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# Effect of Waste Engine Oil and Waste Cooking Oil on Performance Improvement of Aged Asphalt

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**Abstract:** In order to explore the applicability of waste engine oil and waste cooking oil used in aged asphalt, the effect of waste engine oil and waste cooking oil on aged asphalt recycling was studied through the analysis of the improvement of its physical, chemical, and rheological properties. Six aged asphalt binders with different aging times were obtained by indoor test simulation using the Thin Film Oven Test at 163 °C. Then, waste engine oil and waste cooking oil with five different dosages were added to investigate improvement performances. The results clearly demonstrated that waste engine oil and waste cooking oil could soften and recover the work ability of aged asphalt effectively. Furthermore, the physical, chemical, and rheological performances of six aged asphalts could be improved to normal level of virgin asphalt if the content of waste engine oil or waste cooking oil was suitable. The rheological properties of aged asphalt with waste cooking oil had better improvement than that with waste engine oil. Overall, the good applicability would provide waste oil a much wider service range in asphalt pavement recycling field. It also provided a method of developing new rejuvenating agent with the two waste oils to achieve complex synergism effect. Moreover, it realized the waste cyclic utilization and environmental protection.

**Keywords:** asphalt binder; aged asphalt; rejuvenating; properties improvement; waste engine oil; waste cooking oil

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## 1. Introduction

Recent societal and economic developments have resulted in an annual increase in the total mileage of highways in China. Less than 10 years after construction or operation, many asphalt pavements required different degrees of maintenance due to the different damages sustained during their service lives. This in turn led to the generation of large quantities of reclaimed asphalt pavement materials [1]. Asphalt pavement maintenance was facing the problems of disposal of waste asphalt concrete. The continued use of nonrenewable sources caused environmental pollution as a result of improper waste disposal [2,3]. Therefore, the recycling technology of waste asphalt pavement is becoming more and more attractive from the perspective of energy and environmental protection in the world [4,5].

On the other hand, with the development of automobile and living conditions, large amounts of waste oils, such as waste cooking oil (WCO) and waste engine oil (WEO), are generated. In previous studies, many scholars have proposed various asphalt rejuvenators and analyzed their effects on the properties of aged asphalt [6–8]. It was also important to develop new rejuvenators using the WEO and waste cooking oil (WCO). Researchers found that WCO and WEO were similar in molecular structure to asphalt, which was a favorable condition for their reuse. It was well known that the reused of aged asphalt was the key to recycling asphalt pavement. From the chemical point of view, asphalt regeneration is a reversal of its aging. As such, adding new asphalt binders or regenerators with appropriate chemical compositions and properties that had been reduced in aged asphalt could recover the performances of aged asphalt [9]. Asphalt binders consist of four fractions including saturates aromatics resins and asphaltenes. Based on the compatibility theory of rejuvenation, WCO and WEO can be used as a low viscosity component to recycle aged asphalt. Compared with other low viscosity components, flash points of WCO and WEO were above 200°C, which showed that the use of WCO and WEO in hot mix asphalt mixes had high structural safety. The effective treatment or reuse of these materials not only reduced environmental pollution and conserves energy, but also represented an innovative method of recycling waste. The proposed method could recover or improve the properties of aged asphalt and provide an effective method to regenerate the aged asphalt using waste oils. Therefore, it was practically significant and would provide a broad application prospect for WEO and WCO in the asphalt pavement recycling field.

In recent years, many studies were carried out on the use of WEO or WCO as asphalt rejuvenators. Due to similarity in molecular structure of WEO and WCO with asphalt, several attempts had been made to use WEO and WCO in conjunction with asphalt binder. Sara R.M. [10] studied, by thermochemical characterization, the bituminous binders modified with different WEO and recycled engine oil bottoms combined with polymers were studied with penetration, softening point and viscosity tests. The modified bituminous binders had similar penetration values and softening point temperatures higher than those of commercially modified binders. Liu [11] studied the effect of WEO on the rheological properties of asphalt. The results indicated that WEO-modified asphalt had lower elastic portion, heat sensitivity, zero shear viscosity, and antirutting ability, but had higher fatigue resistance and temperature sensitivity. Jia [12] showed the improvement of rheological properties of aged asphalt using dynamic shear rheometer with up to 5% WEO content. However, Hesp and Shurvell [13] found that it could result in poor low temperature performance of rejuvenated asphalt with the WEO content beyond 15%. Hallizza Asili.et.al [14–16] also studied the physical and chemical properties of rejuvenated asphalt to prove its application. Zhang et al. [17] evaluated the effects of WCO viscosity and acid value on the basic, rheological, and chemical properties of typical rejuvenated asphalt. The experimental results indicated that WCO qualities influenced the rejuvenation behaviors of aged asphalt significantly, and that WCO with higher qualities tended to achieve better rejuvenation effects. Recently, Zargar, M [18] conducted a study using natural oils, including WCO and recycled motor oils, as potential natural regenerators. It was found that 3-4% of the WCO could restore the physical properties of the original asphalt binder compared to 20% recycled oil. P. Caputo et al. [19] investigated the efficiency of bitumen rejuvenator investigated through Powder X-ray Diffraction analysis and T2-NMR spectroscopy. The results demonstrated that the HR (an oleic acid-based rejuvenator) additive effectively reconstituted the correct balance between asphaltenes and maltenes, resulting in a reactive colloid network. Yu [20] discussed the rejuvenation effects of WCO on the rheological, microscopic, and chemical characterization of RAP binders. The results demonstrated that adding WCO could restore the rheological properties and reproduce the surface microstructures of aged binders. Zhang [21] studied the physical properties and structure of WCO-rejuvenated asphalt and WEO-rejuvenated asphalt. It was found that both waste oils could significantly reduce sulfoxide groups S=O and carboxyl groups C=O in aged asphalt. These conclusions indicated that WCO or WEO in suitable contents could rejuvenate aged asphalt to achieve the properties of original asphalt and meet all physical requirements. In summary, WEO and WCO could both have positive effects on recovering aged asphalts properties to the original asphalt level with addition of a suitable content.

In this paper, the effects of WEO and WCO on the performances of six aged asphalts with different aging times were studied. The physical, chemical, and rheological properties of the six aged asphalts with different WEO and WCO dosages were analyzed. According to the observed improvements on the properties of aged asphalts, optimum dosages of WEO and WCO were proposed. Thin-layer chromatography with flame ionization detection (TLC-FID), Fourier-transform infrared (FTIR) and dynamic shear rheometer (DSR) tests were conducted to further evaluate the regeneration effects of WCO and WEO. Though some researches indicated that waste oil could rejuvenate aged asphalts, little research has been conducted to investigate different effects of different oils using comparative analysis and to identify shortcomings of some waste oils to rejuvenate aged asphalt. The objective of this paper was to investigate properties improvement of aged asphalt with WEO or WCO and analyze different improvement effects to illustrate which waste oil can lead to better rejuvenated asphalt. This project could lay the foundations for the application of WEO or WCO in the asphalt recycling field and was also an effective attempt to recycle the waste oils and waste asphalt concrete.

## 2. Materials

### 2.1. Asphalt

The virgin asphalt (VA) used in this research was KLMY Pen70 asphalt (One of the petroleum asphalts types named by the Karamay Oilfield), which was produced in western China. TOFT was used to prepare aged asphalts test samples. The TFOT was set up at 163 °C for 300 min in terms of JTG E20 T0609 which was similar with ASTM D1754 [22]. At the same time, in order to improve the accuracy of the test data, the aging time interval was set to 2 h. It simulated different aging times during operation process. The aged asphalt binders were obtained by laboratory test simulations of the TFOT with different aging times of 5 h, 7 h, 9 h, 11 h, 13 h, and 15 h at 163 °C. The different aging times could reflect different service times of actual asphalt pavement. The basic properties of VA and six aged asphalts are shown in Table 1.

**Table 1.** The basic properties of virgin asphalt (VA) and six aged asphalts.

| Categories | Penetration (25 °C)/0.1 mm | Softening Point/°C | Ductility (10 °C)/cm | Viscosity (135 °C)/mPa.s |
|---|---|---|---|---|
| VA | 64.8 | 51.4 | >100 | 752 |
| Aged 5 h | 51.9 | 52.0 | 26.0 | 816 |
| Aged 7 h | 48.6 | 53.2 | 17.1 | 886 |
| Aged 9 h | 45.1 | 54.9 | 15.8 | 981 |
| Aged 11 h | 43.0 | 55.3 | 13.7 | 1140 |
| Aged 13 h | 41.2 | 55.9 | 13.0 | 1190 |
| Aged 15 h | 39.0 | 56.2 | 12.3 | 1270 |

It can be seen from Table 1 that the basic physical properties of the asphalt change most obviously after 7 to 9 h of aging. The penetration of aged asphalts (5–9 h) was reduced by ~20–30% compared to the VA. This stage belongs to the initial rapid aging stage of asphalt. When the aging time increased, the penetration of aged asphalt (11–15 h) decreased by ~33–40%. This stage belonged to the rate aging stage of asphalt. Comparing different aging time in the laboratory with the aged asphalt with different service life of asphalt pavement recovery, it could be considered that the aging time of 5 h, 7 h, 9 h, 11 h, 13 h, and 15 h were chosen to simulate the service time of 1–4 years, which was a simplified simulation method for test [23]. Therefore, these aging times were selected to evaluate the asphalt aging. All the aged asphalts were produced as aged asphalt samples in this research to analyze the different applicability effects of WCO or WEO.

