

## Research Article

# Properties and Components of Recycled Engine Oil Bottom Rejuvenated Asphalt and Its Grey Relationship Analysis

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Numerous environmental pollution and resource waste problems are associated with recycled engine oil bottom (REOB), which cannot be effectively recycled. Based on the similarity compatibility theory and component adjustment theory, comparing the physical and rheological properties of laboratory-aged asphalt under three types of REOB (defined as REOB-1, REOB-2, and REOB-3) with different dosages, the optimum type and dosage of REOB as asphalt regenerant were explored. The rejuvenation mechanism of REOB on aged asphalt was revealed by combined performance, four-component, and infrared spectroscopy analyses. The relationship between the four components and physical rheological indexes in the process of asphalt rejuvenated by REOB was quantitatively obtained by the grey relationship analysis. The results show that only REOB-3 with a dosage of 7% on the aged asphalt has the best comprehensive rejuvenation effect. Also, the high-temperature rutting resistance of rejuvenated asphalt with 7% REOB-3 is better than that of the original asphalt, but the low-temperature flexibility and the crack resistance performance have yet to be improved. The mechanism through which REOB rejuvenates aged asphalt is an incomplete component adjustment; some of the components undergo physical or chemical reactions and transformations. Accordingly, the asphaltene content and the intensity of sulfoxide functional groups in aged asphalt decrease, thereby achieving rejuvenation gradually with the addition of REOB. A grey relationship analysis demonstrates that asphaltenes have the greatest influence on high-temperature performance and that low-temperature performance requires a reasonable combination of four components. Moreover, a comprehensive advantage analysis reveals that REOB is the most sensitive to the softening point and that the asphaltene content has the greatest influence on the physical and rheological properties of REOB-rejuvenated asphalt. Therefore, the asphaltene content should be strictly controlled during the addition of REOB to rejuvenate aged asphalt.

## 1. Introduction

Oxidation, product wear, pollutant mixing, additive consumption, and water absorption all occur throughout the use of engine oil, leading to the decline in the functionality of the oil; unfortunately, these processes invalidate the production of recycled engine oil (REO). In most developed industrial countries at present, REO is primarily subjected to standardized recycling into fuel oil or lubricant oil through various recovery technologies, including ultrafiltration, centrifugal separation, molecular distillation, flocculation treatment, and solvent refining [1]. However, the traditional sulfuric acid-clay regeneration process is still widely used in

China, although some improved regeneration processes, such as distillation-clay refining [2] and distillation-hydrotreating [3], have also been developed in recent years; these new technologies not only improve the quality and efficiency of REO regeneration but also reduce the secondary pollution of acid residue, wastewater, and waste gas in traditional recycling process [1]. Generally, 70–80% of REO can be effectively recycled by the aforementioned processes. However, the remaining residue (accounting for 20–30%) cannot be effectively recycled due to the presence of many impurities; this residue is ultimately called recycled engine oil bottom (REOB). Various functional additives in engine oil containing sulfur, calcium, zinc, phosphorous, and

molybdenum in addition to oxidation products and metals (including mainly iron and copper) that become worn during engine operation all remain in REOB after these regeneration processes; consequently, REOB cannot be effectively utilized. Currently, REOB is mainly treated by directly discarding, burying, or burning the residue, all of which lead to irreparable pollution of the soil, water, and atmosphere; this pollution is detrimental to human health and could even cause cancer once it comes into contact with the human body. Therefore, REOB has been listed as one of the three main control points in the field of environmental protection for the 21st century, and thus, an effective recycling and treatment procedure for REOB is urgently needed [1, 2].

Research on the use of REO and REOB in asphalt materials has already begun both domestically and internationally. Villanueva et al. [4] used different REO contents to soften asphalt and found that the addition of REO increases the penetration of asphalt, decreases the softening point, and reduces the high-temperature performance of asphalt after softening but also significantly improves the low-temperature performance. Through a rheological test on Trinidad Lake Asphalt mixed with different amounts of REO, Ackbarali and Maharaj [5] proved that REO can be used as a modifier to soften hard asphalt. Furthermore, Dedene [6] used REO to modify reclaimed asphalt binders and carried out rutting tests and tensile strength ratio tests on the corresponding asphalt mixture; the results showed that REO can be used as a rejuvenating agent to chemically restore the properties of asphalt pavements containing reclaimed asphalt pavements. In addition, Jia et al. [7] studied the infrared spectra and rheological properties of different asphalt binders containing REO and analysed the microcosmic interactions of REO and different types of asphalt binders. Moreover, Xu [8] studied the pavement performance of an REO-rejuvenated asphalt mixture and analysed its economic and social benefits. In conclusion, research on the modification or rejuvenation of asphalt binders with REO has achieved moderate success. The technology through which REO is employed to regenerate lubricating oil advances daily, and the production source of lubricating oil depends largely on crude oil [1]. Currently, the application of REO to regenerate lubricating oil is more economical, energy-friendly, and promotional than the application of REO in asphalt materials. However, an effective method for utilizing the REOB that remains after the regeneration of REO has yet to be developed, and thus, it will be more necessary to develop application prospects for REOB by conducting research on the use of REOB in asphalt materials.

Through X-ray fluorescence analysis in Canada, Hesp and Shurvell [9] confirmed that REOB can be used in asphalt binders and discussed the adverse effects of REOB on the early cracking of asphalt pavement. Rubab et al. [10] conducted rolling thin film oven (RTFO) and pressure ageing vessel (PAV) tests after mixing REOB with two different types of asphalt binders in Canada and found that REOB may accelerate the oxidation speed of modified asphalt while improving the performance grade of asphalt binders. Ding

et al. [11] used REOB produced by vacuum distillation and atmospheric distillation processes in China to modify straight asphalt; by employing the extended ageing test, it was found that REOB is unfavourable to the ductile fracture performance and cracking resistance performance of asphalt at low temperatures. However, previous studies on REOB are limited to the modification of an asphalt matrix. Moreover, insufficient research has been conducted on the relationship and correlation mechanism between REOB and asphalt binders. In addition, the properties of REOB may vary widely with source and regeneration process, thereby affecting asphalt modification and action performance. Therefore, it is necessary to carry out systematic studies on several types of REOB used for aged asphalt, particularly because the application of REOB to the rejuvenation of aged asphalt can broaden the effective utilization of REOB and have great economic and engineering significance for reclaimed asphalt pavement (RAP).

