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Research Article

Influence of Aggregate and Asphalt Type on the Bond Strength between Asphalt and Aggregates and Its Self-Healing Properties

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Determining the bond strength between asphalt and the aggregate base material is an intuitive way to ascertain the adhesion. In this study, we determined the bond strength between different types of asphalt and aggregates. First, the influence of these types of asphalt and aggregates on the bond strength was evaluated. Second, the bond strength of lime aggregate-based material and five types of asphalt in dry and immersion states was measured, and the influence of water on the bond strength was investigated. Third, the asphalt was extracted into aromatic, saturated, colloidal, and asphaltene by an asphalt component separation test, and the bond strength between each asphalt component and aggregate was tested. Finally, for the specimens with adhesive failure, the bond strength was retested to evaluate the self-healing ability of the adhesive. The results show that adhesion is influenced by asphalt and aggregate types as well as water immersion. The adhesion between asphalt and aggregate is primarily gum-based, followed by aromatics. After adhesion failure, the adhesion recovers to a certain extent (that is, it self-heals), but the healing rate decreases with the increase in test frequency; the adhesion tends to be stable after the third test.

1. Introduction

Asphalt pavement is composed of an asphalt mixture through mixing, paving, and rolling, and it is the most widely used pavement type worldwide [1]. An asphalt mixture is a composite of asphalt and aggregate. In addition to the physical and mechanical properties of the two materials, the adhesion between the asphalt and the aggregate is an important factor that affects the performance of the mixture. When the adhesion performance between the asphalt and aggregate is insufficient, the asphalt film is easily peeled off from the surface of the aggregate under the action of water and traffic loads. This low adhesion performance causes failure on the road surface, such as cracking and potholes, which directly affects the performance and service life of the asphalt pavement [2–4]. Therefore, the quality of the asphalt mixture directly determines the performance of the asphalt pavement.

Although several studies have been conducted on the evaluation methods of water damage to asphalt mixtures and the adhesion performance between asphalt and aggregate, the focus has been on the spalling of the aggregate wrapped with asphalt in water, which is an indirect evaluation method. In recent years, the surface energy theory has been used to evaluate the adhesion between asphalt and aggregate, whereby the surface tension of the asphalt and the contact angle between the asphalt and stone substrate are measured.

The adhesion system of asphalt and aggregate is composed of aggregate and mortar. However, some studies have demonstrated that failures in the asphalt-aggregate adhesion system can be categorized as adhesive and cohesive failures [5]. Adhesive failure occurs at the interface of asphalt and aggregates, and cohesive failure occurs at the asphalt membrane, as shown in Figure 1. To test the adhesion between asphalt and aggregate, the failure of the specimen must be tested at the interface between the asphalt and

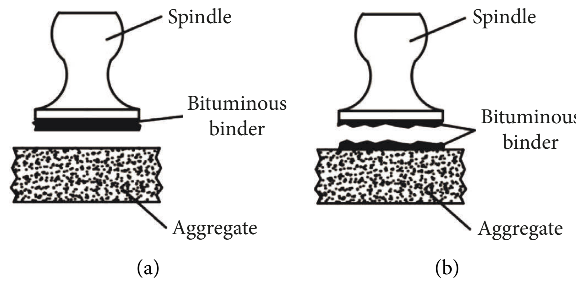

Figure 1: Schematic of adhesive failure and cohesive failure. (a) Adhesion failure. (b) Cohesion failure.

aggregate, rather than the fracture in the middle of the asphalt film. Herein, when the thickness of asphalt film is less than 2 mm, even after repeated tests, the failure between the spindle and stone substrate is referred to as adhesive failure.

