 is used to control the target damage state before the healing interval, whereas D_2 is used to determine the equivalent damage state required for evaluating the recovery of loading-dependent behavior in Section 3.2.

A low damage degree generally corresponds to reversible structural changes, during which asphalt binders can recover their mechanical properties after a sufficient healing time. However, with an increasing damage degree, irreversible damage such as crack initiation and structural discontinuity gradually develops, reducing the effectiveness of self-healing [32]. To investigate the influence of the damage degree on self-healing behavior, asphalt samples were subjected to time scan loading until three predetermined damage levels (10%, 30%, and 50%) were reached. After the healing interval, the samples were subjected to secondary loading to evaluate the recovery capability under different damage states. The tests were performed at 25 °C and 10 Hz, and the results for asphalt binders with different aging conditions are presented in Figure 6.

As shown in Figure 6, an increasing damage degree significantly reduced the asphalt's self-healing capacity. For the unaged HRCMA sample, the healing index (HI) decreased from 1.0 to 0.53 as damage increased from 10% to 50%. After TFOT aging treatment, the HI value further decreased to 0.26 under 50% damage conditions, representing a 51%

reduction compared to the original sample. However, the decrease at 30% damage was only 20%. This demonstrates that aging exerts a more pronounced detrimental effect on asphalt self-healing performance under high damage conditions, revealing a synergistic deterioration effect between aging and damage severity on asphalt self-healing properties. Such aging-induced deterioration is consistent with the molecular-size changes reported for long-term aged rubberized asphalt binders [33].



(**a**) HI_E of SBS modified asphalt (**d**) HI_E of HRCMA

(**b**) HI_t of SBS modified asphalt (**e**) HI_t of HRCMA

(**c**) HI of SBS modified asphalt (**f**) HI of HRCMA

Figure 6. Changes in self-healing index with damage degree.

Comparing the test results of the original asphalt and TFOT asphalt under identical stress conditions reveals that the effect of aging on HI_E is more significant, indicating that aging makes it more difficult for asphalt to recover to its initial state, while its impact on damage rate during loading is relatively minor. The influence patterns of the damage degree and aging level on the self-healing capacity of HRCMA align with those of SBS modified asphalt, though the magnitude of their effects differs significantly. Test results on PAV specimens under identical stress reveal that HRCMA exhibits a higher HI self-healing index and more pronounced HI_E value differences, indicating superior transient recovery capability and enhanced initial state self-healing performance. This advantage becomes more evident at a 30% moderate damage degree. It should be noted that the two binders differ not only in crumb rubber content but also in SBS dosage (4% versus 5%), as discussed in Section 2.1. Therefore, the superior self-healing performance of HRCMA should be attributed to the combined effect of high-content crumb rubber incorporation and the slightly lower SBS dosage within the typical engineering formulation range, rather than to the isolated contribution of crumb rubber. Since the SBS network provides the primary structural stability while crumb rubber contributes the additional elastic recovery and energy dissipation capacity, the observed advantage of HRCMA suggests that the crumb rubber phase plays a dominant role in enhancing the healing capability of the composite binder under thermal aging conditions.

Two-way ANOVA results based on the experimental data confirmed that both the aging level ($F(2, 45) = 62.1$, $p < 0.001$) and the damage degree ($F(2, 45) = 1270.3$, $p < 0.001$) had statistically significant effects on the self-healing indices of the two binders. In particular, the difference in HI between HRCMA and SBSMA under PAV aging at the 30% damage degree was statistically significant ($p = 0.047$).

4.2. Effect of Interval Time

Figure 7 shows the actual mounting state of the binder specimen in the DSR: the specimen is clamped between the parallel plates throughout the test, fatigue-induced damage develops beneath the plate, and the observable difference on the specimen surface after fatigue loading is very small. The healing interval provides sufficient time for asphalt binders to recover from fatigue-induced damage. Under the conditions of 25 °C and a damage degree of 30%, the effects of three healing interval times (3 min, 15 min, and 30 min) on the self-healing performance of different asphalt binders are shown in Figure 8.

