

Article **Study on the Self-Healing Performance of Microcapsules and Microcapsule-Containing Asphalt**

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Abstract: Mixing microcapsules encapsulating asphalt recycling agents into asphalt can effectively enhance its self-healing performance and alleviate the brittle cracking of pavements due to asphalt aging. In practical engineering applications, microcapsules should have good thermal stability, mechanical properties, and uniform dispersion in asphalt or asphalt concrete, for the effective self-healing of micro-cracks which occur as pavements age. The self-healing performance of microcapsule-containing asphalt is affected by several factors. First, the thermal stability, mechanical properties, and dispersibility of microcapsules in the asphalt were investigated by thermogravimetric analysis, nanoindentation tests, and fluorescence microscopy. Then, the effects of the microcapsules' content, temperature, time, degree of damage, and self-healing times on the self-healing performance of microcapsulecontaining asphalt were investigated through two-stage fatigue loading tests. The results show that the microcapsules have good thermal stability, mechanical properties, and dispersibility. They will not thermally decompose when mixing the asphalt concrete, nor will they fracture in the early stages of a pavement's lifetime, or agglomerate in the asphalt. Mixing microcapsules in asphalt effectively improves its self-healing performance. The self-healing index of microcapsule-containing asphalt increases and then decreases with the increase in microcapsule content. It also increases as the temperature and length of time increases, but it decreases as the degree of damage worsens, and thus, the self-healing time increases.

Keywords: microcapsules; asphalt; self-healing performance; contributory factor

1. Introduction

As a very important material for highway engineering, asphalt is widely used in highway construction; however, used asphalt pavements can develop brittle cracking due to aging from the external environment and traffic loads, which significantly reduces their lifespan [\[1,](#page-10-0)[2\]](#page-10-1); therefore, a technology that slows the aging and cracking of asphalt pavements is crucial. Microcapsule self-healing technology is one of the most effective methods to solve this problem. It is implemented by mixing microcapsules that encapsulate asphalt recycling agents into asphalt. As microcracks develop, the microcapsules fracture under the crack tip stress and release the recycling agent [\[3\]](#page-11-0). The recycling agent gradually fills the cracks due to capillarity, and it continues to diffuse into the aged asphalt, thus restoring the asphalt properties and promoting the self-healing capacity of microcracks [\[4\]](#page-11-1).

In practical engineering applications, microcapsules should have good thermal stability [\[5\]](#page-11-2), mechanical properties [\[6\]](#page-11-3), and dispersibility [\[7\]](#page-11-4) to avoid thermal decomposition and failure when mixing and compacting the asphalt concrete. In the meantime, the microcapsules should have good mechanical properties to remain intact and not fracture prematurely under the vehicle loads in the early stages of the pavement's lifetime. As a result, the microcapsules could achieve the self-healing function when microcracks develop in aged

Citation: Jin, J.; Miao, Y.; Zhao, H.; Chen, J.; Qing, L.; Mu, R.; Chen, X.; Li, Z. Study on the Self-Healing Performance of Microcapsules and Microcapsule-Containing Asphalt. *Sustainability* **2022**, *14*, 12231. [https://doi.org/10.3390/](https://doi.org/10.3390/su141912231) [su141912231](https://doi.org/10.3390/su141912231)

Academic Editors: Marco Rosone, Francesca Ceccato, Mingliang Zhou and Stefano Stacul

Received: 5 September 2022 Accepted: 21 September 2022 Published: 27 September 2022

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asphalt pavements. The microcapsules cannot aggregate into clusters in the asphalt, which would affect the performance of asphalt concrete. It has been reported that microcapsules are not uniformly distributed in asphalt, but aggregated at a certain depth [\[8,](#page-11-5)[9\]](#page-11-6); therefore, the thermal stability, mechanical properties, and dispersibility of microcapsules in asphalt necessitate thorough investigations [\[10\]](#page-11-7). The existing findings suggest that the release of the asphalt recycling agent after the microcapsule fractures can promote the self-healing of cracks, and thus effectively improve the self-healing performance of asphalt [\[11,](#page-11-8)[12\]](#page-11-9); however, most of the studies have focused on the effect of the microcapsule content on self-healing performance [\[13\]](#page-11-10). The time, temperature, and self-healing times also affect the self-healing performance of microcapsules containing asphalt [\[14](#page-11-11)[,15\]](#page-11-12). Furthermore, all these factors may affect each other (e.g., when the temperature is high, the healing process of asphalt that containing microcapsules will be faster); therefore, the relationship between the self-healing performance and relevant factors, such as temperature, healing time, and amount of healing agent (dosage of microcapsule) is very important for the practical application of self-healing asphalt containing microcapsules.