Many research studies have proved that the bond strength test can effectively test the adhesive strength between asphalt and aggregate, and then reflect the ability of asphalt mixture to resist water damage. However, the types of aggregates and asphalt binders used in the existing research are not wide enough, especially in the research on the evaluation of the self-healing ability of the bond strength between asphalt and aggregate. This study tested the bond strength between five types of aggregates and five types of asphalt, which can enrich the test cases. The existing research has not yet tested the bond strength between the aggregate and each component of bitumen binders and analyzed the contribution of each component of bitumen binders to adhesion from the perspective of bond strength. In this study, the components of asphalt were separated, and the adhesion strength between each component and the stone substrate was tested, which is conducive to the optimization of asphalt materials from the perspective of asphalt components and adhesion strength.

2. Literature Review

Adhesion performance is evaluated using various methods and indicators, such as the boiling test, agitated water adsorption test, and surface energy test [6–8]. Cucalon et al. used a microcalorimeter to measure the heat transfer between asphalt and aggregate during adhesion and determine the influence of temperature on the adhesion system. Results showed that the adhesion of asphalt and aggregate decreases as the temperature rises, and the adhesion property is the best at 10 ℃-15℃ [9]. Caputo et al. used optical microscopy and X-ray powder diffraction to characterize several mineral aggregates in detail and correlate the boiling test and the contact angle method to investigate the level of bitumen/aggregate affinity [10]. In recent years, surface energy theory has become more common in the study of the adhesion between asphalt and aggregate. Lytton et al. used the method of surface energy theory to study the micromechanism of adhesion between aggregate and asphalt, adhesion durability, and the sensitivity of aggregate and asphalt to water [11]. Han et al. calculated the adhesion of six types of asphalt to granite aggregate before and after adding hydrated lime, and the authors used the surface energy method to evaluate the effect on adhesion when lime is added to the asphalt mixture [12].

Compared with other indirect methods, the bond strength test can directly reflect the bond strength between asphalt and aggregate. Omar et al. summarized a variety of methods used to evaluate the water damage of asphalt mixture and believed that bond strength test is a better test method, which has the advantages of simple operation, strong reproducibility, and wide application range [13]. Aguiar-Moya et al. used the PATTI testing equipment to measure the bond strength between asphalt and various aggregates, and the results revealed that the failure between asphalt and aggregate can be divided into two types: adhesive failure and cohesive failure depending on the aggregate type [14]. The bond strength decreased when the SBR was used. Zhang et al. used the bond strength test and peel test to evaluate the adhesion property between asphalt and aggregate and analyzed the penetration of asphalt to the bond strength. The results revealed that harder (40/60 penetration grade) asphalt exhibits better adhesion property than that of softer (70/100 penetration grade) asphalt [15]. Guo et al. designed a bond strength test using an electrical universal test machine, and the bond strength between the granite substrate and asphalt in different freeze-thaw cycles and test temperatures. The results showed that freeze-thaw cycles would lead to a decline in the bond strength, and at medium and high temperatures, the adhesive strength decreased more significantly with the number of cycles [16]. Wang et al. used bond strength test to test the bond strength between rubber asphalt waterproof adhesive layer and steel bridge gussasphalt pavement and analyzed the influence of water on the bond strength. The results show that water has a significant effect on the adhesive strength; after a freeze-thaw cycle, the measured bond strength decreased by about 60% [17]. Alamrew and Mollenhauer tested the bond strength between three kinds of stone substrate and 70–100 penetration graded bitumen binders adulterated hydrated lime or surfactant by the bond strength test, proving that adulterated hydrated lime and surfactant can improve the adhesion between bitumen binder and aggregate [18]. In addition, the aggregate properties were found not to influence the bonding strength when the cohesive failure occurred. Moreover, some studies have shown that asphalt is a self-healing material, with these properties being directly affected by factors such as temperature [19–21]. García explained the changes in the self-healing rates of asphalt mixture with temperature using the Arrhenius equation and found that there is a minimum temperature below which asphalt cannot be healed [22].

In this study, the PosiTest AT-A automatic bond strength tester was used to measure the bond strength between five types of aggregate and five typical asphalt types under immersion and dry conditions. After adhesive failure, the stubs were immediately returned to their original positions, and the bond strength was retested to determine the degree of self-healing exhibited by the bond.