### 2.2. Waste Engine Oil and Waste Cooking Oil

The WEO utilized in the study was collected from a local auto repair shop. The WCO used in this study was soybean oil, which had been used several times. The basic properties of WEO and WCO were then tested. Based on previous studies, metals such as lead, zinc, calcium, and magnesium, were found in the used engine oil [24,25]. Therefore, WEO and WCO were filtered in order to remove solid impurities to ensure regeneration effectiveness. The basic properties of WEO and WCO were tested and the test results were present in Table 2. A medical absorbent cotton (grade SD-I) was selected over of filtration screens as shown in Figure 1. The WEO was dark in color, while the WCO was yellow, as shown in Figures 2 and 3, respectively.

**Table 2.** The basic properties of waste engine oil (WEO) and waste cooking oil (WCO).

| Categories | Flash Point/°C | Viscosity/Pa·s | Torque/% | Acid Value/mgKOH/g |
|---|---|---|---|---|
| WEO | 195 | 0.093 | 18.6 | 0.63 |
| WCO | 275 | 0.059 | 11.7 | 0.50 |

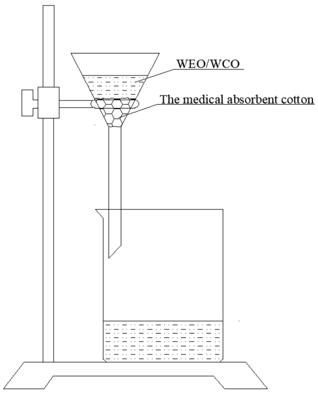



**Figure 1.** The filter device.

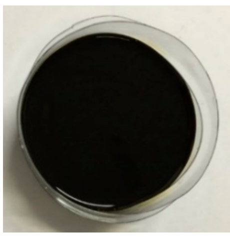


**Figure 2.** Filtered waste engine oil (WEO).

**Figure 3.** Filtered waste cooking oil (WCO).

## 3. Experimental Sections

### *3.1. Rejuvenated Asphalts Preparation*

The rejuvenated asphalt samples were made by mixing six aged asphalts with five WEO and WCO contents (1%, 2%, 3%, 4%, and 5%, by mass of asphalt). A propeller mixer at constant speed (2000 rpm) was used to blend the mixture for 15 min at 135 °C. During the mixing, the time and temperature remained stable to ensure stable experimental results. After blending, the homogenous rejuvenated asphalt binders were produced for the property tests. During testing, when the basic properties of aged asphalt were tested, the waste oils were added to the other aged asphalt samples immediately after recovering the aged asphalt from the TFOT plates to avoid reheat and extra aging.

### 3.2. Physical Properties Tests

In order to evaluate the physical properties of WEO- and WCO-rejuvenated asphalts, the basic tests, such as penetration at 25 °C, softening point, ductility at 10 °C, and rotational viscosity at 135 °C, were conducted in this study, in accordance with JTG E20 T0604-2011, JTG E20 T0606-2011, JTG E20 T0605-2011, and JTG E20 T0625-2011, respectively. All tests were similar to ASTM D5, ASTM D36, ASTM D113, and ASTM D4402. The penetration reflected the viscosity and the degree of hardness of asphalt. Generally, the smaller penetration asphalt, the larger viscosity and harder the asphalt would be. The softening point of the asphalt reflected the viscosity and the temperature sensitivity of asphalt. It would increase with the viscosity values increasing. The asphalt with a higher softening point would have greater viscosity, better temperature and thermal stability. The ductility could be used to measure and characterize the ability of asphalt to undergo tensile deformation and flexibility under external forces without damage and fracture [26,27]. The test processes were shown in Figure 4.

(**a**) Penetration test (**b**) Softening point test (**c**) Rotational viscosity test

**Figure 4.** Basic physical properties tests of rejuvenated asphalt with WEO or WCO.

### 3.3. Chemical Properties Tests

To analyze the influence of WEO or WCO on component composition and structural change of the different degrees of aged asphalts, the samples were tested and characterized with chemical methods. The four components of asphalts were explored and analyzed using TLC-FID. The different samples were prepared by solution of 80 mg asphalt in 4 mL toluene; the concentration of solutions was 2% (*w*/*v*) [28]. It could explain the asphalt components of saturates, aromatics, resins, and asphaltenes by TLC-FID.

Afterwards, the FTIR was used to determine the different functional groups of VA, aged asphalts and rejuvenated asphalts in wave numbers ranging from 4000 $cm^{-1}$ to 500 $cm^{-1}$. The FTIR spectrometer was shown in Figure 5.

**Figure 5.** FTIR spectrometer.

### 3.4. Rheological Properties Tests

To evaluate the high temperature performance recovery of rejuvenated asphalt, the dynamic shear rheometer (DSR) test was often used and analyzed, and the phase angle and the complex modulus imparted basic rutting resistance [29]. Phase angle (δ) is defined as the ratio of loss modulus to storage modulus, and it reflected the viscous response of asphalt binders. Generally, phase angle was much sensitive to asphalt structure. Asphalt with smaller phase angle had better elastic recovery performance. Complex modulus (G*) was defined as the ratio of maximum shear stress to maximum strain, and it provided a measure of total resistance to deformation during shear loading. Asphalt with large complex modulus had better resistance to flow deformation. Rutting resistance factor (G*/sinδ) reflected unrecoverable deformation of asphalts during loading process. Asphalt with higher G*/sinδ, but smaller flow deformation at high temperature, had a higher rutting resistance, which was demonstrated by its better high temperature performance.

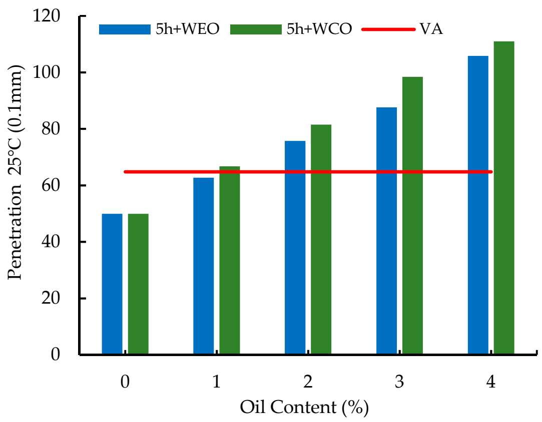
The DSR test was in accordance with JTG E20 T0628, which was similar to ASTM D7175-15. The sinusoidal oscillating load with a frequency of 10 rad/s was applied to the samples. The strain value range of aged asphalt was 8–12%. For the regenerated asphalt, it was better for the strain value to achieve 8–12%, which would prove the regeneration effect of WEO or WCO.

## 4. Results and Discussion

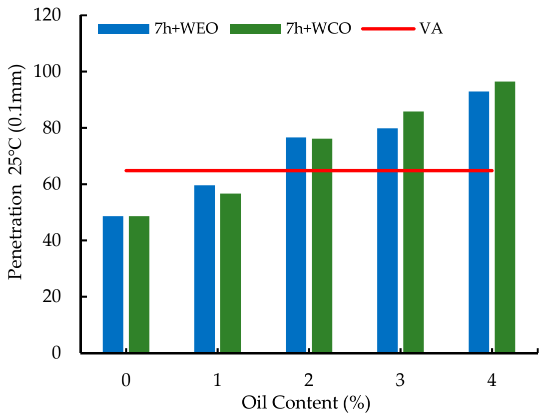
### 4.1. Penetration

The different penetration values at 25 °C for six aged asphalt binders with different aging times are illustrated in Figure 6. It can be clearly observed that the penetration values of all aged asphalts had a similar trend whether it was WEO or WCO that was added, as all the aged asphalts were softened with increased WEO or WCO dosages.