Because the specimen is clamped by the parallel plates throughout the test, the fatigue-induced damage is confined beneath the rotating plate, and the observable difference on the specimen surface after fatigue loading is very small; therefore, the damage and healing states cannot be directly captured by specimen photographs.

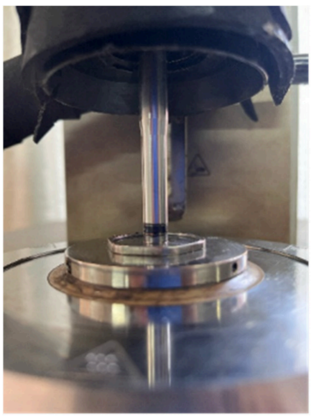
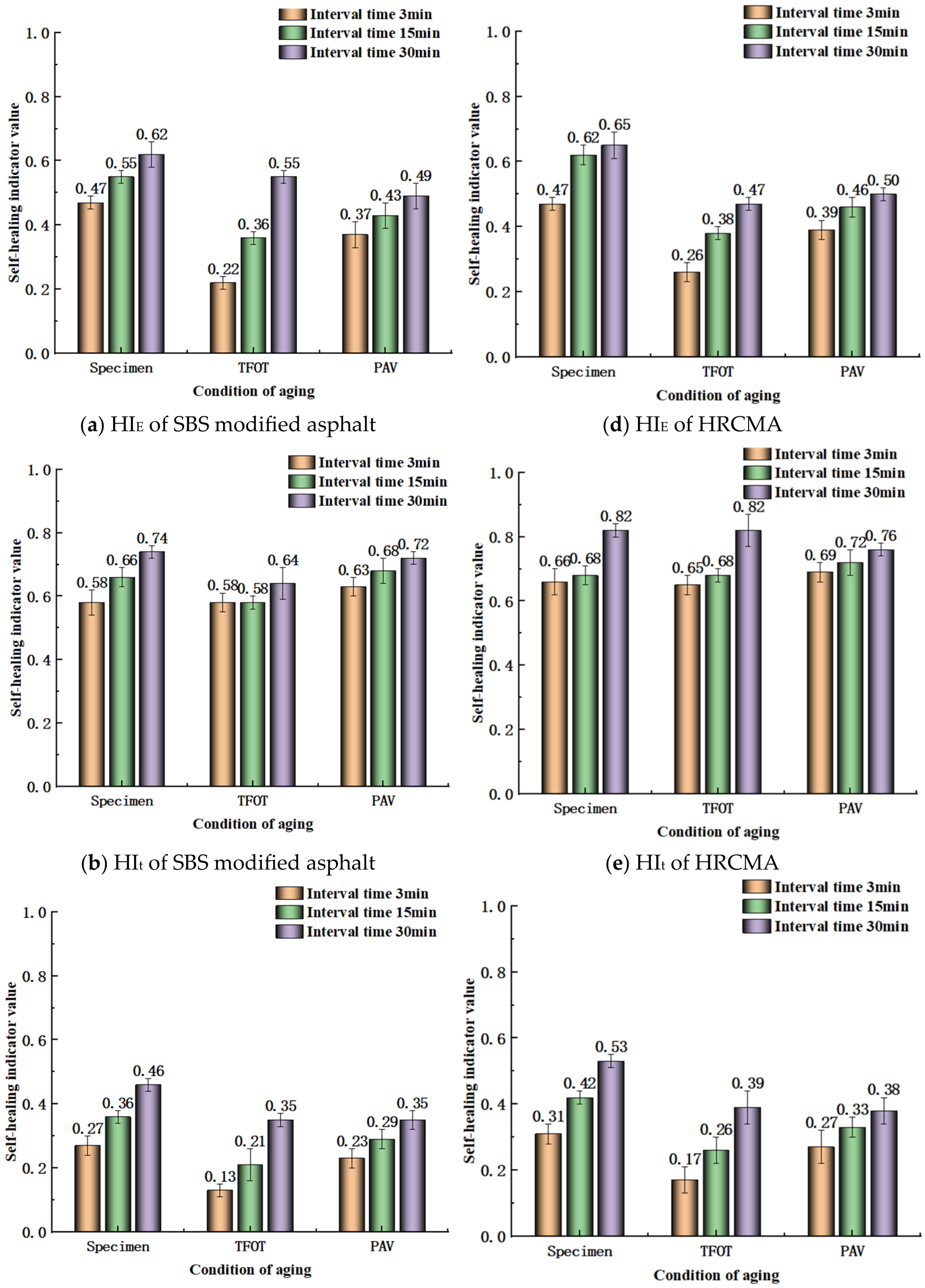