3. Materials

In this study, three types of matrix asphalt binders (KLMY 70# (penetration grade: 60/80), KLMY 90# (penetration grade: 80/100), and SK 90# (penetration grade: 80/100)) and two types of modified asphalt binders (SBS modified asphalt binder and SBR modified asphalt binder) were selected. These materials were obtained by sampling at the asphalt pavement construction site. The primary physical properties of matrix asphalt and modified asphalt were tested according to the Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering (JTG E20-2011) of China [23], as shown in Tables 1–3.

The stone materials used in this study included basalt, dolomite, diorite, granite, and limestone, and their main technical properties were tested according to the Test Methods of Aggregate for Highway Engineering (JTG E42-2005) of China [24] and are shown in Table 4. These stones are collected by the author from the field.

4. Experiments

4.1. Bond Strength Test. The PosiTest AT-A produced by the United States DeFelsko was used in this test, as shown in Figure 2, and its technical parameters are shown in Table 5. The loading rate of the test was set as 0.7 MPa/s, and the thickness of the asphalt film was maintained at approximately 0.1–0.2 mm by controlling the amount of asphalt smearing on the stubs.

As the stone substrate is cut in the cutting process, some stone powder inevitably adheres to its surface, which would affect the bond strength test. Hence, the surface of the substrate was cleaned using an ultrasonic cleaner. The test steps were as follows:

(1) The cutting machine was used to cut the stone into rectangular substrates with a flat surface, and their side length was not less than 30 mm.

(2) The stone substrates were cleaned first with distilled water and then with an ultrasonic cleaner to eliminate surface powder, as shown in Figure 3. When using an ultrasonic cleaner, the ultrasonic bath was set to a temperature of 60°C, and the cleaning time was not less than 15 min.

(3) Once cleaned, the stone substrates were dried at 105°C in an oven for at least 12 h and placed in a cleanroom afterward.

(4) The stone substrates and pull-out stubs were heated at 60°C in an oven for at least 1.5 h, and the asphalt was heated to 160°C (SBS-modified asphalt was heated to 170°C).

(5) The asphalt was smeared on the pull-out stubs, and the thickness of the asphalt film was controlled at approximately 0.1–0.2 mm by controlling the quality of the coating.

(6) The asphalt-coated pull-out stubs were lightly pressed on the stone substrates and placed in a cleanroom to cool them naturally, as shown in Figure 4.

(7) The bond strength between asphalt and stone substrate was measured by PosiTest AT-A bond strength tester with a loading rate of 0.7 MPa/s.

(8) To study the influence of water on bond strength, the limestone substrates were divided into two groups. For the first group, the stone substrates and pull-out stubs were placed in a temperature test box for at least 1.5 h, and the temperature was set to 25°C. The PosiTest AT-A apparatus was used to determine the bond strength with the loading rate of 0.7 MPa/s, and the bond strength $P1$ was recorded.

(9) For the second group, the stone substrates were placed in a 25°C water bath for 24 h, then the bond strength was tested, and the testing results were recorded.

4.2. Bond Strength Healing Rate Test. Some studies have found that when adhesion failure occurs between asphalt and aggregate, if given a certain intermittent period and appropriate temperature, the asphalt-aggregate with adhesion failure will reproduce adhesion, showing certain self-recovery characteristics [25]. In the past, fatigue test, indirect tensile test, and dynamic shear test were mostly used to influence the self-healing performance of asphalt [26–28].

The bond strength between asphalt and aggregates will partly recover after failure. To test the resilience of adhesion, limestone substrates were used as the test object. After the bond strength test, the stubs were immediately returned to their original positions and replaced in the temperature test box for 24 h; the bond strength was then retested. This operation was repeated 4 times, and the bond strengths of 5 types of asphalt and stone substrates were recorded.