In this paper, three types of REOB were selected from the recycling plant. Physical and rheological tests were carried out in addition to four-component and infrared spectroscopic analyses on aged asphalt containing various REOB contents. Upon comparing and analysing the rejuvenation effects of aged asphalt blended with different dosages of REOB, the correlation mechanism between REOB and aged asphalt was revealed based on the combined performance, component, and structure. Furthermore, the relationship between the properties and components of rejuvenated asphalt with REOB was established by means of grey relationship analysis. To explore the feasibility of utilizing REOB for rejuvenating asphalt, this study provides a basis for further research into the application of waste oil to rejuvenate asphalt binders.

## 2. Materials and Methods

**2.1. Raw Materials.** In China, the chemical compositions and properties of REOB produced by different REO regeneration processes may vary greatly due to the different sources of REO and the different degrees of oxidation and wear during their use. Three types of REOB were recovered from a large REO treatment plant after pretreatment and atmospheric and vacuum distillation. Their basic properties and chemical components are shown in Table 1, respectively. The 70-A petroleum asphalt binders, which are widely used in North China, were selected as the control binders; their main technical properties are shown in Table 2.

**2.2. Preparation of the Aged Asphalt Binders.** According to the standard highway engineering testing specification of China for asphalt and asphalt mixtures (JTG E20-2011), an RTFO test was used to simulate the short-term ageing of the original asphalt mixture during mixing and paving. Similarly, a PAV test was used to simulate the long-term ageing of an asphalt pavement in service for 5 years [12]. Finally, the aged asphalt binders were obtained by implementing a PAV test after an RTFO test on the original 70-A asphalt binders.

TABLE 1: Basic properties and chemical components of three types of REOB.

Type	Colour	Viscosity (60°C, cSt)	Density (25°C, g/cm <sup>3</sup> )	Asphaltenes (%)	Resins (%)	Aromatics (%)	Saturates (%)
REOB-1	Brown	293	0.873	0.9	6.8	73.1	19.2
REOB-2	Black brown	535	0.912	8.1	0.1	91.7	0.1
REOB-3	Reddish brown	340	0.885	0.2	15.5	83.5	0.8

TABLE 2: Properties indices of 70-A matrix asphalt binders.

Indices	Test results	Engineering requirements	Test methods	
Penetration (25°C, 0.1 mm)	71	60~80	JTG E20-2011 T0604	
Softening point (°C)	48.7	≥46	JTG E20-2011 T0606	
Ductility (10°C, cm)	22	≥20	JTG E20-2011 T0605	
Brookfield viscosity (135°C, Pa·s)	0.397	—	JTG E20-2011 T0625	
	Mass loss (%)	-0.12	-0.8~0.8	JTG E20-2011 T0610
After RTFO (163°C, 75 min)	Penetration ratio (%)	65.3	≥61	JTG E20-2011 T0604
	Ductility (10°C, cm)	7.2	≥6	JTG E20-2011 T0605

**2.3. Preparation of the Rejuvenated Asphalt Binders.** The aged asphalt binders were heated to 150°C in an oven while adding 3%~8% (mass ratio of aged asphalt) REOB with an interval of 1%. To prepare different proportions of REOB-rejuvenated asphalt during this preparation process, the blended samples consisting of aged asphalt binders and REOB were continuously stirred by a glass rod until the REOB was evenly dispersed within the aged asphalt binders.

**2.4. Physical Property Tests.** To evaluate the influences of the various physical characteristics of REOB on aged asphalt, the conventional physical indices of aged asphalt containing REOB of different contents, including the 25°C penetration, softening point, and 10°C ductility, were tested in accordance with JTG E20-2011 specifications T0604, T0606, and T0605.

**2.5. Rheological Property Tests.** The rutting factor ( $G^*/\sin \delta$ ) obtained by a dynamic shear rheometer (DSR) was used to characterize the high-temperature stability of different proportions of REOB-rejuvenated asphalt according to JTG E20-2011 T0628; the rejuvenated asphalt was measured at a fixed frequency of 10 rad/s with increments of 6°C in the strain control mode. Moreover, the creep stiffness ( $S$ ) and creep rate ( $m$ ) obtained by a bending beam rheometer (BBR) were used to characterize the low-temperature crack resistance of the aged asphalt binders with different content of REOB in accordance with JTG E20-2011 T0627, in which the test temperatures were -12°C, -18°C, and -24°C.

**2.6. Four-Component Analysis.** Four-component analysis was carried out on the asphalt binders and REOB by using the rod-thin-layer chromatography/hydrogen flame method. The specific testing steps are described as follows. The samples were dissolved in toluene and prepared in solutions of 30 mg/mL. Next, samples 0.8–1.0  $\mu$ L in volume were placed into the 15 mm chromatographic rod five to six times, and the diameter of each point sample was controlled at 1–3 mm. Then, the chromatographic frame was placed into

three types of mixtures composed of *N*-heptane, toluene, and toluene/ethanol (volume ratio 55:45) and expanded in sequence on the expansion table. After each expansion, the chromatographic frame was dried, kept warm, and moisturized until the next expansion. Finally, after three rounds of expansion, the chromatographic frame was tested and analysed by a rod-thin-layer chromatography analyser.

**2.7. Infrared Spectra Test.** The molecular structures and functional groups of asphalt binders and REOB were analysed by Fourier transform infrared (FTIR) spectrometry. The specific steps of the test are described as follows. The samples were dissolved in carbon disulfide (CS<sub>2</sub>), an organic solvent, with a concentration of 5 wt.%. Then, one drop of the prepared solution was dropped into a small groove with a diameter of 10 mm on a potassium bromide wafer. After evaporating the CS<sub>2</sub>, the sample film was tested by FTIR spectrometry. The test employed scanning wavenumbers of 4000–500 cm<sup>-1</sup>, and a total of 64 scans were performed.