Figure 7. The binder specimen mounted in the dynamic shear rheometer during the fatigue–healing–fatigue test.



(**a**) HI_E of SBS modified asphalt (**d**) HI_E of HRCMA

(**b**) HI_t of SBS modified asphalt (**e**) HI_t of HRCMA

(**c**) HI of SBS modified asphalt (**f**) HI of HRCMA

Figure 8. Changes in self-healing index with interval time.

As shown in Figure 8, the self-healing indices of all asphalt binders increased with an increasing healing interval time, indicating that a longer recovery period promotes the restoration of damaged internal structures. For HRCMA, the HI increased from 0.31 to 0.53 when the interval time increased from 3 min to 30 min, demonstrating the significant contribution of extended healing time to damage recovery. Similar trends were observed for SBS modified asphalt, although the magnitude of improvement varied depending on the aging condition.

The increase in self-healing performance with interval time is mainly associated with the diffusion and rearrangement of asphalt molecular components. Molecular dynamics simulations have shown that the self-healing capability of asphalt binders is governed by the molecular diffusion rate across the crack interface [11], and Sun et al. [34] demonstrated that the chemical composition and colloidal structure of asphalt binders significantly influence their self-healing capability. During the early healing stage, the closure of microcracks and the recovery of intermolecular interactions occur rapidly due to the high mobility of light components [12]. Thixotropic recovery has also been identified as an additional mechanism contributing to the rapid early-stage healing of bituminous mastics [35]. With prolonged healing time, molecular diffusion and structural rearrangement continue, resulting in further recovery of mechanical properties. However, the healing rate gradually decreases because the easily recoverable damage is repaired first, while the remaining damage requires longer molecular movement and reconstruction.

Comparison among different aging conditions shows that aging reduces the sensitivity of self-healing performance to the healing interval time. For PAV-aged samples, the difference in HI values among different interval times was relatively small compared with those of unaged and TFOT-aged samples. This behavior can be attributed to the oxidation-induced increase in asphalt polarity, molecular association, and viscosity, which restrict molecular mobility and diffusion ability. As a result, although extending the healing time provides additional opportunities for recovery, the rearrangement of oxidized asphalt components becomes more difficult, limiting the improvement in self-healing performance.