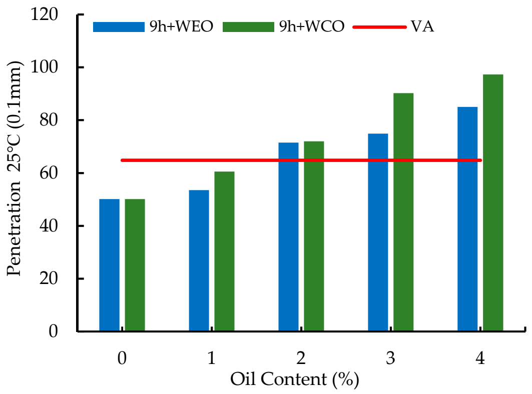
The penetration values of the aged asphalts were significantly larger than that of VA. As revealed in Figure 6, different aged asphalts could reach or exceed the hardness degree of VA with different oil contents. Compared with the VA, the penetration of 5-h- and 7-h-aged asphalts recovered up to 96.8% and 92.0% with 1% WEO content, 9 h- and 11 h-aged asphalts recovered up to 110.3% and 97.7% with 2% WEO content, while 13-h- and 15-h-aged asphalts recovered up to 105.2% and 99.4% with 3% and 4% WEO contents, respectively. This proved that WEO could soften aged asphalt. The effect of WEO content on asphalt penetration was better for specific aged asphalt. Although WCO had similar effects on penetration, it could achieve the same regeneration effects in lesser contents, thereby confirming its better regeneration efficiency than WEO. With 1%, 2%, and 3% WCO content, the recovery extents for asphalts aged 5 h, 7 h, 9 h, 11 h, 13 h, and 15 h were 102.9%, 87.3%, 93.4%, 98.1%, 107.7%, and 102.9%, respectively. Moreover, it could be inferred that there were more light components in the WCO which gave the aged asphalt better fluidity. Although more WEO or WCO could achieve better improvements, excessive oil content affected other properties of aged asphalt. There was a suitable WEO or WCO content range, which depended on the aging degree of asphalt. The asphalt recovered from real reclaimed asphalt pavement (RAP) had a more severe aging state. Therefore, the optimum dosage of regenerating agent in actual road engineering should be determined by considering the actual aging conditions.



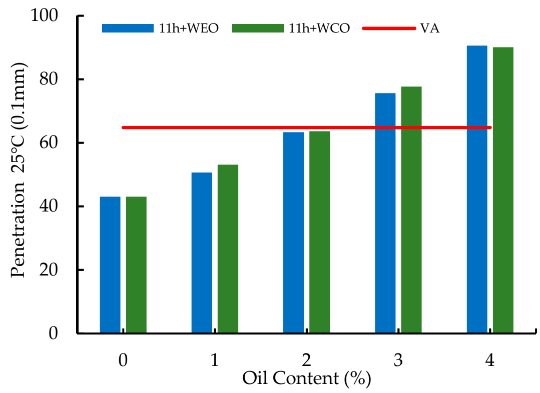

(**a**) Rejuvenated asphalt (5 h aging time of VA)



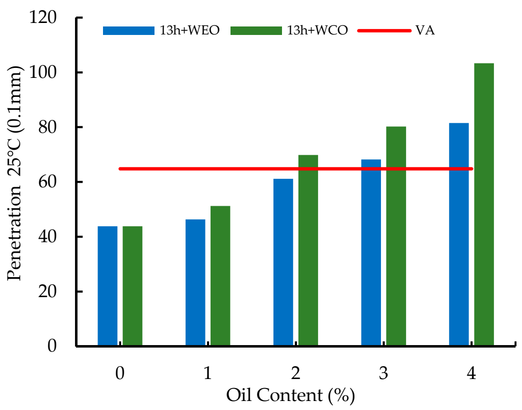

(**b**) Rejuvenated asphalt (7 h aging time of VA)



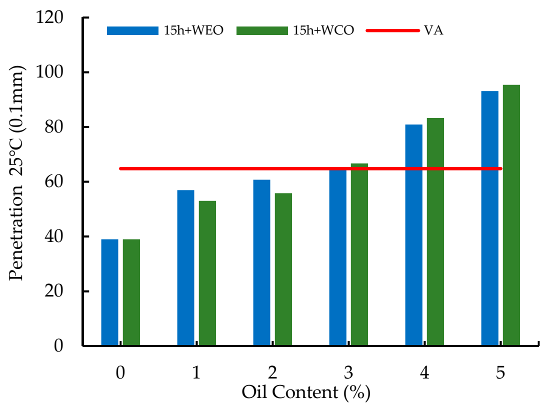

(**c**) Rejuvenated asphalt (9 h aging time of VA)




(**d**) Rejuvenated asphalt (11 h aging time of VA)




(**e**) Rejuvenated asphalt (13 h aging time of VA)

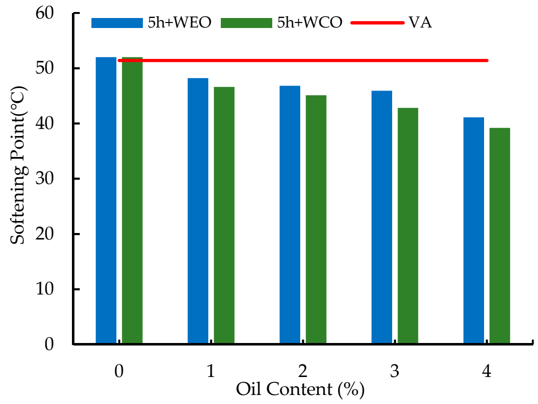



(**f**) Rejuvenated asphalt (15 h aging time of VA)

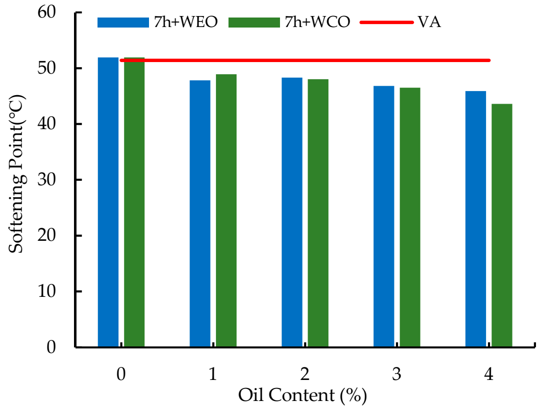
**Figure 6.** Effects of WEO or WCO on penetration of different aged asphalt.

### *4.2. Softening Point*

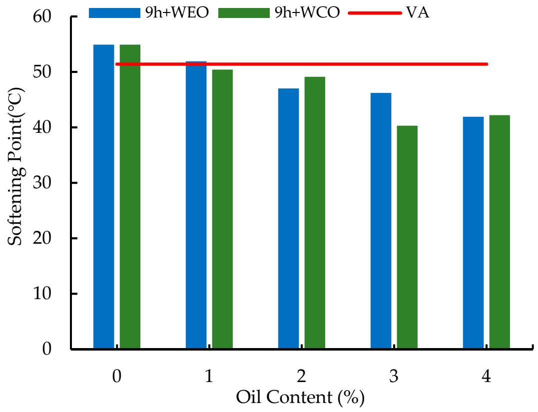
The effects of WEO and WCO on the softening point of aged asphalts with different aging times are displayed in Figure 7. It could also be observed that the softening point values of the aged asphalt binders were consistent with the previous conclusion in 4.1 as they decreased with increase in WEO or WCO contents.



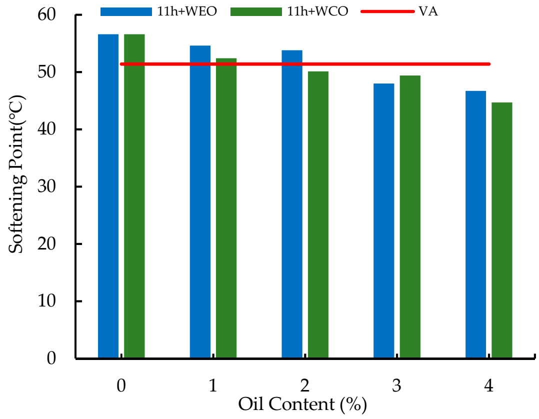

(**a**) Rejuvenated asphalt (5 h aging time of VA)



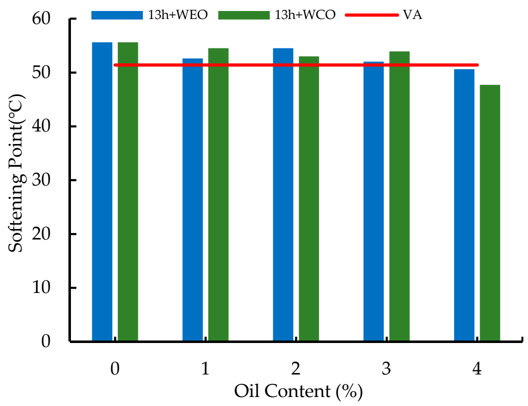

(**b**) Rejuvenated asphalt (7 h aging time of VA)



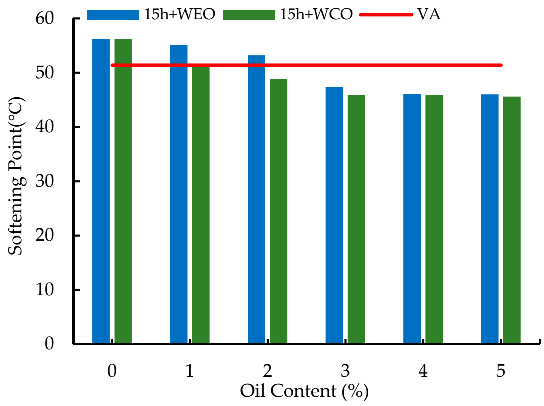

(**c**) Rejuvenated asphalt (9 h aging time of VA)




(**d**) Rejuvenated asphalt (11 h aging time of VA)




(**e**) Rejuvenated asphalt (13 h aging time of VA)




(**f**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 7.** Effects of WEO and WCO on softening point of different aged asphalt.