This study defines the bond strength self-healing rate to evaluate the adhesion self-healing properties between asphalt and stone substrates; bond strength self-healing rate can be calculated by the following formula:

$$\eta = \frac{P_n}{P_1}, \quad (1)$$

where η is the bond strength self-healing rate (%); P_1 is the bond strength tested the first time (MPa); P_n is the adhesion strength after the nth self-healing.

Table 1: Technique performance of matrix asphalt.

Types of asphalt	Penetration (25°C, 100 g, 5 s)/0.1	Ductility (5 cm·min^{-1}, 10°C) (cm)	Kinetic viscosity at 60°C (Pa·s)	Softening points (°C)	Density (25°C) (g/cm^3)
KLMY70#	67.5	>100	253	49	0.976
KLMY90#	87.3	>100	175	46	0.998
SK90#	92.6	>100	167	46.5	0.974

Table 2: Technique performance of SBS-modified asphalt.

Penetration (25°C, 100 g, 5 s)/0.1	Ductility (5 cm·min^{-1}, 5°C) (cm)	Kinematic viscosity at 135°C (Pa·s)	Softening points (°C)	Solubility (%)	Elastic recovery rate at 25°C (%)
75	59	1.723	65.5	99.62	94.6

Table 3: Technique performance of SBR-modified asphalt.

Penetration (25°C, 100 g, 5 s)/0.1	Ductility (5 cm·min^{-1}, 5°C) (cm)	Kinematic viscosity at 135°C (Pa·s)	Softening points (°C)	Penetration after aging (25°C, 100 g, 5 s)/0.1	Ductility after aging (5 cm·min^{-1}, 5°C) (cm)
113.1	71.1	0.681	47.8	99.53	38.5

Table 4: Technical index of test stone.

Lithology	Crushing values (%)	Los Angeles abrasion loss (%)	Adhesion grade with asphalt	Water absorption (%)	Apparent relative density	Relative density of gross volume
Diorite	17.5	17.1	4	1.63	2.791	2.747
Dolomite	18.4	19.7	4	0.71	2.797	2.853
Limestone	16.9	18.1	4	1.03	2.749	2.687
Basalt	13.6	15.1	4	1.23	2.949	2.846
Granite	18.6	22.7	3	0.79	2.722	2.651

Figure 2: Automatic bond strength tester of PosiTest AT-A.

Table 5: Technical parameters of PosiTest AT-A.

Sizes of pull-out stub (mm)	Resolution (MPa)	Accuracy	Test range (MPa)
20	0.01	±1%	0–20

Figure 3: Ultrasonic cleaner.

Figure 4: Specimen for bond strength test.

4.3. Bond Strength Test of Asphalt Components and Stone Base Material. Road petroleum asphalt is a mixture of hydrocarbons and nonhydrocarbons, with large molecular contents and complex chemical components [29, 30]. Some researchers extracted and divided the components under different conditions [31–33]. Owing to different separation methods, the division of asphalt components is varied. Among the many division methods of asphalt components, Corbett's four-component division method is widely used, where asphalt is divided into four components: saturated component, aromatic component, gum, and asphaltene [34]. This method was used in this study to extract the asphalt into four components: aromatics, saturates, gum, and asphaltenes.

First, the KLMY 70# asphalt was extracted into the four components, after which the components were heated to a molten state and evenly coated on the spindle, as shown in Figure 5. Finally, the bond strength of the asphalt components and stone substrate in a dry state was tested at 25°C by the method discussed in Section 4.1.

Since the purpose of this test is to reveal the contribution of each component in asphalt to the bond strength, the mass ratio of each component in asphalt cement is not weighed. Considering that asphaltene is very sensitive to temperature, it generally hardens before it can be applied onto the spindle after heating and melting. Even if the spindle is reheated, it cannot be applied evenly, and it is more difficult to bond with stone effectively. Therefore, the bond strength between asphaltene and stone substrate was not tested in this instance.