### 3. Results and Discussion

#### 3.1. Physical Properties

**3.1.1. Penetration.** As one of the important classification indices of asphalt binders, the penetration, which represents the conditional viscosity of an asphalt binder at a specified temperature, can reflect the degree of softening and consistency of an asphalt binder. Figure 1 shows the influence of three types of REOB on the penetration of aged asphalt under different contents. Figure 1 explains that the penetration of the aged asphalt binders increases with the addition of all three types of REOB, which indicates that REOB can soften aged asphalt and reduce its consistency and viscosity. However, the improvement effect of the three types of REOB differs. Among the types of REOB analysed, the increase degree of REOB-1 is the largest, and the rate of increase is basically unchanged throughout the process; the increase degree of REOB-2 is the smallest, and when both the content of REOB-2 and REOB-3 ranges from 3% to 6%, the rate of increase is obviously higher than that when the

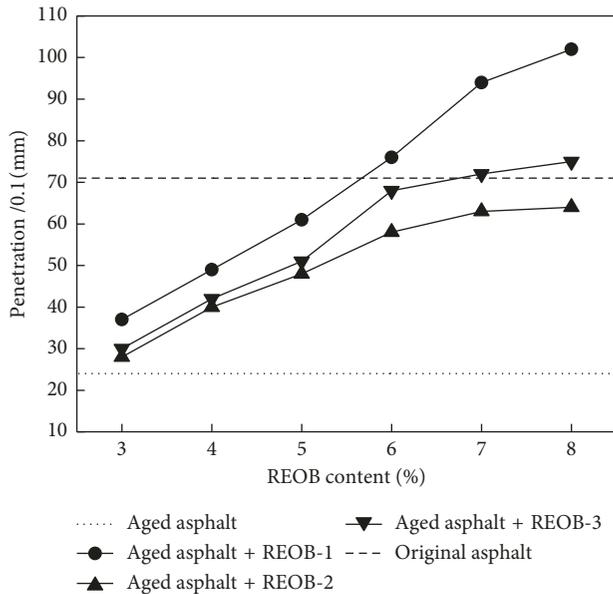


FIGURE 1: Influence of three types of REOB on penetration of aged asphalt.

content ranges from 6% to 8%. Furthermore, when REOB-1 content is 6% and REOB-3 content is 7%, penetration can be restored to the original asphalt level; when REOB-2 content is more than 6%, penetration tends to be stable and lower than that of the original asphalt. These differences are observed because the viscosities of all three types of REOB are lower than that of aged asphalt; thus, REOB can infiltrate deeper to aged asphalt, thereby softening and increasing the overall penetration.

**3.1.2. Softening Point.** The softening point represents the temperature of an asphalt binder when it reaches a certain viscosity under a particular test condition. A higher softening point represents a greater asphalt viscosity and a better stability at high temperatures. The influences of the three types of REOB on the softening point of aged asphalt are illustrated in Figure 2, which demonstrates that the softening point of aged asphalt binders decreases with an increase in REOB dosage ranging from 3% to 7%. Moreover, when the dosage of REOB-1 reaches 6% and the dosage of REOB-3 reaches 7%, the softening points of the corresponding aged asphalt binders decrease to nearly the level of an original 70-A asphalt, thereby showing that the high-temperature viscosity and stability of binders are reduced by the addition of REOB. When the three REOB contents all reach 8%, the softening point of aged asphalt continued to decrease with REOB-2, but the decrease rate of that obviously slowed; conversely, both REOB-1 and REOB-3 increase the softening point of binders to a certain extent, indicating increases in both high-temperature stability and viscosity. However, there is a difference between the penetration analysis in Section 3.1.1 and the above analysis at an 8% dosage of both REOB-1 and REOB-3. The possible reason for this difference is that an oil film forms on the surface of aged asphalt samples containing 8%

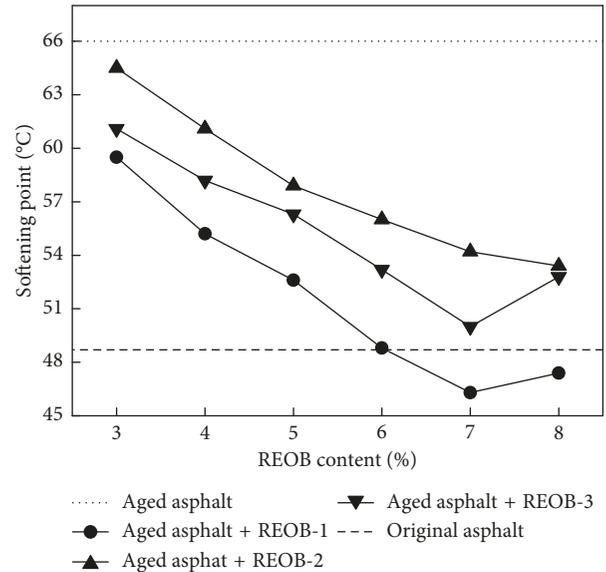


FIGURE 2: Influence of three types of REOB on softening point of aged asphalt.

REOB-1 or 8% REOB-3 that are kept at room temperature over a period of time. As a result, the penetration value is affected by the thickness of the surface oil film, and the formation of the surface oil film has an impact on the softening point of asphalt binders.

**3.1.3. Ductility.** The ductility is an index used to evaluate the plastic deformation capacity of asphalt; this index can measure the ability to undergo tensile deformation without being destroyed under an external force. The influences of three types of REOB on the ductility of aged asphalt are displayed in Figure 3, which shows that the ductility of aged asphalt binders with an REOB dosage ranging from 3% to 7% increases with an increase in REOB dosage, revealing that the plastic deformation and crack resistance of binders at low temperatures will gradually increase with the increase in REOB dosage. In addition, when the REOB content reaches 7%, the ductility of aged asphalt with REOB-3 increases to nearly the level of an original 70-A asphalt; REOB-1 maximizes the ductility of aged asphalt but cannot meet the requirements of the asphalt pavement specification. However, when the amount of REOB reaches 8%, the ductility of aged asphalt with REOB-2 continues to increase but remains much lower than that of the original asphalt. Additionally, the ductility of aged asphalt binders containing either 8% REOB-1 or 8% REOB-3 suddenly decreases, which clearly indicates that the low-temperature crack resistance of binders is poor, which is not in accordance with the technical pavement performance requirements.