In this study, the thermal stability, mechanical properties, and dispersibility of microcapsules in the asphalt were investigated through thermogravimetric analysis, nanoindentation tests, and fluorescence microscopy. Then, the effects of the microcapsule content, time, temperature, degree of damage, and self-healing times on the self-healing performance of microcapsule-containing asphalt were investigated using two-stage fatigue loading tests using the dynamic shear rheometer (DSR).

2. Experimental Tests

2.1. Raw Material

Lunte®70# and 90# petroleum asphalt were used in the tests, and their basic technical properties were tested according to the Standard Test Methods for Bitumen and Bituminous Mixtures for Highway Engineering (JTG E20-2011). The test results are shown in Table [1.](#page-1-0) The microcapsule adopted was an industrial product, and it had an average particle size of 10 µm. The capsule used for the microcapsules was methylated melamine formaldehyde resin, and the encapsulated product was a high-performance asphalt recycling agent with a density of 0.958 $g/cm³$. The main components of the asphalt recycling agent include saturates and aromatics, which can dissolve and disperse the asphaltene in the aging asphalt, thus softening the asphalt and restoring its properties. As the purpose of this paper is to investigate the effect of the addition of self-healing microcapsules on the behavior of asphalt, the manufacturers and the types of the microcapsule are not included.

Table 1. Basic performance indexes of the asphalt.

2.2. Test Method

2.2.1. Sample Preparation

The microcapsule-containing asphalt was prepared using a high-speed shearing machine. The asphalt was heated in the oven to a flowable state and then poured into a stainless-steel basin, which was placed on a preheated electric stove to maintain the temperature of the asphalt at about 160 ◦C. Then, the desired mass fraction of microcapsules was slowly added to the asphalt. Finally, the microcapsule-containing asphalt was obtained by stirring at 800 to 1200 r/min for 10 to 15 min with the high-speed shearing machine.

2.2.2. Microcapsule Performance Test

The microcapsules should remain intact during the mixing and compaction of the asphalt concrete, particularly during the early stages of the pavement's lifetime, so that they can play their self-healing role at a later stage. In the meantime, the dispersibility of microcapsules in asphalt is also crucial. Uniformly distributed microcapsules can promptly respond to cracks from different directions, which is significantly more conducive to crack healing. In summary, microcapsules should have good thermal stability, mechanical properties, and dispersibility. In this section, the thermal stability, mechanical properties, and dispersibility of microcapsules in the asphalt were investigated through thermogravimetric analysis, nanoindentation tests, and fluorescence microscopy.

(1) Thermogravimetric analysis

The thermal stability of the microcapsules was investigated using thermogravimetric analysis. About 10 mg of microcapsules were put into the HCT-3 thermal analysis system for thermogravimetric analysis in the N_2 atmosphere, and the microcapsule mass changes as a result of temperature changes were recorded. During the test, the heating rate was 10 °C/min, the flow rate of protective gas N_2 was 50 mL/min, and the temperature ranged from room temperature to 700 °C.

(2) Nanoindentation test

With the nanoindentation test, the mechanical properties of materials can be tested at a microscopic level, and the force-displacement curve recorded by the nanoindenter can yield the mechanical properties of the material. A clean piece of glass was prepared, and a layer of glue was applied to it. Then, a layer of microcapsules was sprinkled on it. The piece of glass was left on the table until the glue was completely solidified. The force-displacement curves of the microcapsules were acquired through tests and recorded using the nanoindenter, and the hardness and elastic modulus of the microcapsules were calculated. The schematic diagram of the nanoindentation test is shown in Figure [1.](#page-2-0) In the test, the maximum load was 10 mN, the loading time was 30 s, the unloading time was 30 s, and the load retention time was 10 s. The Oliver–Pharr (O–P) method was used to calculate the elastic modulus E_{1T} and hardness H_{1T} of the microcapsules, where the elastic modulus E_{1T} was derived from Equations (1) and (2), and the hardness H_{1T} was calculated with Equation (3). √