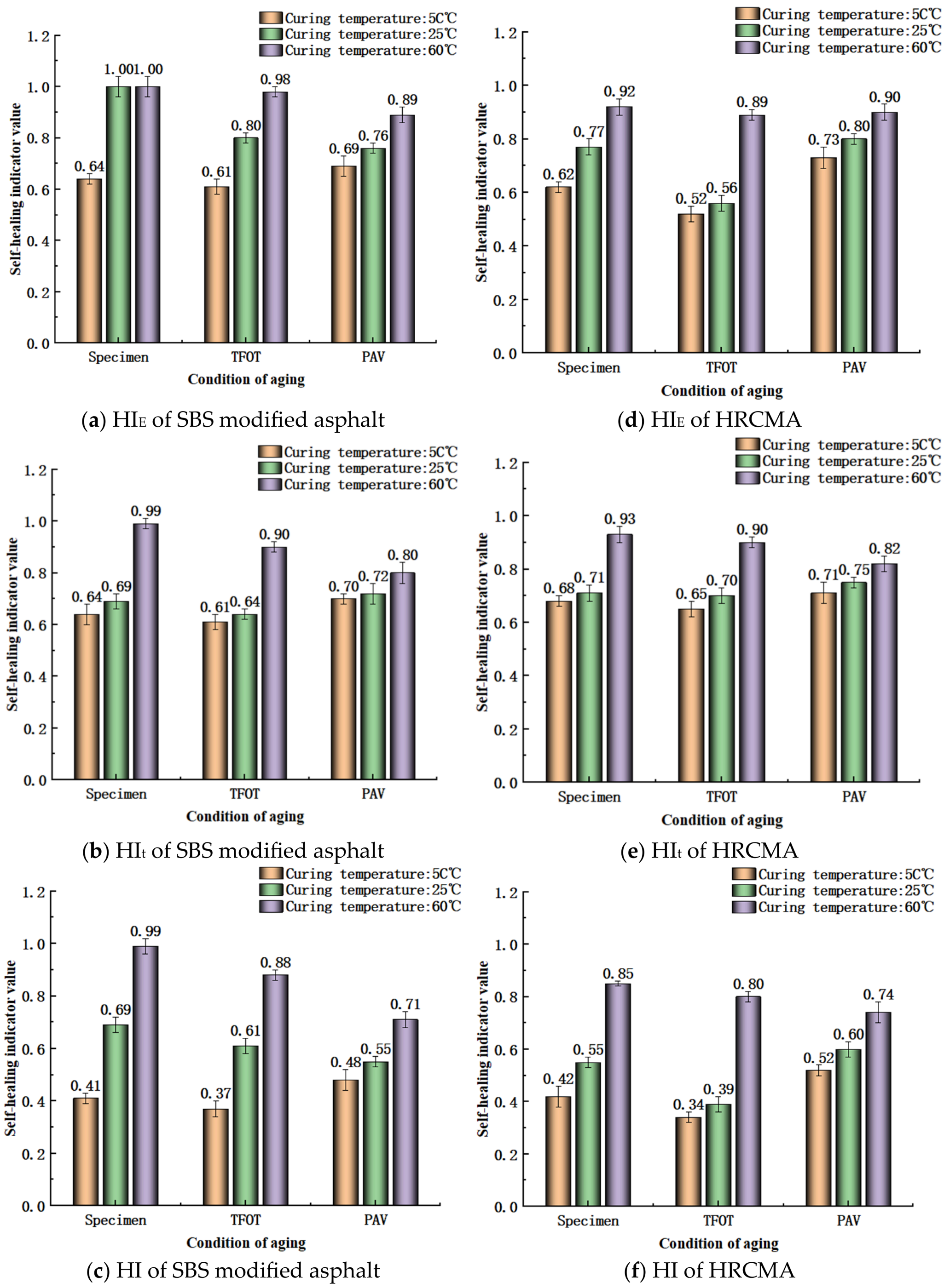
The variations in HI_E and HI_t further demonstrate the different effects of aging on the two recovery mechanisms. Both indices increased with an increasing healing interval time, but exhibited different aging sensitivities. TFOT aging reduced HI_E by more than 50%, indicating that aging mainly weakens the recovery of initial mechanical properties. In contrast, the influence of aging on HI_t was mainly reflected in the suppression of recovery during short healing intervals (3–15 min), suggesting that aging primarily affects the early-stage restoration of loading-dependent behavior.

Overall, the results indicate that the healing interval time plays an important role in determining asphalt self-healing performance, but its effectiveness is strongly dependent on the molecular mobility of asphalt binders. Aging decreases the contribution of prolonged healing time by restricting molecular diffusion and structural rearrangement, resulting in reduced interval time sensitivity.