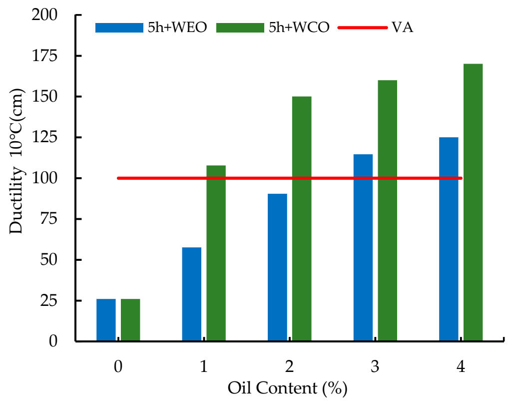
As shown in Figure 7, different aged asphalts could reach or exceed the softening point of VA with different oil content. The softening point of asphalts aged 5 h, 7 h, and 9 h recovered up to 93.8%, 93.0%, and 101.0%, respectively, with 1% WEO content. 11 h-aged asphalt recovered up to 104.7% with 2% WEO content. 13-h- and 15-h-aged asphalts recovered up to 101.2% and 103.5% with 3% WEO content, respectively. This proved that WEO could recover some of the aged asphalt's properties, but the effect of WEO content on aged asphalt was better for specific aged asphalt. Meanwhile, the WCO also had a similar effect on softening point. With 1% and 2% WCO contents, asphalts aged 5 h, 7 h, 9 h, 11 h, 13 h, and 15 h were able to recover up to 90.7%, 95.1%, 98.1%, 97.5%, 103.1%, and 94.9%, respectively;

though more WEO or WCO could result in better improvement performance, an excessive amount of oil content would affect other properties of aged asphalt. There was a suitable oil content related to the aging degree of asphalt.

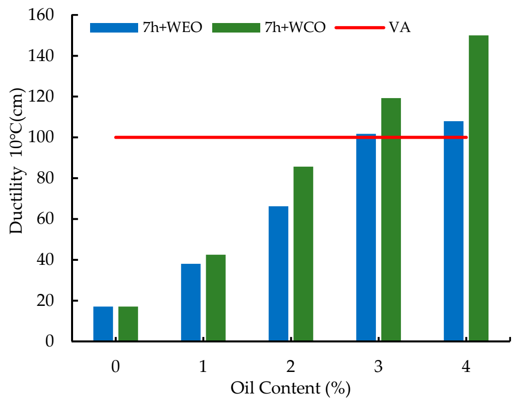
### *4.3. Ductility*

The test results of ductility at 10 °C were shown in Figure 8. Obvious differences could be observed between the effects of WEO and WCO on ductility at 10 °C. Regenerated asphalt with WCO had much better ductility than that with WEO, indicating that WCO could result in better low cracking resistance and temperature flexibility of aged asphalt binders.

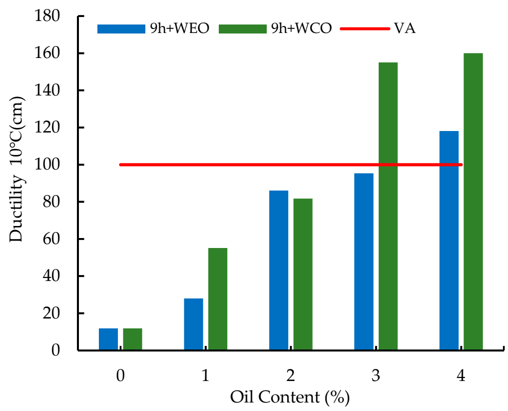
Compared with the VA, the ductility at 10 °C of 5-h- and 7-h- aged asphalts totally recovered up to the VA's ductility with 3% WEO content. When the WEO content was 4%, the 9-h-, 11-h-, and 13-h-aged asphalts achieved a ductility value also similar to that of the VA at 10 °C, while the WCO also had similar effects on ductility, although the influence of the WEO on the basic physical properties of aged asphalt was different. With 1% WCO content, the ductility for 5-h-aged asphalt recovered up to the VA's ductility value at 10 °C. For 7-h-, 9-h-, and 11-h- aged asphalts with WCO content of 3%, the ductility of the VA at 10 °C was also achieved. When the WCO content was up to 4%, the 13-h- and 15-h-aged asphalts also showed similar ductility with VA at 10 °C. It could therefore be concluded that WEO or WCO could improve the different property indexes of aged asphalt. After fully considering the performance of other aspects, it was concluded that, though more WEO or WCO could improve the low temperature performance, an excessive amount of oil content was not a favorable choice. There should be a suitable WEO or WCO content to give WEO and WCO better regeneration function.



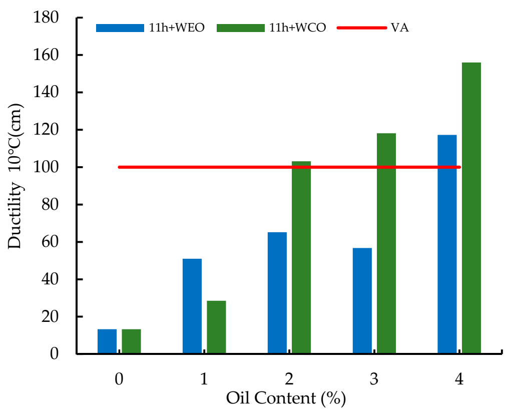

(**a**) Rejuvenated asphalt (5 h aging time of VA)




(**b**) Rejuvenated asphalt (7 h aging time of VA)

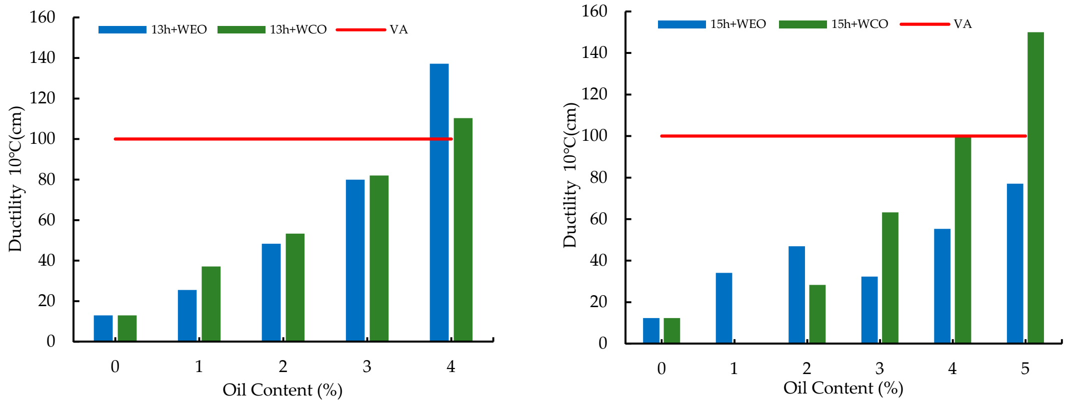



(**c**) Rejuvenated asphalt (9 h aging time of VA)




(**d**) Rejuvenated asphalt (11 h aging time of VA)

**Figure 8.** *Cont.*



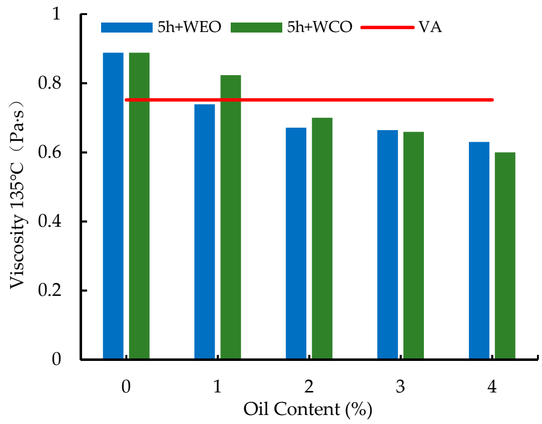

(**e**) Rejuvenated asphalt (13 h aging time of VA) (**f**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 8.** Effects of WEO and WCO on ductility of different aged asphalts at 10 °C.