**3.1.4. Determination of the REOB Optimal Type and Dosage.** A comprehensive analysis of the penetration, softening point, and ductility indices shows that the three types of REOB have a certain recovery effect on the physical properties of aged asphalt and that the recovery effect is closely

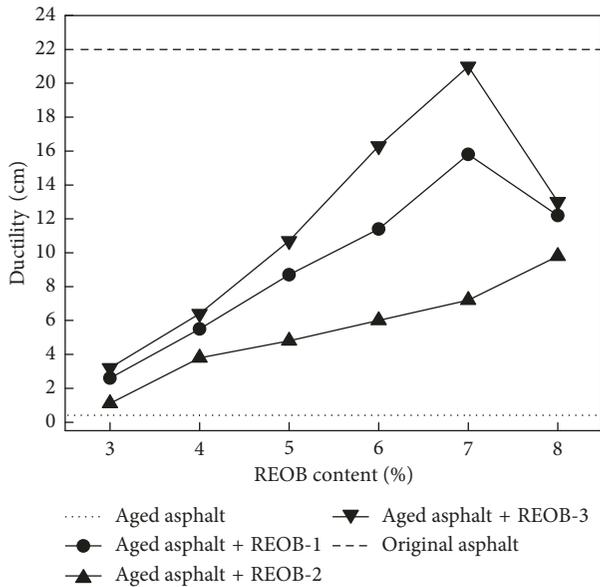


FIGURE 3: Influence of three types of REOB on ductility of aged asphalt.

related to the type and content of REOB. With these three conventional physical indices of an original 70-A asphalt as benchmark values, the ratios between the three conventional physical indices of aged asphalt binders containing different types and dosages of REOB and the benchmark values are defined as the degrees of recovery; the closer the degree of recovery to 100%, the better the rejuvenation effect of REOB on aged asphalt [13]. Therefore, the degrees of recovery for the ductility of aged asphalt with 6% REOB-1 and 8% REOB-2 are 51.8% and 44.5%, respectively, which are far from the standard of original asphalt. For an REOB-3 dosage of 7%, the degrees of recovery for penetration, softening point and ductility are 101.4%, 102.7%, and 95.5%, respectively. The comprehensive recovery effect is the best, and these three conventional physical indices can meet the technical pavement performance requirements; thus, according to physical properties, the best choice for the rejuvenation of aged asphalt is 7% REOB-3. In addition, based on the above test results, REOB-3 was selected for the following rheological tests and the rejuvenation mechanism was discussed; hereafter, REOB refers to REOB-3.

### 3.2. Rheological Properties

**3.2.1. Rutting Factor.** Because the rutting of asphalt pavement mainly occurs during the initial stage of pavement construction, the Strategic Highway Research Program (SHRP) proposed the rutting factor ( $G^*/\sin \delta$ ) of original asphalt or asphalt subjected to short-term ageing using the RTFO test as an evaluation index of the high-temperature stability. The SHRP requires that the  $G^*/\sin \delta$  value of original asphalt is no less than 1.0 kPa and that the  $G^*/\sin \delta$  value of asphalt after RTFO ageing is no less than 2.2 kPa. Figures 4(a) and 4(b) show the influence of REOB content on the  $G^*/\sin \delta$  values of original asphalt and asphalt after

RTFO ageing, respectively. Both figures show that the  $G^*/\sin \delta$  value of asphalt binders decreases with an increase in the testing temperature for both original and RTFO-aged asphalt, indicating that rutting occurs easily within asphalt pavement at high temperatures. At the same testing temperature, the  $G^*/\sin \delta$  value of aged asphalt decreases with an increase in the REOB content from 4% to 7%; this finding indicates that the addition of REOB decreases the rutting resistance of aged asphalt at high temperatures. However, the sudden increase in the  $G^*/\sin \delta$  value of asphalt rejuvenated by 8% REOB indicates that the rutting resistance of binders will increase at this dosage, which is consistent with the softening point analysis in Section 3.1.2. For original binders and RTFO-aged binders, the  $G^*/\sin \delta$  values of aged asphalt with an REOB dosage of 4% to 8% are higher than that of original asphalt; simultaneously, the stability of rejuvenated asphalt with 7% REOB at a high temperature is the closest to that of original asphalt. Additionally, comparing Figure 4(a) with 4(b), the  $G^*/\sin \delta$  values of all binders increase at the same temperature after short-term ageing, which indicates that asphalt binders are hardened. Meanwhile, the high-temperature grade of aged asphalt containing various REOB contents does not lose after short-term ageing.

**3.2.2. Creep Stiffness and Creep Rate.** Contrary to its ability to resist rutting at high temperatures, asphalt pavement will exhibit shrinkage cracking at low temperatures; this cracking is usually due to an increase in the stiffness modulus in addition to the constant ageing of asphalt binders during use and the gradual transition of asphalt from flexible to brittle at low temperatures [12]. Therefore, the SHRP claims that the creep stiffness ( $S$ ) and creep rate ( $m$ ) of asphalt binders after performing RTFO and PAV tests should be used as evaluation indices of the low-temperature cracking resistance, where  $S$  should not exceed 300 MPa and  $m$  should not be less than 0.3 after 60 seconds of testing. The  $m$  value reflects the rate of release of the shrinkage stress during the viscoelastic flow of asphalt binders. Larger values of  $m$  and smaller values of  $S$  correlate with a more favourable cracking resistance of asphalt pavement at low temperatures. The influence of REOB content on the creep stiffness and creep rate of aged asphalt is illustrated in Figures 5(a) and 5(b), respectively, both of which show that the  $S$  value of an asphalt binder will increase and the  $m$  value will decrease with a decrease in the testing temperature, indicating that cracking will occur more easily in asphalt pavement at lower service temperatures. At the same testing temperature, the  $S$  value of aged asphalt decreases and the  $m$  value increases with an increase in the REOB content from 4% to 7%. However, the  $S$  value of rejuvenated asphalt with 8% REOB increases, while the  $m$  value suddenly decreases; this reflects the drop in the low-temperature performance, which is consistent with the ductility analysis in Section 3.1.3. At  $-12^\circ\text{C}$ , an  $S$  value of less than 300 MPa and an  $m$  value exceeding 0.3 will meet the technical standard requirement for aged asphalt containing either 6% or 7% REOB; in addition, the