4.3. Effect of Healing Temperature

The healing temperature directly affects the mobility of asphalt components and consequently influences the recovery of fatigue-induced damage. In this study, asphalt samples were first subjected to loading at 25 °C until reaching a damage degree of 50%. Subsequently, the damaged samples were heated to different healing temperatures (5 °C, 25 °C, and 60 °C) for 15 min. After the healing process, the samples were cooled back to 25 °C for 15 min to eliminate the influence of temperature on the subsequent fatigue loading stage. The self-healing performance of different asphalt binders under various healing temperatures is shown in Figure 9.

As shown in Figure 9, all self-healing indices increased with an increasing healing temperature from 5 °C to 60 °C, indicating that elevated temperature promotes the recovery of fatigue-induced damage. This improvement can be attributed to the temperature-dependent viscoelastic behavior of asphalt binders. Increasing the temperature reduces binder viscosity and increases the mobility of asphalt molecular components, thereby facilitating molecular diffusion, crack closure, and the reconstruction of intermolecular interactions during the healing process. This temperature-dependent healing behavior is consistent with the thermally activated healing tests reported by Liu et al. [29], who found that asphalt binders can completely recover their fatigue damage when heated to temperatures related to their softening points. Induction heating experiments on porous asphalt concrete have similarly demonstrated that crack closure accelerates as the binder temperature increases [36].



(**a**) HIE of SBS modified asphalt (**d**) HIE of HRCMA

(**b**) HIt of SBS modified asphalt (**e**) HIt of HRCMA

(**c**) HI of SBS modified asphalt (**f**) HI of HRCMA

Figure 9. Changes in self-healing index with healing temperature highlighted.

The effect of temperature was not linear over the investigated range. Compared with the temperature increase from 5 °C to 25 °C, the increase from 25 °C to 60 °C resulted in a more significant improvement in HI values. This behavior indicates that asphalt binders require sufficient thermal energy to overcome the resistance associated with molecular movement. Once the temperature reaches a higher mobility range, the diffusion and rearrangement processes become more active, leading to accelerated healing.

The influence of the healing temperature varied among different aging conditions. For unaged asphalt binders, a relatively high self-healing capability was observed even at moderate temperature conditions due to their relatively high molecular mobility. However, aged binders exhibited a stronger temperature dependence. When the healing temperature increased from 25 °C to 60 °C, TFOT- and PAV-aged samples showed a greater increase in HI compared with unaged samples. A similar suppression of the healing efficiency with an increasing aging level has also been reported for aged asphalt mixtures [30]. Likewise, the induction healing efficiency of porous asphalt concrete has been shown to decrease significantly with asphalt aging [37].

This phenomenon can be explained by the structural changes induced by aging. Oxidative aging increases the content of polar components and promotes the formation of stronger intermolecular associations, resulting in higher viscosity and reduced molecular mobility [38], which is consistent with the oxidation-induced evolution of the structure, morphology, and rheology of polymer-modified bitumen reported by Wu et al. Therefore, aged asphalt binders require higher thermal energy to activate molecular diffusion and structural rearrangement during the healing process. The enhanced temperature sensitivity of aged binders indicates that an elevated healing temperature can partially compensate for the loss of self-healing capability caused by aging.

Furthermore, the variations in HI_E and HI_t demonstrate that temperature affects both the recovery of initial mechanical properties and the recovery of loading-dependent behavior. The increase in HI_E with temperature reflects the restoration of the asphalt structure, while the improvement of HI_t indicates an enhanced recovery of resistance to subsequent loading. Therefore, the proposed HI provides a comprehensive evaluation of temperature-dependent self-healing behavior by integrating these two recovery mechanisms.