5. Results and Discussion

5.1. Bond Strength between Asphalt and Stone. The test results of bond strength between five types of stone and asphalt are presented in Table 6.

The following key observations can be made from Table 6 and Figure 6. First, the adhesion of five types of stone materials to five types of asphalt was in the order of dolomite > basalt > limestone > diorite > granite, and the adhesion strength from high to low is SBS-modified asphalt > KLMY70 # matrix asphalt > KLMY 90 # matrix asphalt > SK 90 # matrix asphalt > SBR modified asphalt. Second, the adhesion of KLMY 70 # matrix asphalt and that of KLMY 90 # matrix asphalt to five types of aggregates were similar, indicating that the origin of asphalt is closely related to the adhesion performance. Third, by contrast, the adhesion between SBS-modified asphalt and granite aggregate was the strongest among the five types of asphalt, but the increase was not apparent compared with that of the other four types of asphalt. Finally, the adhesion of SBR-modified asphalt to aggregate was not better than that of matrix asphalt, that is, the SBR modifier did not improve the adhesion of asphalt to aggregate.

In addition, variance analysis was used to evaluate the influence of stone type and asphalt type on adhesion. The fundamental idea of ANOVA is to evaluate the significance of each factor's influence on the results by studying the influence of variation from different sources on the total variation [35]. To further analyze the significance of the influence of stone type and asphalt type on the adhesion, two-factor ANOVA in Microsoft Excel software was used to analyze the difference between the influence of stone type and asphalt type on the bond strength. At the time of analysis, the significance level was set at 0.05, and the results are shown in Tables 7 and 8.

From the above results, at <95% guarantee rate, the test statistics F of stone type and asphalt type were greater than the critical value, indicating that both have a significant impact on adhesion.

5.2. Influence of Water on Bond Strength. Table 9 lists the testing results of the bond strength of the two groups of stone substrates. As shown in Table 9, at 25°C, the testing results for the bond strength were approximately 0.31–0.68 MPa, and the order of bond strength from the largest to the smallest was SBS > KLMY 70# > KLMY 90# > SK 90# > SBR. The bond strength of KLMY 70# matrix asphalt was larger than that of KLMY 90# matrix asphalt, indicating that the adhesion of asphalt was closely related to penetration.

After the water immersion, the bond strength between the five types of asphalt and stone substrates was reduced. The testing results for the bond strength were approximately 0.2–0.54 MPa, and the descending order of the bond strength was SBS > KLMY 70# > SK 90# > SBR > KLMY 90#. SBS-modified asphalt had the best adhesion performance with stone substrates, irrespective of whether it was immersed. The ratio between the two groups of data reflects the change in bond strength before and after immersion, as shown in Figure 7. The bond strength of SBS-modified asphalt had a minimal change, and KLMY 90# matrix asphalt had the maximum change before

(a) (b) (c)

Figure 5: Spindle coated with asphalt components. (a) Aromatics. (b) Colloid. (c) Saturation.

Table 6: Bond strength test results.

Lithology	KLMY 70#	KLMY 90#	SK 90#	SBS	SBR
Diorite	0.59	0.61	0.55	0.76	0.41
Dolomite	1.05	0.86	0.74	1.22	0.52
Limestone	0.68	0.64	0.61	0.85	0.42
Basalt	0.82	0.79	0.69	1.13	0.49
Granite	0.42	0.45	0.4	0.49	0.39