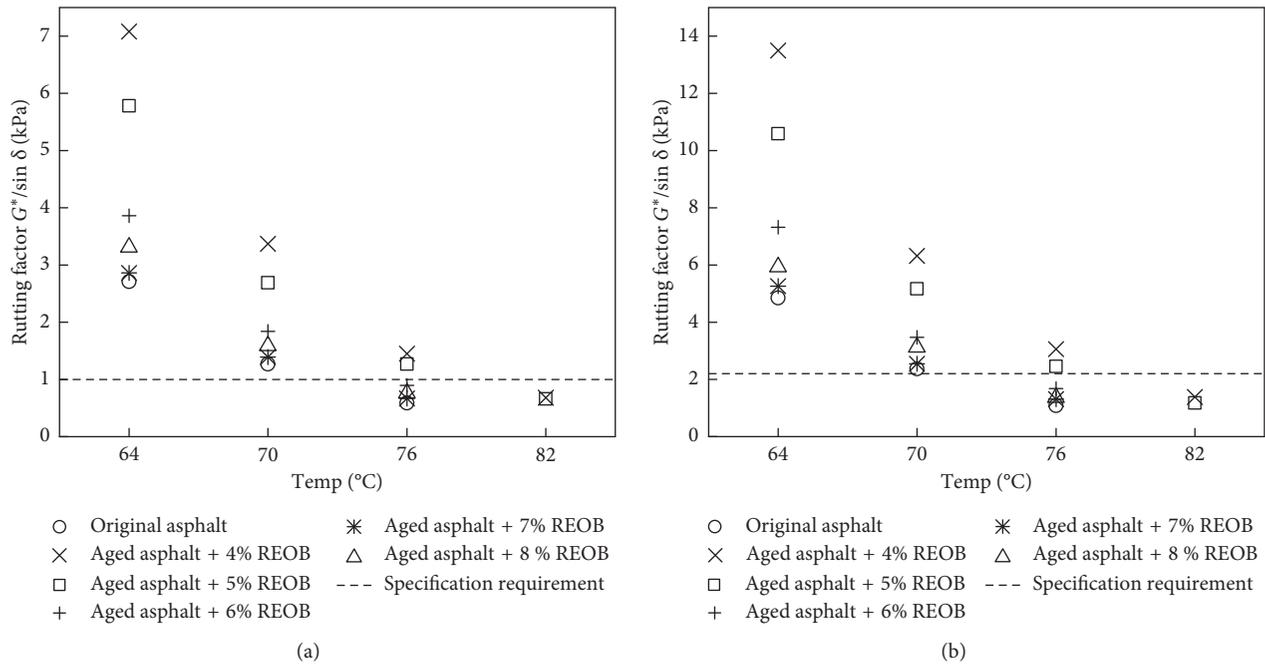


FIGURE 4:  $G^*/\sin \delta$ —temperature for aged asphalt with different REOB content. (a) Original. (b) After RTFO.

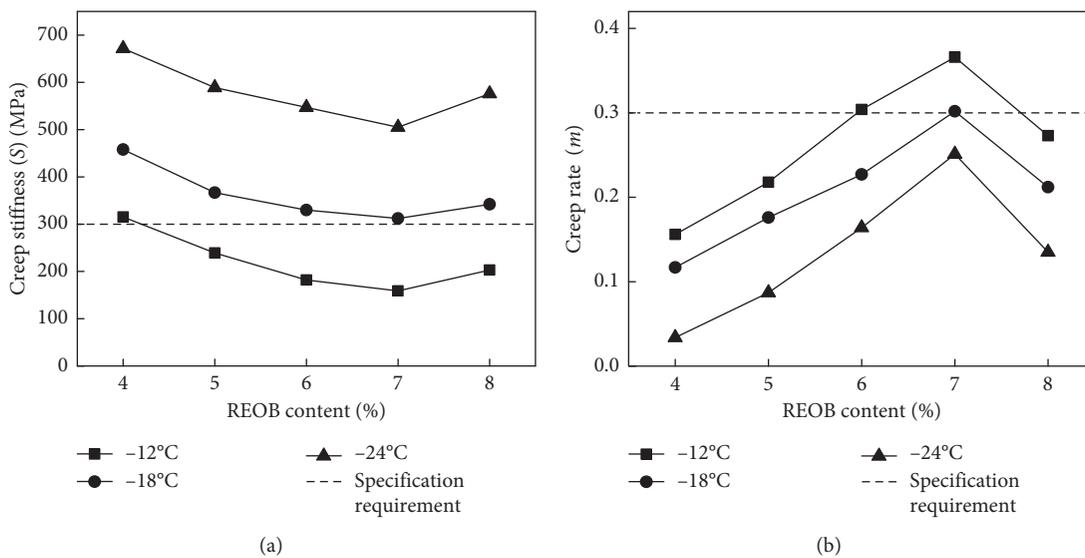


FIGURE 5: Influence of REOB content on creep stiffness and creep rate of aged asphalt. (a) Creep stiffness. (b) Creep rate.

low-temperature cracking resistance of asphalt rejuvenated by 7% REOB is superior to that of asphalt rejuvenated by 6% REOB.

**3.2.3. Performance Grade Classification.** The performance grade can be obtained based on the above analysis performed with a DSR and a BBR, as shown in Table 3. As asphalt continues to age, the high-temperature grade will increase, while the low temperature grade will decrease. Therefore, adding an appropriate REOB dosage can improve the low-temperature performance of aged asphalt, but this will compromise its high-temperature performance.

Nevertheless, under the optimum dosage of REOB for aged asphalt, the performance grade of asphalt rejuvenated by 7% REOB will reach PG70-22. Moreover, its high-temperature grade will be the same as that of original asphalt, while the low-temperature grade of the rejuvenated asphalt will be lower by one grade. At excessively high or low REOB content, the low-temperature performance of rejuvenated asphalt will be even worse, and it will no longer satisfy the SHRP requirements.

**3.3. Four-Component Analysis.** Table 4 shows the content of the four components of different asphalt binders and REOB

TABLE 3: Performance grade of different asphalt binders.

Test objects	High temperature grade	Low temperature grade	Performance grade (PG)
Original asphalt	70	-28	70-28
Aged asphalt	76	-10	76-10
Aged asphalt + 4% REOB	76	-10	76-10
Aged asphalt + 5% REOB	76	-16	76-16
Aged asphalt + 6% REOB	70	-22	70-22
Aged asphalt + 7% REOB	70	-22	70-22
Aged asphalt + 8% REOB	70	-16	76-16

TABLE 4: Content of four components of REOB and different asphalt binders (%).

Test objects		Asphaltenes	Resins	Aromatics	Saturates
Original asphalt		5.4	13.4	39.1	42.1
REOB		0.2	15.5	83.5	0.8
Aged asphalt		13	38.1	11.1	37.8
	Theoretical value	12.23	36.74	15.45	35.58
Rejuvenated asphalt with 6% REOB	Measured value	8	38.2	15.7	38.1
	Difference value	4.23	-1.46	-0.25	-2.52
	Theoretical value	12.1	36.52	16.17	35.21
Rejuvenated asphalt with 7% REOB	Measured value	4.8	41.5	18.4	35.3
	Difference value	7.3	-4.98	-2.23	-0.09
	Theoretical value	11.98	36.29	16.89	34.84
Rejuvenated asphalt with 8% REOB	Measured value	10.6	14.9	25.8	48.7
	Difference value	1.38	21.39	-8.91	-13.86

and demonstrates that the content of asphaltenes and resins will increase as asphalt ages, while the content of aromatics and saturates will correspondingly decrease. The main reason for this is that the light components composed of aromatics and saturates will oxidize and polymerize as the asphalt ages, and they will gradually transform into recombinant components comprising resins and asphaltenes.