$$
E_r = \frac{\sqrt{\pi}}{2\beta} \frac{S}{\sqrt{A_c(h_c)}}\tag{1}
$$

$$
\frac{1}{E_r} = \frac{1 - v_i^2}{E_i} + \frac{1 - v^2}{E_{IT}} = \frac{9951}{10000E_i} + \frac{15}{16E_{IT}}
$$
(2)

$$
H_{IT} = \frac{F_m}{A_{\text{cmax}}} \tag{3}
$$

where the elastic modulus (E_i) of the diamond indenter is set to 1141 GPa, and the Poisson's ratio is set to 0.07; the Poisson's ratio ($v_{\rm i}$) of the microcapsules is set to 0.25; A_c is the contact area between the indenter and the microcapsule, and A_{cmax} is the maximum contact area between the indenter and the microcapsule; E_r is the indentation-reduced modulus (Pa); F_m is the maximum indentation load, h_c is the indentation depth (m), and S is the contact stiffness (N/m); β is the Berkovich indenter constant, which is set to 1.034. **Sustainability in the contract of 14**, x FOR PEER REVIEW 4 of 14^{, x} FOR PEER REVIEW 4 of 14, x FOR PEER REVIEW 4 of 12, x FOR PEER REVIEW 4 of 14, x FOR PEER REVIEW 4 of 14, x FOR PEER REVIEW 4 of 14, x FOR PEER REVIEW

Figure 1. Schematic diagram of nanoindentation test. **Figure 1.** Schematic diagram of nanoindentation test.

(3) Fluorescence microscopy

Microcapsule agglomerations in the asphalt affect the performance of asphalt concrete and are not conducive to self-healing. Through fluorescence microscopy, the distribution of microcapsules in asphalt can be observed. Briefly, a drop of the prepared microcapsulecontaining asphalt was placed on a piece of clean glass and observed under a fluorescence microscope after the microcapsule-containing asphalt was cooled to room temperature; thus, the distribution of the microcapsules in the asphalt after high temperature and highspeed mixing was obtained.

2.2.3. DSR Two-Stage Fatigue Loading Test

A dynamic shear rheometer (DSR) was used for the fatigue-healing-fatigue two-stage fatigue loading test on the microcapsule-containing asphalt. The working principle of DSR is shown in Figure [2.](#page-4-0) The asphalt sample was placed between the fixed disc and the oscillating disc. The fatigue loading on the sample was exerted by the reciprocating shear of the oscillating disc, and the complex shear modulus of the asphalt decreased continuously with the loading process. The complex shear modulus, G, reflects the ability of the asphalt to resist deformation, and its value is the ratio of the maximum shear stress to the maximum shear strain of the asphalt during the DSR test. The larger the complex shear modulus (G), the stronger the asphalt's ability to resist deformation. Research has shown that microcracks develop in asphalt when the complex shear modulus decreases to 50% of its initial value [\[12\]](#page-11-9). At this point, the microcapsules will fracture under the tip stress generated by the microcracks and release the recycling agent. The recycling agent gradually fills the microcracks due to capillarity and continues to diffuse into the asphalt at the tips of the microcracks, thus softening the asphalt and promoting the self-healing of microcracks. This test is divided into two loading phases, as shown in Figure [3.](#page-4-1) The test is suspended when the complex shear modulus of the sample is reduced to the specified degree of damage (defined as the ratio of the complex shear modulus of the sample after loading to the value before loading), and a period of time is allowed for the self-healing of asphalt. Then, the second phase of loading commences. The test stops when the complex shear modulus of the sample drops to the specified value (50% of the initial complex shear modulus is adopted when considering the effect of dosage, temperature and time, and 10%, 30#, 50% and 70% of the initial complex shear modulus are adopted for the analysis of the effect of degree of damage). In this paper, the self-healing index HI is defined as the ratio of the complex shear modulus increment after asphalt self-healing to the difference between the initial complex shear modulus; upon suspension of the first phase, the magnitude of the HI reflects the self-healing performance of the asphalt, and a higher HI means better self-healing performance. The expression for HI is presented in Equation (4):

$$
HI = \frac{G_2 - G_1}{G_0 - G_1} \tag{4}
$$

where G_0 is the initial complex shear modulus, G_1 is the complex shear modulus upon suspension of the first phase, and G_2 is the complex shear modulus after a period of selfhealing. In this test, 8 mm parallel discs with a disc spacing of 2 mm are used. The strain control mode is adopted, and the strain level is 3% (i.e., the maximum shear strain of the sample during the shear fatigue loading is 3%). The loading frequency is 10 Hz, and the test temperature is 25° C.