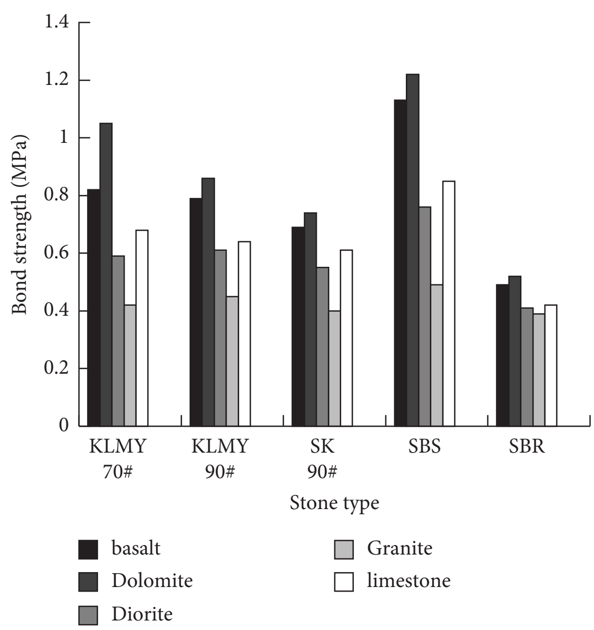

Figure 6: Comparison of bond strength between different asphalt and stone base materials.

Table 7: Two-factor ANOVA without replication.

Summary	Count	Sum	Average	Variance
Row 1	5	3.92	0.784	0.05408
Row 2	5	4.39	0.878	0.07352
Row 3	5	2.92	0.584	0.01578
Row 4	5	2.15	0.43	0.00165
Row 5	5	3.2	0.64	0.02375
Column 1	5	3.56	0.712	0.05677
Column 2	5	3.35	0.67	0.02585
Column 3	5	2.99	0.598	0.01757
Column 4	5	4.45	0.89	0.08625
Column 5	5	2.23	0.446	0.00313

Table 8: Results of ANOVA.

Source of variation	SS	df	MS	F	P value	F crit
Rows	0.609624	4	0.152406	16.4036164	$1.62147E-05$	3.00691728
Columns	0.526464	4	0.131616	14.16596706	$3.99998E-05$	3.00691728
Error	0.148656	16	0.009291			
Total	1.284744	24				

Table 9: Testing results of bond strength.

Asphalt type	KLMY 70#	KLMY 90#	SK 90#	SBS	SBR
Group 1	0.45	0.36	0.38	0.68	0.31
Group 2	0.30	0.20	0.24	0.54	0.23

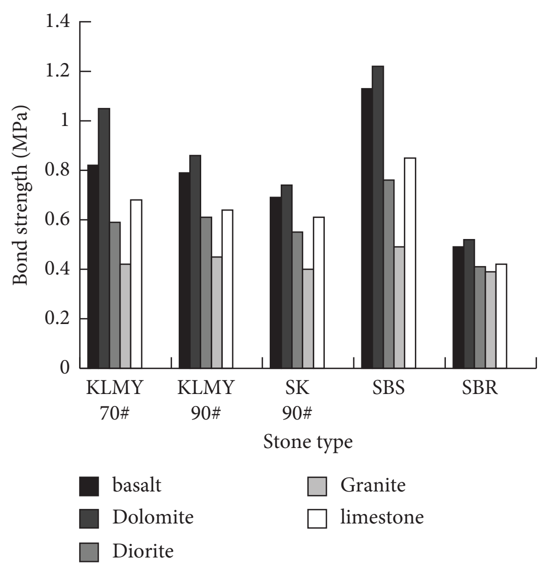

Figure 7: Ratio of two groups of tests.

and after immersion. The change in bond strength ratio before and after water immersion indicates the sensitivity of adhesion to water, with the bond strength ratio being inversely proportional to the sensitivity to water. Consequently, the adhesiveness of SBS-modified asphalt is least sensitive to water, and the KLMY 90# matrix asphalt is most sensitive to water.

The effect of water to adhesiveness is mainly reflected in the following two aspects.

(1) Moisture replacement

Owing to the polarity of aggregates and moisture, aggregates were more likely than asphalt to adsorb water. Moisture penetrated from the interface of aggregates and asphalt, leading to the replacement of the aggregate-asphalt interface with aggregate-moisture interface, under the effect of gravity, capillary action, and traffic loads.