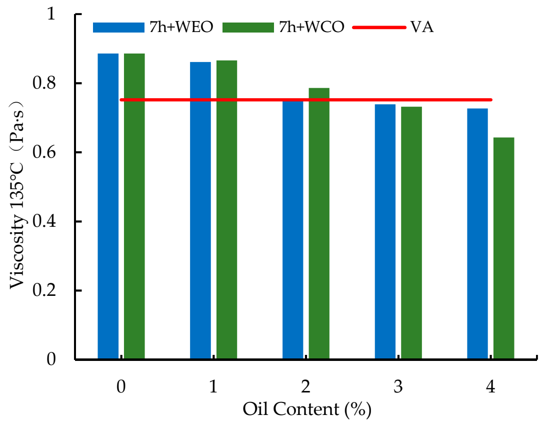
### 4.4. Viscosity

Viscosity reflected the frictional resistance between the internal molecules of the asphalt during flow deformation. According to mixing and compaction temperature of the asphalt mixture, the rotational viscosity of asphalt at 135 °C was close related to the frictional resistance between the internal molecules of the asphalt. The effects of WEO and WCO on the viscosity of aged asphalt at 135 °C with different aging are shown in Figure 6. All viscosity values decreased as the WEO or WCO content increased.

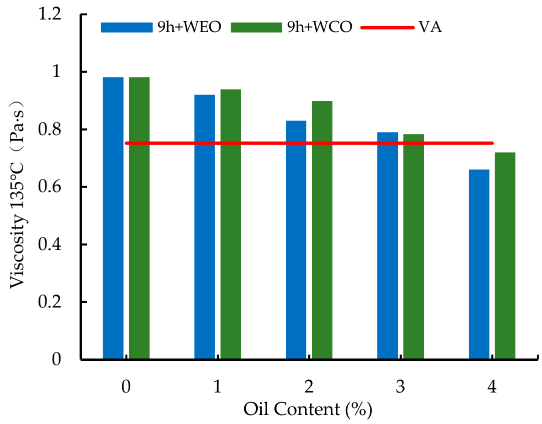
As shown in Figure 9, different aged asphalts could reach the 135 °C viscosity of VA with different oil contents. The 5-h-aged asphalt recovered up to 98.3% of the VA's viscosity at 135 °C with 1% WEO content. With 2% WEO content, it reached 100.3% for the 7-h-aged asphalt. For 9-h-aged asphalt and 3%WEO content, the 135 °C viscosity reached 105.1%. For 11-h- and 13-h-aged asphalts with WEO content of 3%, 97.7% and 91.8% of the VA were recovered, respectively. However, for the asphalt aged 15 h with WEO content of 5%, the viscosity could only reach 96.8%. The improvement effect on viscosity decreased gradually with increased aged asphalt. The WCO displayed similar effects on asphalts viscosity with 5-h-, 7-h-, 9-h-, 11-h-, 13-h-, and 15-h-aged asphalts containing 2%, 3%, 4%, and 5% WCO contents, recovering 93.1%, 97.3%, 104.1%, 101.7%, 98.3%, and 114.5%, respectively. Though more WEO or WCO could improve the frictional resistance of aged asphalt, excessive oil content would affect other properties of aged asphalt. If the viscosity of recycled asphalt becomes too high, the asphalt mixture would not be uniformly dispersed, segregation would then occur, which could adversely affects mixture performance. If the viscosity of recycled asphalt becomes too low, the asphalt roads would be prone to rutting. To satisfy the adhesion performance of recycled asphalt, the WEO or WCO dosage should be in a reasonable range.



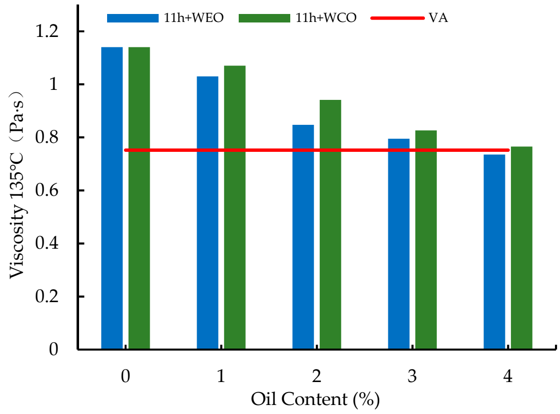

(**a**) Rejuvenated asphalt (5 h aging time of VA)



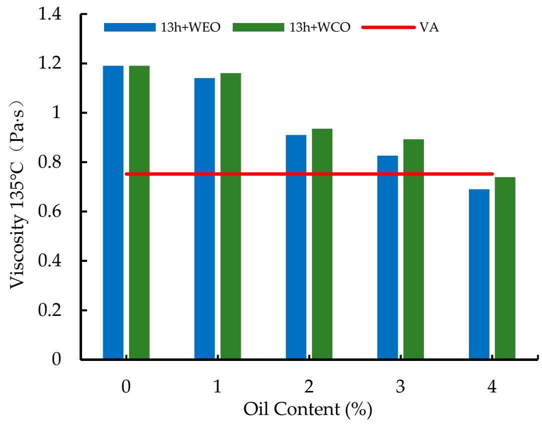

(**b**) Rejuvenated asphalt (7 h aging time of VA)



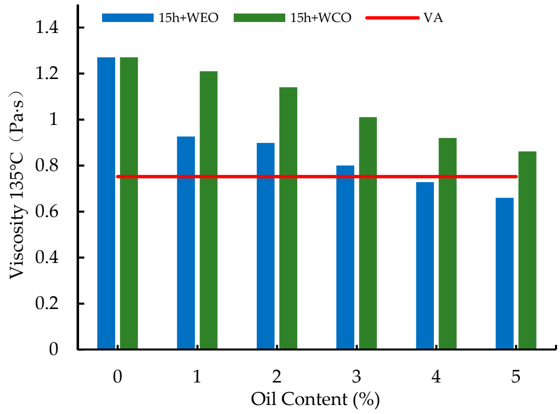

(**c**) Rejuvenated asphalt (9 h aging time of VA)




(**d**) Rejuvenated asphalt (11 h aging time of VA)




(**e**) Rejuvenated asphalt (13 h aging time of VA)




(**f**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 9.** Effects of WEO and WCO on viscosity of different aged asphalt.

Based on the physical test conclusions, WEO and WCO showed different effects on rejuvenation of aged asphalts. After a comprehensive analysis considering the regeneration effect of different aged asphalt, the proposed optimal ranges are shown in Tables 3 and 4.

**Table 3.** Effect of different WEO contents on physical properties in different aged asphalts.

| Aging Time | 5–7 h | 7–11 h | 11–13 h | >13 h |
|---|---|---|---|---|
| penetration | 1–2% | 1–2% | 2–3% | 3–4% |
| softening point | 1–2% | 2–3% | 2–3% | 3–4% |
| ductility | 1–3% | 3–5% | 4–5% | >5% |
| viscosity | 1–3% | 3–4% | 3–4% | >4% |

**Table 4.** Effect of different WCO contents on physical properties in different aged asphalts.

| Aging Time | 5–7 h | 7–11 h | 11–13 h | >13 h |
|---|---|---|---|---|
| penetration | 1–2% | 1–2% | 2–3% | 3–4% |
| softening point | 1–2% | 1–2% | 1–2% | >2% |
| ductility | 1–3% | 1–3% | 3–4% | >4% |
| viscosity | 1–3% | 3–4% | 3–4% | 4–5% |

From the above analysis, it could be observed that WEO or WCO could improve the different property index of aged asphalt. Generally, less WCO content could achieve the same regeneration effect when compared with WEO which ascertains that WCO was a better regenerative agent. According to the different aging time, aged asphalt could recover its basic physical performance with 1–5% WEO content and 1–4% WCO content (by mass of asphalt).

### *4.5. Chemical Properties*

#### 4.5.1. Four Components

Compared with 7–h-aged asphalt and VA, the components composition results of rejuvenated asphalt with 3% WEO and 2% WCO are shown in Table 5. Correspondingly, for the components of aged asphalt, VA had higher saturates and aromatics, but less resins and asphaltenes which was the same with changing rule of aged asphalt. It also illustrated that the percentages of components composition of aged asphalt could be slightly changed by WEO and WCO. Moreover, compared with common aged asphalt, the waste oils could slightly reduce the asphaltenes and increase aroma components in aged asphalt the result was drawn from the 7-h-aged asphalt; similar conclusions could be speculated with longer aging time.