Figure 2. DSR working principle diagram. **Figure 2.** DSR working principle diagram. **Figure 2.** DSR working principle diagram.

Figure 3. Principle of DSR two-stage loading test. **Figure 3.** Principle of DSR two-stage loading test.

3. Results and Analysis 3. Results and Analysis 3. Results and Analysis

- *3.1. Performance of Microcapsules 3.1. Performance of Microcapsules 3.1. Performance of Microcapsules*
- 3.1.1. Thermal Stability of Microcapsules 3.1.1. Thermal Stability of Microcapsules 3.1.1. Thermal Stability of Microcapsules

Figure 4 shows the thermal gravity (TG) of the microcapsule, which is the residual Figure [4](#page-5-0) shows the thermal gravity (TG) of the microcapsule, which is the residual mass percentage of the microcapsule according to temperature. It is evident that the microcapsule residual mass percentage decreases continuously with the increase in temperature. As the temperature increases from room temperature to 172 °C, the microcapsule mass loss is 2.4%, which is mainly attributed to the water evaporation from the microcapsule samples. As the temperature increases from 172 °C to 350 °C, the microcapsule mass loss is 11.86%, which is mainly attributed to the water evaporation from the capsule and the decomposition of some incomplete microcapsules. As the temperature continues to rise, the microcapsule residual mass percentage decreases rapidly, which is attributed to the fracturing of the microcapsules and the dissipation of the recycling agent. Thermogravimetric analysis indicates that the thermal decomposition of microcapsules begins at about 350 °C. Asphalt mixes are generally mixed at about 180 °C. Thus, the microcapsules will not decompose prematurely during the mixing of asphalt concrete.

3.1.2. Mechanical Properties of Microcapsules

Microcapsules with three different particle sizes were selected for the nanoindentation test. The load-displacement curves obtained are shown in Figure [5.](#page-5-1) The elastic modulus and hardness are important mechanical properties of microcapsules, and a higher elastic modulus means better resistance to deformation. The elastic modulus and hardness of microcapsules with different particle sizes were calculated using the Oliver–Pharr (O–P) method and Equations (1)–(3). The results are shown in Table [2.](#page-5-2)

Figure 4. The TG curve of microcapsules.

Figure 5. Microcapsule test sample and load-displacement curve. (**a**) Force-displacement curves. **Figure 5.** Microcapsule test sample and load-displacement curve. (**a**) Force-displacement curves. (**b**) SEM photo of microcapsules. (**b**) SEM photo of microcapsules.

Table 2. Microcapsule hardness and elastic modulus. **Table 2.** Microcapsule hardness and elastic modulus.

Project	Μ1	M2	M3
Elastic modulus (GPa) Hardness (MPa)	3.1 147.2	82.6	52.9

Figure 5. Microcapsule test sample and load-displacement curve. (**a**) Force-displacement curves. According to Table 2[, t](#page-5-2)he elastic modulus of the microcapsules ranges from 1.1 to 3.1 and 3.1 to 3.1 and 3.1 a that the elastic modulus and hardness of microcapsules vary with particle sizes, and larger elastic modulus and hardness are dependent on wall of the particle. The thicker the particle wall, the higher the elastic modulus and hardness. The microcapsules with larger particle sizes always have thicker capsules, which lead to a higher elastic modulus and hardness. a pressure no less than 25 mN in asphalt concrete, to prevent them from breaking under vehicle loads [\[6\]](#page-11-3). Based on this, it is estimated that for the microcapsules with a 10 μ m diameter in this study, withstanding a pressure no less than 0.25 mN in asphalt will not cause the breaking of microcapsules due to vehicle loads. As shown in Figure 5, the failure load of microcapsules with different particle sizes is 3 to 9 mN; therefore, the microcapsules can remain intact in the initial period of the asphalt concrete pavement's lifetime. As the asphalt concrete pavement grows older, microcracks appear in the aging pavement, whereas microcapsules fracture under the crack tip stress and release the recycling agent. GPa, and the hardness of the microcapsules ranges from 52.9 to 147.2 MPa. It is evident 3.1 GPa, and the hardness of the microcapsules ranges from 52.9 to 147.2 MPa. It is evident particles (M1) have a higher elastic modulus and hardness. The main reason is that the particles (M1) have a higher elastic modulus and hardness. The main reason is that the Research has shown that microcapsules with a particle size of $100 \mu m$ should withstand