Most of the components of REOB are aromatics with some resins and few asphaltenes and saturates. Therefore, on the one hand, adding certain dosages of REOB to aged asphalt will supplement the aged asphalt with aromatics that are otherwise missing, thereby harmonizing the four components and achieving a stable asphalt colloid structure. On the other hand, the volatile saturates content in REOB is very small, indicating that REOB exhibits a good heat resistance as an asphalt rejuvenator, which is consistent with the technical requirements of asphalt rejuvenators [14].

According to the theory of pure component adjustment, the mechanism through which REOB rejuvenates aged asphalt is the pure supplement of components, that is, no reactions and transformations occur among the four components [15]. So each component conforms to the formula (1):

$$P = m_a P_a + m_r P_r, \quad (1)$$

where  $P$  is the proportion of one component in rejuvenated asphalt (%);  $m_a$  is the blending ratio of aged asphalt in rejuvenated asphalt;  $P_a$  is the proportion of one component in aged asphalt (%);  $m_r$  is the blending ratio of REOB in rejuvenated asphalt; and  $P_r$  is the proportion of one component in REOB (%).

However, Table 4 illustrates that the theoretical results for the four components calculated by formula (1) of pure adjustment theory are substantially different from those measured through actual testing, which indicates that the rejuvenation mechanism of REOB in aged asphalt can be defined as incomplete component adjustment, in which some components undergo physical or chemical reactions and transformations during the rejuvenation process.

Comparing with the results calculated by formula (1) of pure component adjustment, the measured asphaltene content of rejuvenated asphalt with 6% REOB is reduced by 4.23%, while the contents of resins, aromatics, and saturates correspondingly increase to varying degrees. These differences are observed because when 6% REOB is added to aged asphalt, aromatics are used as a dispersing medium for the asphalt colloidal structure; these aromatics can peptize and disperse asphaltenes, making them undergo an inverse transformation, and some of the asphaltenes are converted into resins, aromatics, and saturates. For rejuvenated asphalt with 7% REOB, the degree of asphaltene dissolution is greater and the amount of transference out of the structure reaches 7.3%; thus, its content is reduced to the nearest level of original asphalt; the amounts of resins and aromatics taken in are also more for rejuvenated asphalt with 7% REOB than for that with 6% REOB. However, different conditions are observed for rejuvenated asphalt with 8% REOB; the amount of asphaltenes transferred out is sharply reduced at this time, and thus, the asphaltene content reaches 10.6% compared with the amount in rejuvenated asphalt with 6% and 7% REOB. The largest proportion of resin transferred out reaches 21.5%, and the largest proportions of aromatics and saturates transferred in are 13.7% and 9.3%, respectively.

According to asphalt colloid structure theory, asphaltene are hydrophobic to oil components, which are composed of aromatics and saturates, but they exhibit an affinity to resins. Therefore, asphaltene need to be encapsulated by a sufficient amount of resins to form micelles and be dispersed in oil components to develop stable colloidal structures [16]. However, the resin content is too small relative to the asphaltene content for rejuvenated asphalt with 8% REOB; asphaltene would flocculate too easily due to the lack of resins for encapsulating the asphaltene, and as a consequence, a stable colloidal structure cannot be formed. In summary, the colloidal structure of rejuvenated asphalt with 7% REOB is the most stable. Moreover, adding REOB can achieve a good rejuvenation effect by reducing the asphaltene content and increasing the resin content in aged asphalt.

**3.4. Infrared Spectroscopic Analysis.** The technical properties of asphalt are related not only to the content of chemical components but also to the chemical structure. Figure 6 shows infrared spectrograms of REOB, original asphalt, aged asphalt, and rejuvenated asphalt with 7% REOB. The infrared spectrogram of REOB demonstrates a strong asymmetric stretching vibration peak at  $2925\text{ cm}^{-1}$  and the symmetric stretching vibration peak of methylene ( $-\text{CH}_2-$ ) at  $2854\text{ cm}^{-1}$ . At  $1464\text{ cm}^{-1}$  and  $1377\text{ cm}^{-1}$ , the asymmetric and symmetric bending vibration peaks of methyl ( $-\text{CH}_3$ ), respectively, are observed. The strong absorption peak at  $722\text{ cm}^{-1}$  is the bending vibration peak of long-chain methylene  $-(\text{CH}_2)_n-$ , and the position of the absorption peak can be used to determine that  $n > 4$ . The strong absorption peaks at  $1744\text{ cm}^{-1}$  and  $1711\text{ cm}^{-1}$  can prove the existence of carbonyl groups ( $\text{C}=\text{O}$ ) composed of acids and ketones in REOB. The vibration absorption peak of the carbon skeleton ( $\text{C}=\text{C}$ ) on the benzene ring is near  $1450\text{ cm}^{-1}$ , and the absorption peak near  $900\text{ cm}^{-1}$  is caused by C-H on the benzene ring. In addition, there are acid amides (at  $1260\text{ cm}^{-1}$ ), phosphorous compounds (at  $1247\text{ cm}^{-1}$ ), and sulfur compounds (at  $1163\text{ cm}^{-1}$ ) in the REOB. In conclusion, REOB is composed of base oil and complex mixtures containing a variety of polar functional groups. The main components of REOB are alkanes, naphthenes, and aromatic hydrocarbons, which have chemical compositions that are very similar to that of asphalt. According to the theory of similarity compatibility, REOB is compatible with asphalt and can form a stable colloidal structure.