**Table 5.** Four components of VA, aged asphalts, and rejuvenated asphalts.

| Asphalt | VA | Aged Asphalt (7 h) | Rejuvenated Asphalt (3% WEO) | Rejuvenated Asphalt (2% WCO) |
|---|---|---|---|---|
| Saturates (%) | 5.68 | 4.25 | 5.93 | 6.22 |
| Aromatics (%) | 61.59 | 51.96 | 48.51 | 46.57 |
| Resins (%) | 20.01 | 26.33 | 29.19 | 31.57 |
| Asphaltenes (%) | 12.73 | 17.46 | 16.37 | 15.64 |

On the macrolevel, penetration, softening point, ductility, and viscosity of aged asphalt showed a decrease in varying aging degrees. Therefore, the different changes of the four components were consistent with asphalt physical properties. From the view of decreasing asphaltenes and aromatics content and increasing resins and saturates content of rejuvenated asphalt, waste oil had an obvious effect on recycled aged asphalt, which made it possible to use it as a regenerator. In addition, WCO could achieve better regeneration effect than that of WEO with less content as proven earlier.

#### 4.5.2. Functional Groups

The rejuvenated asphalt with 2% WCO was selected for the FTIR microexperiment. Based on the results of the four components, if the rejuvenated asphalt with WCO could improve the asphalt's properties, then so should the rejuvenated asphalt with WEO. The infrared spectroscopy analysis curves of VA, aged asphalt (7 h) and rejuvenated asphalt at wavenumbers between 4000 $cm^{-1}$ and 500 $cm^{-1}$ are shown in Figure 10.

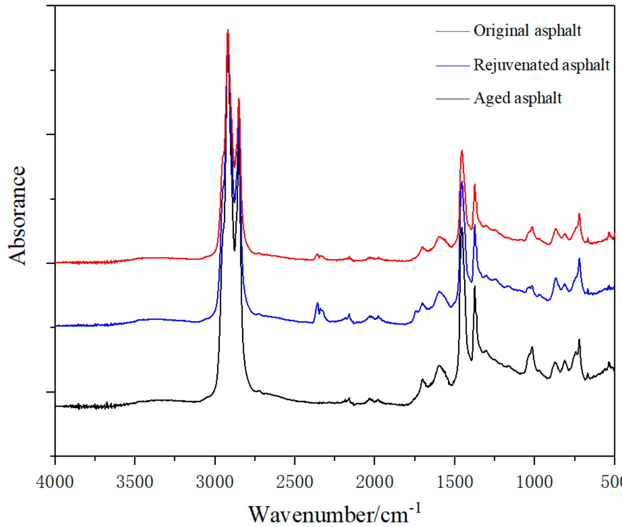



**Figure 10.** FTIR spectra results of VA, aged asphalt, and rejuvenated asphalt.

As illustrated in Figure 10, the absorption peaks of the aged asphalt (AH-70) and the VA (AH-70) were similar. However, the tensile vibration absorption peak of methane –CH2– of aged asphalt at 2923 $cm^{-1}$ and 2851 $cm^{-1}$ was enhanced. Carbonyl (C=O) and sulfoxide (S=O) peak area intensities reflected the aging and rejuvenating degree of asphalt. Compared with the VA, aged asphalt exhibited a notable increase in C=O (1600 $cm^{-1}$) and S=O (1031 $cm^{-1}$) peak areas. The absorption peaks indicating aromatics (813 $cm^{-1}$ and 865 $cm^{-1}$) were also enhanced.

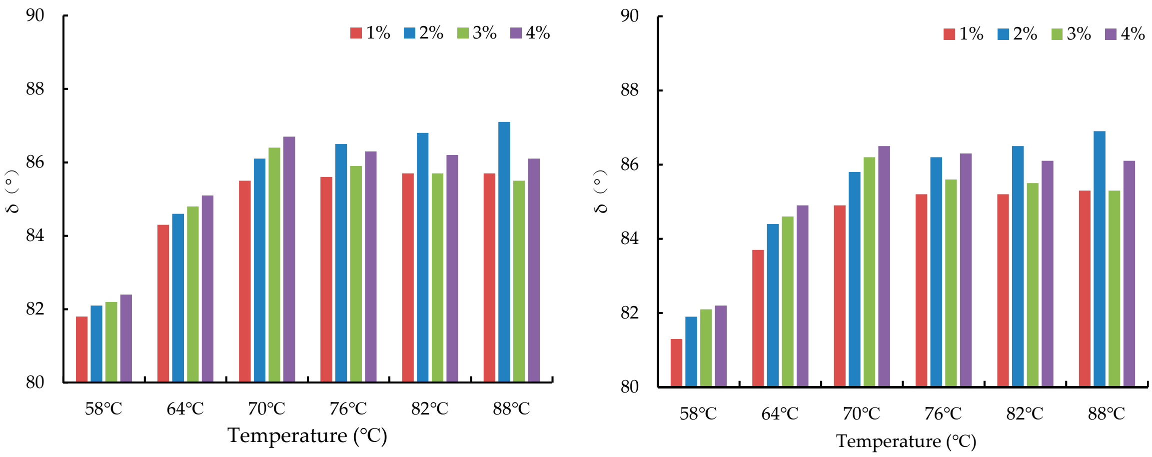
The absorption peaks of the rejuvenated asphalt and the VA were also basically similar, while their aromatic absorption peaks were also significantly higher than that of the aged asphalt. However, the rejuvenated asphalt had two new absorption peaks, which appeared at 1746 $cm^{-1}$ and 1164 $cm^{-1}$, belonging to C=O stretching of carbonyl. The new absorption peaks are the characteristic peaks of soybean oil. Moreover, the oxygenated function (C=O and S=O) peak area intensity of the rejuvenated asphalt showed a notable decrease in carbonyl peak compared to that of aged asphalt. This was due to the proportionality of the decrease in intensity of band from C=O and S=O for recycled asphalts, and decrease in asphaltene content had an effective impact on rejuvenated asphalt performance with WEO or WCO.

### 4.6. Rheological Properties

#### 4.6.1. Phase Angle (δ)

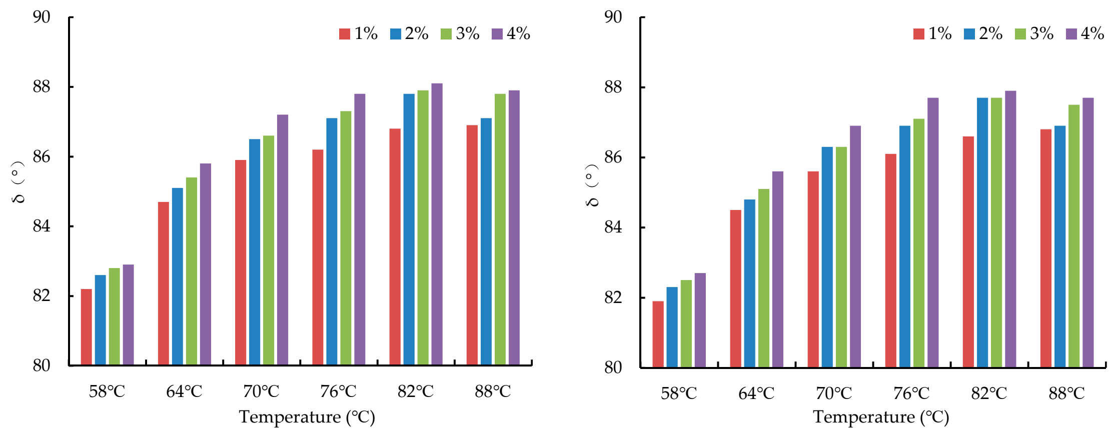
The temperature dependence of the phase angle of rejuvenated asphalts is illustrated in Figures 11 and 12. It revealed that viscous property performance increased with increasing temperature. Also, the phase angle increased with increasing waste oil content. Compared with the 5 h aging time, it had much better improvement than that of 15 h aging time at the same temperature and waste oil content.

Meanwhile, different elastic recovery performance occurred with WEO and WCO. At the same temperature and oil content, rejuvenated asphalt with WCO had a slightly larger phase angle than that of WEO. Asphalt with a smaller phase angle has better elastic recovery, and WEO had better impact on elastic recovery performance than WCO. With the temperature increasing, all the phase angles increased, but the rejuvenated asphalt with WCO had a much more stable change. Thus maintaining stable elastic recovery performance and reducing the occurrence of damages due to the viscous response change. The phase angles increased obviously when the temperature increased from 58 °C to 70 °C. There was no significant difference when the oil content was 2%, 3%, and 4% with the temperature rising. This indicated there would be a suitable dosage for better elastic recovery performance of aged asphalt which depended on the aged asphalt types and field construction requirements.




(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 11.** Influences of WEO on phase angel of rejuvenated asphalt (different aging time of VA).