\mathcal{V} 1.2. Diagonalistics of Minus concrete, to prevent them for \mathcal{V} 3.1.3. Dispersibility of Microcapsules with a microcapsules with a microcapsules with a structure of the microcapsules with a structure of the microcapsules with a microcapsules with a microcapsule of the microcapsules wit

Figure [6](#page-6-0) shows the distribution of microcapsules in asphalt after high-speed mixing at high temperatures, where the bright green spots are microcapsules, and the rest is

asphalt. The microcapsules are evenly dispersed and remain intact in the asphalt after phalt. The microcapsules are evenly dispersed and remain intact in the asphalt after memechanical mixing at high temperatures, and no aggregation or agglomeration is observed.
— These observations indicate that the microcapsules have good dispersibility in asphalt, These observations indicate that the microcapsules have good dispersibility in asphalt, which facilitates their prompt responses to microcracks from all directions (i.e., releasing the recycling agent and promoting the healing of cracks). In the meantime, the intact the recycling agent and promoting the healing of cracks). In the meantime, the intact microcapsules indicate that they can withstand the high temperatures of molten asphalt, and that they have certain mechanical strength. that they have certain mechanical strength. asphalt. The microcapsules are evenly dispersed and remain intact in the asphalt after rese observations indicate that the microcapsules have good dispersibility in asphalt,
the recycling the integration and promotion the microcapsules from all directions (i.e., releasing The recycling agent and promoung the healing of cracks). In the meantime, the miact
microcapsules indicate that they can withstand the high temperatures of molten certail

Figure 6. Fluorescence microscopy image of microcapsule-containing asphalt.

3.2. Analysis of the Contributory Factors of the Self-Healing Performance of Microcapsule-3.2. Analysis of the Contributory Factors of the Self-Healing Performance of Containing Asphalt Containing Asphalt Microcapsule-Containing Asphalt

3.2.1. Effect of microcapsule content on the self-healing performance of asphalt 3.2.1. Effect of Microcapsule Content on the Self-Healing Performance of Asphalt 3.2.1. Effect of microcapsule content on the self-healing performance of asphalt

microcapsule-containing asphalt (microcapsule contents of 0.15 wt%, 0.35 wt%, 0.55 wt%, and 0.75 wt%), measured through DSR two-stage fatigue loading tests, are shown in Figure [7.](#page-6-1) In the first stage test, when the complex shear modulus drops to 50% of its initial value, the machine was switched off, and we waited for 60 min to allow for self-healing, before carrying on with the second stage of fatigue loading. The temperature remained at carrying on with the second stage of fatigue loading. The temperature remained at 25 °C 25 ◦C throughout the testing process. throughout the testing process. The self-healing indexes (HI) of 70# and 90# asphalt, and the corresponding The self-healing indexes (HI) of 70# and 90# asphalt, and the corresponding micro-

Figure 7. Self-healing indexes of the 70# and 90# microcapsule-containing asphalt.

As shown in Figure [7,](#page-6-1) the self-healing index of asphalt increases continuously as the microcapsule content increases from 0 to 0.55 wt%. The self-healing indexes of asphalt, with 0.55 wt% microcapsules, improved by 80.4% (70#) and 53.0% (90#), compared with the matrix asphalt. As the microcapsule content increased to 0.75 wt%, the self-healing index decreased by 13.8% (70#) and 15.2% (90#), respectively, compared with that of microcapsulecontaining asphalt with 0.55 wt% microcapsules. These results indicate that an appropriate microcapsule content can effectively improve the self-healing index of asphalt, but an excessive number of microcapsules can hinder the self-healing of asphalt.