Comparing the infrared spectrum of aged asphalt with that of original asphalt, the positions of the vibration peaks of both asphalts are basically the same, but some peak strengths are different. Owing to the oxidation reaction of asphalt during ageing, the ketone carbonyl ( $\text{C}=\text{O}$ ) vibration peak appears at  $1710\text{ cm}^{-1}$ , which is the characteristic peak of ageing asphalt, and the sulfoxide group ( $\text{S}=\text{O}$ ) vibration peak at  $1060\text{ cm}^{-1}$  is significantly enhanced. In addition, the methylene ( $-\text{CH}_2-$ ) and methyl ( $-\text{CH}_3$ ) vibration peaks at  $2925\text{ cm}^{-1}$ ,  $2854\text{ cm}^{-1}$ ,  $1475\text{ cm}^{-1}$ , and  $1376\text{ cm}^{-1}$  are weakened, and the C-H vibration peaks on benzene rings at  $750\text{--}900\text{ cm}^{-1}$  are also weakened; these effects are the result

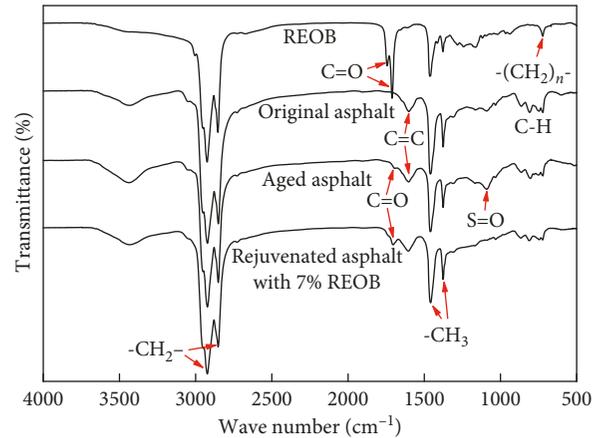


FIGURE 6: Infrared spectrograms of REOB and asphalt binders.

of condensation and dehydrogenation during asphalt ageing.

Due to the small content of REOB, the infrared spectrum of rejuvenated asphalt with 7% REOB is similar to that of aged asphalt. Compared with that of aged asphalt, the ketone carbonyl ( $\text{C}=\text{O}$ ) vibration peak of rejuvenated asphalt at  $1710\text{ cm}^{-1}$  is obviously enhanced because of the addition of REOB carbonyl groups to the rejuvenated asphalt, but the sulfoxide ( $\text{S}=\text{O}$ ) vibration peak at  $1060\text{ cm}^{-1}$  is significantly weakened. In summary, the characteristic peaks of REOB also exist in rejuvenated asphalt, and some vibration peaks of functional groups evidently change both before and after rejuvenation, indicating that chemical reaction and transformation of the structures occur within rejuvenated asphalt; these findings are consistent with the correlation mechanism between REOB and aged asphalt ascertained in the four-component analysis presented in Section 3.3.

#### 4. Grey Relationship Analysis of Properties and Components

The differences in the asphalt properties depend on the chemical composition of the asphalt [17]. The addition of REOB changes the components in addition to the physical and rheological properties of aged asphalt. The relationship between the four components and the physical rheological properties of asphalt rejuvenated by REOB are obtained quantitatively by the grey relationship analysis to improve the asphalt pavement performance by controlling the REOB content.

The penetration, softening point, ductility, rutting factor (after RTFO ageing at a testing temperature of  $70^\circ\text{C}$ ), creep stiffness, and creep rate (at a testing temperature of  $-12^\circ\text{C}$ ) of aged asphalt and rejuvenated asphalt with 6%, 7%, and 8% REOB were selected as reference sequences  $X_{01}$ ,  $X_{02}$ ,  $X_{03}$ ,  $X_{04}$ ,  $X_{05}$ , and  $X_{06}$ , respectively, and the content of asphaltene, resins, aromatics, and saturates were used as comparison sequences  $X_1$ ,  $X_2$ ,  $X_3$ , and  $X_4$ , respectively. The relationship degrees in Table 5 are calculated from the data in Table 6 according to the calculation steps of grey relationship analysis method [17, 18].

TABLE 5: Relationship degrees between the properties indices and four components.

Projects	Asphaltenes $X_1$	Resins $X_2$	Aromatics $X_3$	Saturates $X_4$
Penetration $X_{01}$	0.524	0.545	0.656	0.563
Softening point $X_{02}$	0.860	0.785	0.587	0.802
Ductility $X_{03}$	0.544	0.545	0.550	0.547
Rutting factor $X_{04}$	0.905	0.788	0.565	0.701
Creep stiffness $X_{05}$	0.787	0.730	0.552	0.655
Creep rate $X_{06}$	0.539	0.545	0.594	0.558

TABLE 6: Raw data of aged asphalt and rejuvenated asphalts with REOB.

Projects	Aged asphalt	Rejuvenated asphalt with 6% REOB	Rejuvenated asphalt with 7% REOB	Rejuvenated asphalt with 8% REOB
$X_{01}$	24	68	72	75
$X_{02}$	66	53.2	50	52.8
$X_{03}$	0.4	16.3	21	13
$X_{04}$	6.84	3.47	2.55	3.13
$X_{05}$	1172	182	159	203
$X_{06}$	0.057	0.304	0.366	0.273
$X_1$	13	8	4.8	10.6
$X_2$	38.1	38.2	41.5	14.9
$X_3$	11.1	15.7	18.4	25.8
$X_4$	37.8	38.1	35.3	48.7

For asphalt rejuvenated by REOB, Table 5 shows that the relationship degree with the penetration is greatest for aromatics, followed by saturates, resins, and asphaltenes, while the relationship degree with the softening point is greatest for asphaltenes, followed by saturates, resins, and aromatics; for ductility, the difference in relationship degree between the four components is very small, indicating that good ductility requires a reasonable combination of the four components. These correlations are basically consistent with the qualitative relationships between the four components and the properties of asphalt in the blending method [19]. As the dispersing medium of an asphalt colloid solution, aromatics can play the role of a glue dissolution and softening agent; that is, it can make the asphalt softer, thereby increasing penetration. In contrast, asphaltenes are thickening agents, and saturates are used as a softener. The plasticization of aromatics is stronger than that of saturates, and asphaltenes require more softener than resins. Therefore, asphaltenes could increase the softening point, while saturates could reduce the softening point. Furthermore, the reasonable combination of the four components can improve the plasticity and ductility of asphalt binders at low temperature.

For the rheological indices, the order of the relationship degree with  $G^*/\sin \delta$  is as follows: asphaltenes > resins > saturates > aromatics; the order of the relationship degree with  $S$  is the same as that of  $G^*/\sin \delta$ , which indicates that asphaltenes have the greatest influence on the deformation resistance of binders at both high and low temperatures. The order of the relationship degree with  $m$  is as follows: aromatics > saturates > resins > asphaltenes; this order is consistent with the correlation order of ductility, and the difference in relationship degrees among the four components is very small, which indicates that the reasonable combination of four components can ensure a good deformation rate.