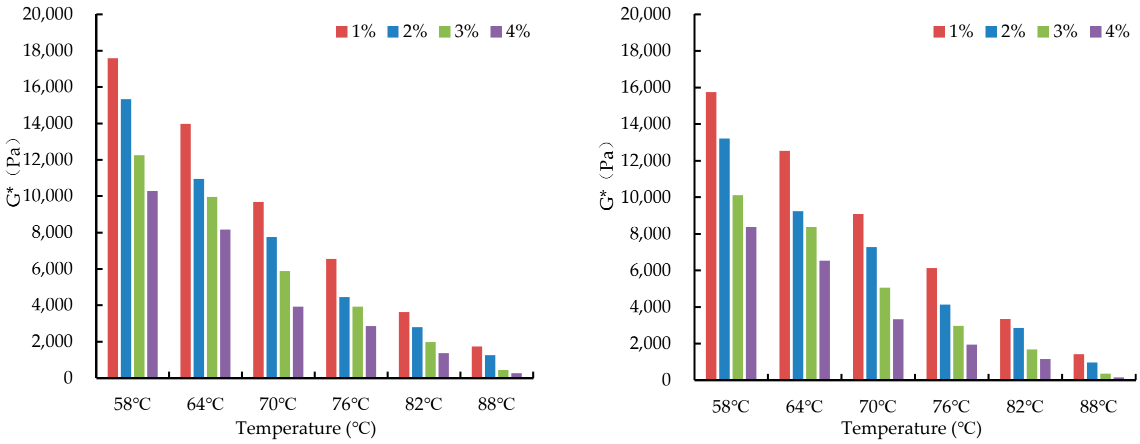

(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 12.** Influences of WCO on phase angel of rejuvenated asphalt (different aging time of VA).

#### 4.6.2. Complex Modulus (G*)

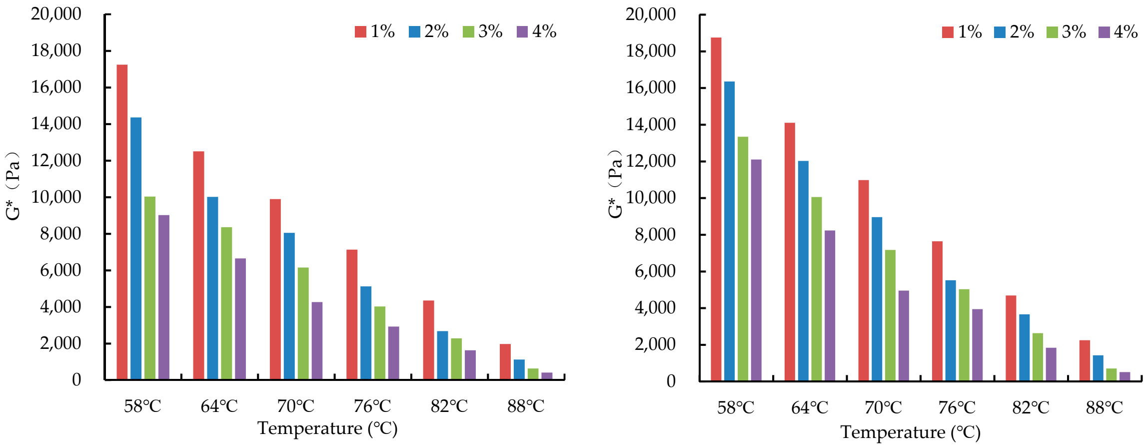
Asphalt with large complex modulus has better resistance to flow deformation. Figures 13 and 14 illustrated the effects of WEO, WCO, and different dosages on the complex modulus of rejuvenated asphalts at different temperatures.

It was well-known that the higher the |G*|, the higher the resistance to flow deformation. As seen in Figures 15 and 16, the complex modulus of rejuvenated asphalt with WEO and WCO had the similar trend. Complex modulus decreased as the oil content and temperature increased, which appeared much more obvious after the temperature exceeded 76 °C. With the temperature rising, all the resistance to flow deformation of rejuvenated asphalts kept decreasing; especially when the oil content was larger than 3%. At the same time, the corresponding anti-rutting performance of rejuvenated asphalt was deteriorating. However, when the temperature was above 76 °C, the rutting factor of rejuvenated asphalt was close to that of the aged asphalt. This indicated that the effect of waste oil would be weakened at high temperature. Therefore, a proper range should be selected to get better high temperature performance for rejuvenated asphalts with WEO or WCO.




(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 13.** Effects of WEO on complex modulus of rejuvenated asphalt (different aging time of VA).




(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 14.** Effects of WCO on complex modulus of rejuvenated asphalt (different aging time of VA).

In addition, with the aging time increasing, complex modulus of both rejuvenated asphalts decreased notably. A similar trend with rutting resistance factor was observed. Though the WEO and WCO had a similar trend, the WCO-rejuvenated asphalt kept higher complex modulus at the same temperature and aging time, which would show good performance at high temperatures. The conclusions also confirmed the previous analysis of the different regeneration effect on WEO and WCO in different contents.

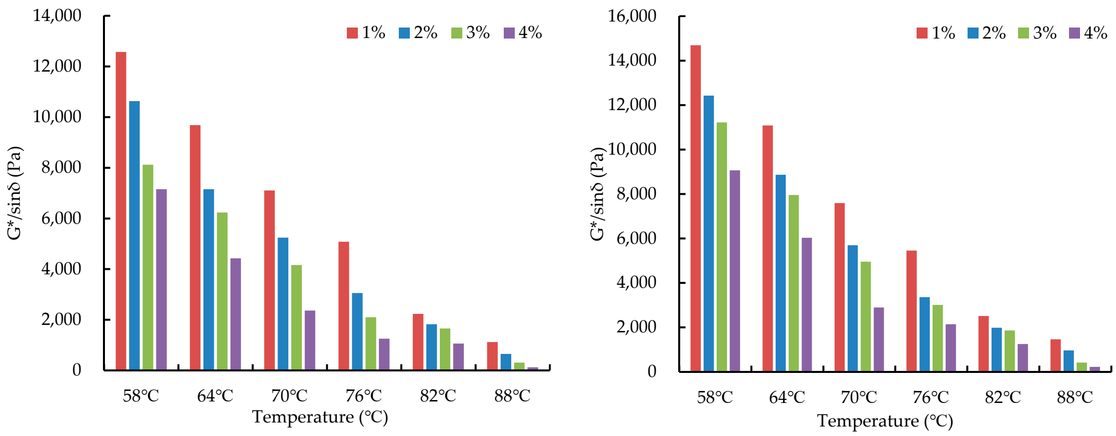
4.6.3. Rutting Resistance Factor (G*/sinδ)

The test results of rutting resistance factor of rejuvenated asphalt with WEO and WCO at 5 h and 15 h are presented in Figures 15 and 16, respectively. WCO and WEO affect the rutting resistance of the rejuvenated asphalt by improving its viscosity. As shown in Figures 15 and 16, the rutting resistance factor decreased with increasing temperature. With the same temperature, the rejuvenated asphalt of aging 15 h had smaller rutting resistance than that of aging 5 h, which proved that longer aging time had a negative influence on the high-temperature property. At the same aging time, the rutting resistance factor decreased notably with the content and temperature increasing.

When the temperature was up to 70 °C, the rutting resistance factor reduced notably and it decreased sharply with a temperature higher than 76 °C. Therefore, the increase in waste oil content did not lead to a larger rutting resistance factor. Though the WCO and WEO were much more important for property recovery of aged asphalt and high-temperature performance of rejuvenated asphalt, there should be reasonable choice according to the asphalt performance requirements and maximum temperature in the construction region.

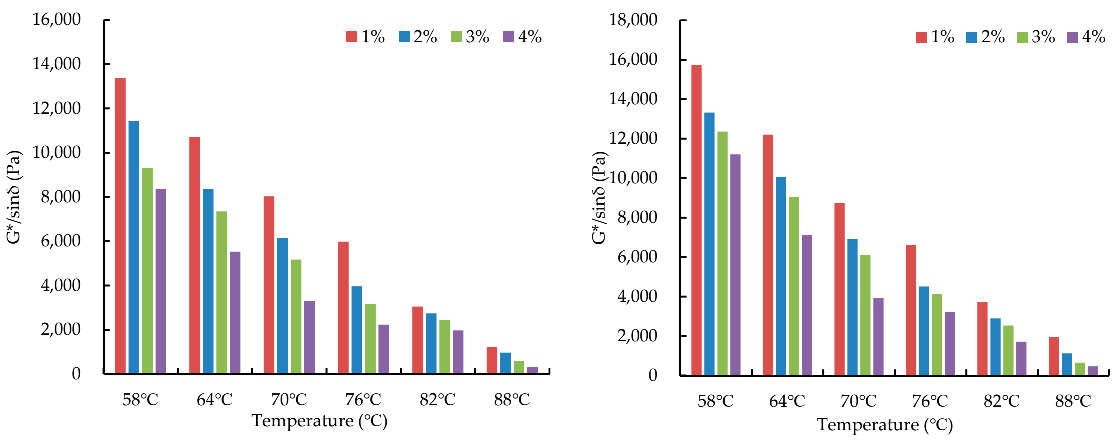
There were different reductions for rejuvenated asphalt in rutting resistance factor. At the same temperature, aging time and waste oil dosage, rejuvenated asphalt with WEO had much smaller rutting resistance factor than that with WCO. With temperature increasing, the decreasing extent of rejuvenated asphalt with WCO was lower than that with WEO, especially when the temperature was higher than 76 °C (this was similar with the conclusions of softening point and viscosity). Meanwhile, this illustrated that the WCO and WEO could both recover part of the performance of aged asphalt, but WCO could achieve the same results with less content. The conclusions confirmed the previous analysis which WCO had better regeneration effect than WEO with different aged asphalts.