By fitting the data in Figure [7,](#page-6-1) which show the two types of microcapsules which contain asphalts, it is evident that the self-healing equations for the self-healing index of asphalt, and the microcapsule content, are quadratic functions with maximum points. The self-healing index of microcapsule-containing asphalt first increased and then decreased with the increase of microcapsule content, peaking at the microcapsule content of 0.55 wt%; therefore, higher microcapsule contents are not always better, and the optimal microcapsule content is about 0.55 wt%.

3.2.2. Effect of Temperature on the Self-Healing Performance of Asphalt

The variation in self-healing indexes as a result of temperature changes was investigated through DSR two-stage fatigue loading tests on 70# and 90# asphalt and the corresponding microcapsule-containing asphalt (microcapsule content of 0.55 wt%). The procedures of the tests are identical to the previous section (i.e., when the complex shear modulus of the sample drops to 50% of its initial value, the first stage is finished, and then a 60 min waiting period is allowed for self-healing, followed by the second stage of fatigue loading). The temperatures are 25 °C, 30 °C, 35 °C, and 40 °C, respectively, for this test.

Figure [8](#page-7-0) shows the self-healing index variation as a result of temperature changes for 70# and 90# asphalt, and the corresponding microcapsule-containing asphalt, where the self-healing indexes of both the matrix asphalt and microcapsule-containing asphalt increase continuously as the temperature also increases. At 40° C, the self-healing indexes of the 70# matrix asphalt and microcapsule-containing asphalt are 5.59 and 3.27 times those at 25 \degree C, respectively, and the self-healing indexes of the 90# matrix asphalt and microcapsule-containing asphalt are 4.15 and 2.77 times of those at 25 ◦C, respectively. The self-healing indexes of microcapsule-containing asphalt are higher than those of the matrix asphalt under the same conditions; therefore, the fatigue loading on the samples successfully fractured the microcapsules and released the recycling agent, which softened the asphalt and promoted the healing of microcracks.

Figure 8. Self-healing index variations between 70# and 90# asphalt and the corresponding micro-**Figure 8.** Self-healing index variations between 70# and 90# asphalt and the corresponding capsule-containing asphalt as a result of changes in temperature. microcapsule-containing asphalt as a result of changes in temperature.

By fitting the self-healing index data of microcapsules at different temperatures, it is evident that the relationship between the self-healing indexes of asphalt and temperature is a power function. That is, the self-healing indexes of asphalt increase as a power function as the temperature increases, and the asphalt performance will eventually be fully restored.
— These results indicate that the increase in temperature facilitates the self-healing of asphalt, and the self-healing indexes of asphalt with microcapsules are always higher than those of the diffusion of the recycling agent in the asphalt and further promote the self-healing of microcracks. the matrix asphalt at the same temperature. Thus, increasing the temperature can promote

3.2.3. Effect of Time on the Self-Healing Performance of Asphalt

0.2 presented in Figure [9.](#page-8-0) In the tests, the degree of damage was 50%, the time was 600 s, 900 s, tained using DSR two-stage fatigue loading tests on 70# and 90# asphalt and the corre-*HI*=0.003e0.0013*^x* +0.119 1800 s, and 3600 s, and the temperature was 25 $^{\circ}$ C. The variations in self-healing indexes in accordance with duration of time were ob-*HI*=0.032e0.0008*^x* +0.109 contan *HI*=0.834e0.0002*^x* −0.759 sponding microcapsule-containing asphalt (microcapsule content of 0.55 wt%), and are

Figure 9. Self-healing index variations between 70# and 90# asphalt and the corresponding micro-**Figure 9.** Self-healing index variations between 70# and 90# asphalt and the corresponding capsule-containing asphalt as a result of time changes. microcapsule-containing asphalt as a result of time changes.

As is evident from Figure [9,](#page-8-0) the self-healing indexes of 70# and 90# asphalt and the As is evident from Figure 9, the self-healing indexes of 70# and 90# asphalt and the microcapsule-containing asphalt increase continuously as the duration of time increases. microcapsule-containing asphalt increase continuously as the duration of time increases. As the length of time reaches 3600 s, both the self-healing index of 70# asphalt, and the As the length of time reaches 3600 s, both the self-healing index of 70# asphalt, and the corresponding microcapsule-containing asphalt, improved by 326% and 407%, respectively, tively, compared with those at 600 s, and the self-healing indexes of the 90# matrix asphalt compared with those at 600 s, and the self-healing indexes of the 90# matrix asphalt and the corresponding microcapsule-containing asphalt improved by 277% and 322%, respectively. The self-healing indexes of the microcapsule-containing asphalt are greater than those of matrix asphalt under the same conditions. The reason for these results is that after after being released from the microcapsules, the recycling agent diffuses more widely and being released from the microcapsules, the recycling agent diffuses more widely and more uniformly in the asphalt over time; this is conducive to the self-healing of microcracks.