(2) Emulsification of asphalt

Under the effect of moisture, the asphalt film was spontaneously emulsified, decreasing its viscosity and surface tension, and subsequently decreasing the cohesion and adhesion to the aggregate interface.

5.3. Bond Strength of Asphalt Components and Stone Base Material. The bond strength test of several components and limestone-based material was conducted, and the results are shown in Table 10.

As the saturated component was flowing at room temperature, it had almost no bonding ability to the stone; therefore, the measurement of the saturation content and the adhesion value between asphaltene and stone substrate was impossible.

Table 10 shows that the adhesion between asphalt and stone primarily derives from a colloid and secondarily from aromatics. Therefore, it is imperative to select the asphalt with suitable gum content to improve the adhesion between asphalt and stone.

5.4. Self-Healing Properties of Bond Strength. Table 11 lists the testing results of bond strength.

Table 12 lists the bond strength ratio between five types of asphalt and stone substrates. As shown in the table, after the failure of the adhesion interface, the bond strength exhibits self-healing ability. The bond strength ratio decreases with the failure times, and it reduced the

Table 10: Test results of bond strength between asphalt components and stone substrates.

Asphalt components	Aromatics	Colloid	Saturation	Asphaltene
Bond strength (MPa)	0.19	0.61	—	—

Table 11: Bond strength of each adhesion failure.

Test indexes	Test times	KLMY 70#	KLMY 90#	SK 90#	SBS	SBR
Bond strength (MPa)	1st	0.45	0.36	0.38	0.68	0.31
	2nd	0.36	0.29	0.32	0.49	0.27
	3rd	0.24	0.20	0.21	0.42	0.23
	4th	0.23	0.19	0.19	0.42	0.21
	5th	0.23	0.18	0.19	0.41	0.20

Table 12: Calculation results of bond strength self-healing rate.

Test indexes	Test times	KLMY 70#	KLMY 90#	SK 90#	SBS	SBR
Bond strength ratio (%)	2nd	80.9	81.2	85.2	71.8	85.7
	3rd	52.9	56.5	54.1	62.4	73.8
	4th	51.5	52.2	50.8	61.2	66.7
	5th	50.0	50.7	50.8	60.0	64.3

most at the second and third times. The SBR-modified asphalt exhibited the greatest healing rate, indicating that the SBR-modified asphalt shows the best self-healing properties.

6. Conclusions

In this study, the influence of asphalt and stone types on the bond strength between asphalt binder and stone substrate was studied. In addition, the test results of bond strength before and after water immersion were compared, and the influence of water on bond strength was evaluated. The components of asphalt were also extracted, and the bond strength between each component and stone substrate was tested. Furthermore, bond strength recovery between asphalt and aggregate after failure was studied. Several conclusions were drawn from this study which are as follows:

(1) SBS-modified asphalt has the best adhesion performance with stone substrates whether it is immersed in water or not. The effect of water to adhesiveness was mainly reflected in the following two aspects: moisture replacement and the emulsification of asphalt.

(2) The adhesion between asphalt and stone primarily derives from a colloid and secondarily from aromatics. Therefore, selecting an asphalt with suitable gum content is crucial to improving the adhesion between asphalt and stone.

(3) The water immersion has a crucial influence on the adhesion between aggregate and asphalt, with the types of asphalt and aggregate having a clear influence on their adhesion.

(4) After the failure of the adhesion interface, the bond strength exhibited a self-healing ability. The bond strength ratio decreased with the number of failures and reduced the most at the second and third failures. The SBR-modified asphalt exhibited the best self-healing properties.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Conceptualization was done by X. G. and W. Z.; methodology by X. G.; validation by J. L. and H. T.; formal analysis by X. G. and P. Z.; investigation by J. L. and H. T.; resources by J. L. and H. T.; data curation by X. G. and P. Z.; writing—original draft preparation by X. G.; writing—review and editing by X. G.; visualization by X. G.; funding acquisition by J. L. and H. T. All authors have read and agreed to the published version of the manuscript.

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