In addition, rutting resistance factor reflected unrecoverable deformation of asphalt during the loading process. At high temperature, bitumen with higher G*/sinδ but smaller flow deformation would have much higher rutting resistance. All these facts indicated that the rutting resistance had satisfactory improvement with waste oil content range of 1 to 3%. When the dosage was more than 3% and the temperature was higher 76 °C, the WCO could keep better high temperature property. Therefore, the range of 1 to 3% was a more suitable dosage with WEO and WCO for better high temperature performance, which depended on the aged asphalt condition and actual construction requirements.




(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 15.** Influences of WEO on rutting resistance factor of rejuvenated asphalt (different aging time of VA).




(**a**) Rejuvenated asphalt (5 h aging time of VA) (**b**) Rejuvenated asphalt (15 h aging time of VA)

**Figure 16.** Influences of WCO on rutting resistance factor of rejuvenated asphalt (different aging time of VA).

### 4.7. Scanning Electron Microscopy (SEM) Test

The SEM device is shown in Figure 17. The images of VA, aged asphalt, and rejuvenated asphalt are shown in Figure 18. As seen from Figure 18a, VA had good homogeneity and was close to a homogeneous structure. After 7 h of aging time, some small folds and little impurities could be found at some surfaces, which proved the asphalt structure was affected. When the WCO was added into the aged asphalt, there was not obvious change in the appearance compared to the aged asphalt. While some little tiny white points could be seen in some asphalt surface, which appeared as extremely small particles of WCO related to the filtered sieve holes during the lab test preparation. What is more, it speculated that WEO had the same situation. As seen in Figure 18c, though rejuvenated asphalt was apparently no longer homogeneous, its properties were improved which was proven by the previous test results. It illustrated that WEO or WCO collected from the factory could be regenerative agents after concentrate filtering system, and the existing extremely small particles had little effect on the performance improvement.

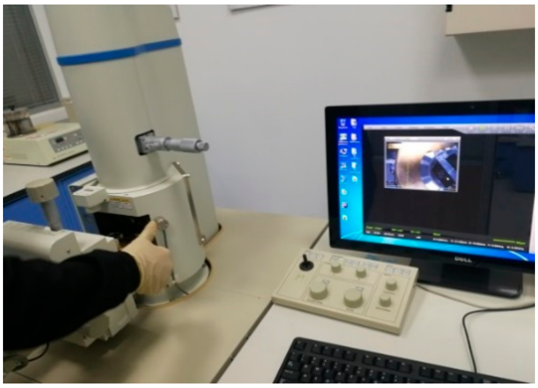
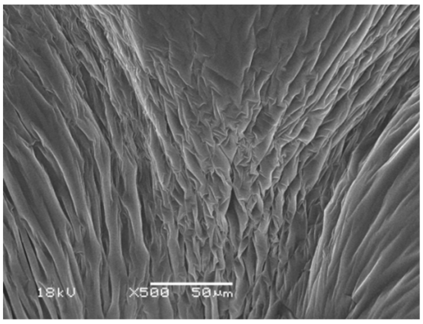
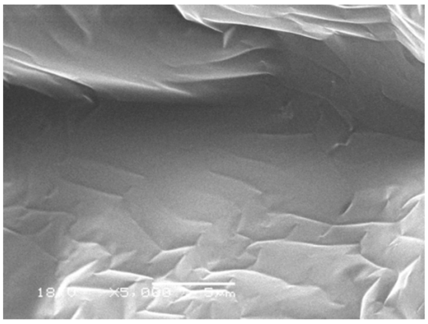


**Figure 17.** Scanning electron microscope test device.

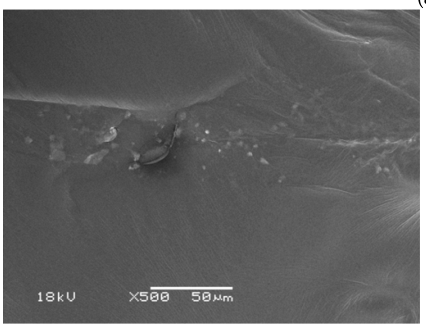



× 500 times

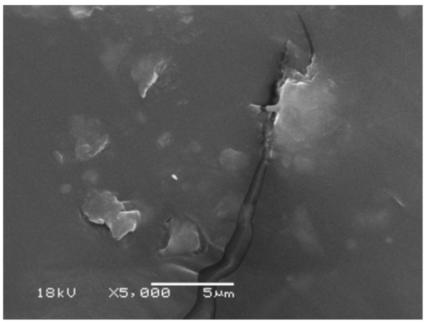


× 5000 times

(**a**) VA



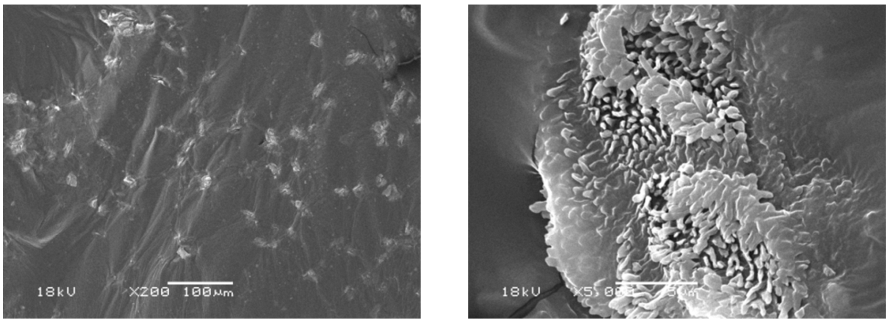

× 500 times




× 5000 times

(**b**) Aged asphalt (7 h)

**Figure 18.** *Cont.*




× 200 times × 5000 times

(**c**) Rejuvenated asphalt (2% WCO)

**Figure 18.** The SEM images of different asphalt states.

## 5. Conclusions

In this study, some conventional and specific experiments were conducted to evaluate the regeneration properties of the aged asphalts. Based on the experiments results and analysis from different aged asphalts, the following conclusions can be drawn.

(1) WEO and WCO could improve the basic physical properties of aged asphalts to the normal level. To satisfy better performance of regenerated asphalt, the WEO or WCO dosage should be in a reasonable range, which was different for different aged asphalts.

(2) In comparison, WCO had better function than WEO as a regenerative agent. Less WCO content could achieve better regeneration properties. According to the different aging time, aged asphalt could recover its basic physical performance with 1–5% WEO content and 1–4% WCO content (by mass of asphalt).

(3) The addition of WEO or WCO could increase saturate, resin, and asphaltene content and decrease aromatics dosage, intensity of carbonyl, and sulfoxide of aged asphalt, but it could not change its colloidal structure. Moreover, there was a suitable oil content related to the aged asphalt condition and actual construction requirements. An excessive amount of oil content would affect other properties of aged asphalt.

(4) The rheological properties of aged asphalts could also be regained to normal level of VA with different contents of WEO or WCO. Overall, WEO or WCO were adaptable regenerative agents to improve aged asphalt performances. It provided a new application method for WEO or WCO in asphalt pavement recycling field, which realized the wastes cyclic utilization and environmental protection.

## 6. Future Researches

The scope of this study proved the adaptability of WEO and WCO for aged asphalt through basic properties, partial rheological, and chemical characterization. This study helps form the foundation to develop a new type of regenerative agent with WEO and WCO as the basis materials. Some other performances, such as resistance to fatigue cracking and moisture sensitivity, were not evaluated. Also, it was uncertain whether waste oil would affect the bonding ability of asphalt and aggregates. The research team is conducting these above related experiments and performance detection of re-aging asphalt. After obtaining the test data, a new regenerative agent will be developed. Afterwards, it will be used in the hot in-place recycling field and related performance indexes will be evaluated. Based on the testing results, components will be adjusted to obtain better asphalt property. Finally, a stable regenerative agent product will be formed by recycling waste oil.

**Author Contributions:** All authors discussed extensively. H.L. and B.D. conceived the experiments. H.L., B.D., P.G., and G.Z. conducted the experiments. H.L., W.W., G.Z., and Q.M. analyzed the results. All authors reviewed the manuscript.

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**Conflicts of Interest:** The authors declare no conflicts of interest.

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