By measuring the self-healing indices of microcapsules at different repair times, it is evident that the relationship between the self-healing indexes of asphalt and duration time
evident that the relationship between the self-healing indexes of asphalt and duration time can be expressed using exponential equations (Figure [9\)](#page-8-0). That is, the self-healing indexes of asphalt increase exponentially with time, and the asphalt performance will eventually indexes in the self-h be fully restored; thus, the asphalt performance is more thoroughly restored with time.
The about the contract of the c The self-healing indexes of asphalt with microcapsules are always higher than those of the matrix asphalt over the same period of time, which indicates that the incorporation of microcapsules is beneficial to the self-healing of asphalt.

3.2.4. Effect of Damage Degree on the Self-Healing Performance of Asphalt 3.2.4. Effect of Damage Degree on the Self-Healing Performance of Asphalt

The self-healing indexes of 70# and 90# asphalt and the corresponding microcapsule-The self-healing indexes of 70# and 90# asphalt and the corresponding microcapsulecontaining asphalt (microcapsule content of 0.55 wt%), with different degrees of damage, containing asphalt (microcapsule content of 0.55 wt%), with different degrees of damage, are measured using DSR two-stage fatigue loading tests, which are shown in Figure [10.](#page-8-1) In are measured using DSR two-stage fatigue loading tests, which are shown in Figure 10. In the tests, the degrees of damage were 10%, 30%, 50%, and 70%, the time was 1800 s, and the tests, the degrees of damage were 10%, 30%, 50%, and 70%, the time was 1800 s, and the temperature was $25 °C$.

Figure 10. Self-healing index variations between 70# and 90# asphalt and the corresponding micro-**Figure 10.** Self-healing index variations between 70# and 90# asphalt and the corresponding capsule-containing asphalt as a result of the degree of damage. microcapsule-containing asphalt as a result of the degree of damage.

As is evident in Figure [10,](#page-8-1) the self-healing indexes of 70# and 90# asphalt and microcapsule-containing asphalt decrease continuously as the degree of damage increases. The self-healing indexes of the 70# matrix asphalt and microcapsule-containing asphalt, with a degree of damage of 70%, are lower by 86.7% and 82.2%, respectively, than those with a degree of damage of 10%. The self-healing indexes of the 90# matrix asphalt and microcapsule-containing asphalt with a degree of damage of 70% are lower by 76.4% and 71.0%, respectively, than those with a degree of damage of 10%. This is due to the higher number of microcracks in asphalt with a higher degree of damage, which renders the restoration of asphalt performance more difficult, despite undergoing the same length of time and temperature. In the meantime, the self-healing indexes of microcapsule-containing asphalt are always greater than those of matrix asphalt under the same conditions. These results suggest that microcapsules can effectively promote the self-healing of microcracks despite their higher numbers in the asphalt.

By fitting the self-healing indices of microcapsules according to different degrees of damage, it is evident that the relationship between the self-healing indexes of asphalt and the degree of damage can be modelled using exponential equations. That is, the self-healing index of asphalt decreases exponentially as the degree of damage worsens; thus, the increased degree of damage to the asphalt are not conducive to the self-healing of asphalt, as higher degrees of asphalt damage mean that more microcracks will occur, which render the self-healing of asphalt more difficult. If the degree of damage is the same, the self-healing indexes of asphalt with microcapsules are always higher than those of matrix asphalt, which indicates that the released recycling agent from the fractured microcapsules can effectively promote the self-healing of asphalt.

3.2.5. Effect of Self-Healing Times on the Self-Healing Performance of Asphalt

The effect of self-healing times on the self-healing indexes of 70# and 90# asphalt and the corresponding microcapsule-containing asphalt (microcapsule content of 0.55 wt\%) was investigated through three DSR two-stage fatigue loading tests. The results are presented in Figure [11.](#page-9-0) The degrees of damage in the tests were 50%, the time was 1800 s, and the temperature was 25 °C .