4.4. Practical Implications for Sustainability and Cost-Effectiveness

The findings of this study also have practical implications for the sustainability and cost-effectiveness of asphalt pavements. The utilization of a high content (25% by mass) of crumb rubber derived from end-of-life tires (ELTs) directly consumes a large number of end-of-life tires, alleviating the environmental burden associated with tire disposal while reducing the consumption of virgin asphalt binder. It should be acknowledged that the higher viscosity of HRCMA requires elevated production and construction temperatures, which increases the plant energy demand; previous analyses of the energy consumption and environmental impact of rubberized asphalt pavements have quantified this additional burden [39]. Nevertheless, life cycle assessment studies have demonstrated that asphalt mixtures containing crumb rubber can achieve lower overall environmental impacts than conventional mixtures when the avoided disposal of end-of-life tires and the reduced binder demand are taken into account [40,41]. From a cost perspective, although the initial material cost of HRCMA is higher than that of conventional modified asphalt, the longer fatigue life and the slower deterioration of the self-healing capability after aging, as demonstrated in this study, are expected to reduce the life cycle maintenance cost of asphalt pavements. A quantitative life cycle cost analysis combined with pavement-scale validation will be conducted in future studies.

5. Conclusions

Based on the proposed comprehensive self-healing index, this study systematically investigated the effects of damage degree, healing interval time, and healing temperature on the self-healing behavior of SBS modified asphalt and HRCMA under different aging conditions. The main conclusions are summarized as follows:

(1) A comprehensive self-healing index (HI) was developed by integrating the recovery of initial mechanical properties and the recovery of loading-dependent behavior. In this index, the initial state recovery was characterized by the modulus recovery component (HI_E), while the recovery of loading-response characteristics was evaluated using the damage-equivalent loading time component (HI_t). The proposed index provides a more comprehensive evaluation of asphalt self-healing behavior than conventional single-parameter indices.
(2) An increasing damage degree significantly reduces the self-healing capability of asphalt binders. Aging and damage degree exhibit a coupled deteriorating effect, where aging intensifies the reduction of self-healing performance under severe damage conditions. Under the investigated aging conditions, HRCMA showed better resistance to aging-induced degradation than SBS modified asphalt, particularly at higher damage levels. Two-way ANOVA confirmed that the effects of the aging level and the damage degree on the self-healing indices were statistically significant (both $p < 0.001$).
(3) Extending the healing interval time promotes the recovery of asphalt self-healing performance; however, the improvement becomes limited after severe aging due to reduced molecular mobility and diffusion ability. Compared with SBS modified asphalt, HRCMA exhibited higher self-healing indices under equivalent aging and healing conditions, indicating its superior recovery capability during the healing process.
(4) An increasing healing temperature enhances asphalt self-healing performance by improving molecular mobility and facilitating structural recovery. However, the temperature sensitivity of self-healing behavior varies with aging conditions. Aged asphalt binders require higher healing temperatures to achieve comparable recovery because oxidative aging restricts molecular movement and increases resistance to structural rearrangement.
(5) The variations in HI_E, HI_t, and the comprehensive HI index showed similar trends under different testing conditions and aging levels. However, the influence of damage, aging, interval time, and temperature was more significant on HI_E than on HI_t, indicating that aging primarily affects the recovery of initial mechanical properties rather than the recovery of loading-dependent damage evolution behavior.

The findings of this study demonstrate that HRCMA possesses enhanced self-healing capability and aging resistance compared with SBS modified asphalt, suggesting its potential application in improving pavement durability and extending service life under long-term traffic loading conditions. By maintaining a higher recovery capability after thermal aging, HRCMA may contribute to reducing the fatigue-related deterioration and maintenance frequency of asphalt pavements. From the perspective of sustainability, the superior retention of the self-healing capability after thermal aging further supports the use of high-content crumb rubber as a promising modifier for durable and low-maintenance asphalt pavements.

Although this study provides new insights into the effect of thermal aging on the self-healing behavior of HRCMA, several limitations should be acknowledged. The present investigation was conducted at the binder level, and direct molecular-level experiments (e.g., spectroscopy or molecular dynamics simulations) were beyond the scope of this binder-level study; such techniques, together with asphalt mixture tests, microstructural characterization, and pavement-scale evaluations, will be pursued in future work to further validate the applicability of the obtained findings and clarify the underlying healing mechanisms.

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X.M. and A.D.A.; project administration, X.M.; validation, Y.L. and M.K.S.; funding acquisition, M.K.S. and A.D.A. All authors have read and agreed to the published version of the manuscript.

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