Figure 11. Changes in the self-healing indexes of 70# and 90# asphalt and microcapsule-containing **Figure 11.** Changes in the self-healing indexes of 70# and 90# asphalt and microcapsule-containing asphalt for three self-healing processes. asphalt for three self-healing processes.

Figure [11](#page-9-0) shows the results of three DSR fatigue loading tests on 70# and 90# asphalt Figure 11 shows the results of three DSR fatigue loading tests on 70# and 90# asphalt and the microcapsule-containing asphalt, where the self-healing indexes of 70# and 90# and the microcapsule-containing asphalt, where the self-healing indexes of 70# and 90# matrix asphalt and microcapsule-containing asphalt decrease continuously with self-healing ing times. As the self-healing times reach three, the self-healing indexes of 70# and 90# times. As the self-healing times reach three, the self-healing indexes of 70# and 90# matrix matrix asphalt decrease by 48.4% and 15.5%, respectively, and the self-healing indexes of asphalt decrease by 48.4% and 15.5%, respectively, and the self-healing indexes of the asphalt with 0.55 wt% microcapsules decrease by 10.9% and 8.5%, respectively. The reason for this is that more microcracks are generated in the asphalt as the number of fatigue loading increases, and some are not healed during the allowed time, which aggravates microcrack generation after the subsequent fatigue loading. As a result, the self-healing

indexes of matrix asphalt and microcapsule-containing asphalt decrease. The self-healing indexes of asphalt with microcapsules decrease relatively more slowly with self-healing times, thus indicating that the released recycling agent from the fractured microcapsules softens the asphalt and enhances its self-healing performance.

By fitting the data of the microcapsule self-repair index with different self-repair times, it is evident that the relationship between asphalt self-healing indexes and self-healing times can be described using exponential equations. That is, the asphalt self-healing index decreases exponentially as the self-healing times increase. The reason for this is that as the self-healing times increase, some microcracks are not completely healed before the subsequent fatigue loading, which intensifies the microcrack generation in the asphalt and renders the self-healing of the asphalt more difficult. With the same self-healing times, the self-healing indexes of asphalt with microcapsules are always higher than those of matrix asphalt, thus suggesting that the recycling agent released from the fractured microcapsules can soften the asphalt and promote its self-healing.

4. Conclusions

- (1) The microcapsules had good thermal stability, mechanical properties, and dispersibility. These advantages ensured that they would not thermally decompose when mixing the asphalt concrete, would not fracture under the vehicle loads at the early stage of pavement service, and would not agglomerate in the asphalt.
- (2) Adding microcapsules could effectively improve the self-healing performance, and the recycling agent released from the fractured microcapsules could soften the asphalt and promote its self-healing.
- (3) Higher microcapsule contents were not always better, as the excessive release of the recycling agent from the fractured microcapsules could overly soften the asphalt. The optimal content for the microcapsules in this study was about 0.55 wt%.
- (4) With the increase in time and temperature, the diffusion recycling agent was more uniform in the asphalt, which was conducive to asphalt self-healing; therefore, the self-healing index of the microcapsule-containing asphalt increased with the increase in time and temperature.
- (5) As the degree of damage to the asphalt and self-healing times increased, more microcracks appeared. Thus, the self-healing index of microcapsule-containing asphalt decreased with the degree of damage to the asphalt and self-healing times.

Author Contributions: J.J.: Conceptualization, Methodology, Investigation Writing-original draft, Funding acquisition. Y.M.: Investigation, Writing—review and editing. H.Z.: Methodology, Supervision, Project administration, Writing—Review and Editing, Funding acquisition. J.C.: Investigation. L.Q.: Conceptualization, Methodology, Investigation, Formal analysis. R.M.: Conceptualization, Methodology, Investigation, Formal analysis. X.C.: Investigation, Formal analysis. Z.L.: Investigation, Formal analysis. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (Grant No. 52022027 and 51878239), Science and Technology Project of Department of Transportation of Hebei Province (Grant No. RW-202003).

Informed Consent Statement: Any research article describing a study involving humans should contain this statement.

Data Availability Statement: Data is contained within the article.

Conflicts of Interest: The authors declare no conflict of interest.

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