In order to better compare each row or column in Table 5, the correlation and advantage between the four-component and physical rheological indexes of rejuvenated asphalt with different REOB contents are comprehensively analysed, and the grey relationship matrix ( $\gamma$ ) with the coefficients  $\gamma_{0xy}$  according to Table 5 is established, as shown below. Among them,  $x$  corresponds to the row order of performance indexes in Table 5, and  $y$  corresponds to the column order of four-component in Table 5.

$$\gamma = \begin{bmatrix} \gamma_{011} & \gamma_{012} & \gamma_{013} & \gamma_{014} \\ \gamma_{021} & \gamma_{022} & \gamma_{023} & \gamma_{024} \\ \gamma_{031} & \gamma_{032} & \gamma_{033} & \gamma_{034} \\ \gamma_{041} & \gamma_{042} & \gamma_{043} & \gamma_{044} \\ \gamma_{051} & \gamma_{052} & \gamma_{053} & \gamma_{054} \\ \gamma_{061} & \gamma_{062} & \gamma_{063} & \gamma_{064} \end{bmatrix} = \begin{bmatrix} 0.524 & 0.545 & 0.656 & 0.563 \\ 0.860 & 0.785 & 0.587 & 0.802 \\ 0.544 & 0.545 & 0.550 & 0.547 \\ 0.905 & 0.788 & 0.565 & 0.701 \\ 0.787 & 0.730 & 0.552 & 0.655 \\ 0.539 & 0.545 & 0.594 & 0.558 \end{bmatrix}. \quad (2)$$

Comparing the relationship degree of each row in the matrix, the relationship degrees among the four components and the softening point in the second row are relatively large, indicating that REOB is the most sensitive to softening point; thus, the main function of REOB is to reduce the viscosity of aged asphalt, which meets the relevant technical standards of asphalt rejuvenators at home and abroad [14]. Moreover, a comparison of the relationship degree of each column in the matrix reveals that the relationship degree between the physical rheological indices and asphaltenes in the first column is relatively large, which reflects the fact that the asphaltene content has the greatest influence on the properties of rejuvenated asphalt. Therefore, the asphaltene content should be strictly controlled during the addition of REOB to rejuvenate aged asphalt. These results also confirm the analysis of the four components in Section 3.3.

## 5. Conclusions

- (1) An analysis of physical properties shows that the three types of REOB can increase the penetration and ductility and decrease the softening point of aged asphalt under proper dosage, but the recovery effect of REOB-1 and REOB-2 on ductility is too poor. Only when 7% REOB-3 is used are the three physical indices of aged asphalt restored to the level of original asphalt.
- (2) An analysis of the rheological properties shows that a suitable dosage of REOB can reduce the rutting factor  $G^*/\sin \delta$  of aged asphalt and decrease the creep stiffness while increasing the creep rate. A performance grade classification indicates that a moderate REOB content can decrease the high-temperature grade and increase the low-temperature grade of aged asphalt, thereby showing a good consistency with the analysis of physical properties. In comparison, the high-temperature and low-temperature performances of rejuvenated asphalt with 7% REOB are the closest to those of original asphalt, but its low-temperature performance is slightly worse and thus needs further improvement.
- (3) Four-component analysis indicates that most of the components of REOB are aromatics. The mechanism through which REOB rejuvenates aged asphalt is incomplete component adjustment; that is, some of the components undergo physical or chemical reactions and transformations. The addition of REOB can achieve a rejuvenation effect mainly by reducing the asphaltene content and increasing the resin content in aged asphalt.
- (4) Infrared spectra analysis indicates that the main components of REOB are similar and compatible with asphalt. Adding 7% REOB to aged asphalt can significantly reduce the strength of sulfoxide functional groups, thereby achieving a rejuvenation effect.
- (5) The grey relationship analysis reveals that asphaltenes have the greatest influence on the high-temperature performance of asphalt rejuvenated by REOB and low-temperature performance requires a reasonable combination of the four components. Moreover, a comprehensive advantage analysis demonstrates that the addition of REOB has the greatest sensitivity to softening point, and asphaltene content has the greatest influence on the physical and rheological properties of asphalt rejuvenated by REOB. In addition, the rejuvenation performance of modified asphalt should be further studied by using different REOB.

## Data Availability

The experimental data in this paper are from the pavement material laboratory of Shandong Jiaotong University, which is the provincial key laboratory. The experimental data in this paper are real and reliable.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

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## Supplementary Materials

S1: the basic theory of grey relationship analysis. S2: the calculation steps of grey relationship analysis. (*Supplementary Materials*)

## References

- [1] Y.-F. Ma, D.-X. Liu, W.-L. Xu et al., "Study on the current situation and key technology of waste engine oil regeneration in the transportation industry," *Comprehensive Utilization of China's Resources*, vol. 28, no. 11, pp. 25–29, 2010.
- [2] S.-R. Zhou, *Heat and Mass Transfer in Short-Path Distillation Process and Application of the Process to Regeneration of Waste Lubricating Oil*, Sichuan University, Chengdu, China, 2007.
- [3] H.-Q. Xie, Z.-T. Ma, Y. Han et al., "Test of regeneration processes of waste gasoline engine oil," *Petroleum Processing and Petrochemicals*, vol. 48, no. 1, pp. 85–88, 2007.
- [4] A. Villanueva, S. Ho, and L. Zanzotto, "Asphalt modification with used lubricating oil," *Canadian Journal of Civil Engineering*, vol. 35, no. 2, pp. 148–157, 2008.
- [5] D. S. Ackbarali and R. Maharaj, "The viscoelastic properties of Trinidad Lake asphalt-used engine oil blends," *International Journal of Applied Chemistry*, vol. 7, pp. 1–8, 2011.
- [6] C. Dedene, "Investigation of using waste engine oil blended with reclaimed asphalt materials to improve pavement recyclability," Master Thesis, Michigan Technological University, Houghton, Michigan, 2011.
- [7] X. Jia, B. Huang, B. F. Bowers, and S. Zhao, "Infrared spectra and rheological properties of asphalt cement containing waste engine oil residues," *Construction and Building Materials*, vol. 50, no. 3, pp. 683–691, 2014.
- [8] P.-P. Xu, *Study on the Properties of Waste Oil Recycled Asphalt and its Mixtures*, Jilin University, Changchun, China, 2015.
- [9] S. A. M. Hesp and H. F. Shurvell, "X-ray fluorescence detection of waste engine oil residue in asphalt and its effect on cracking in service," *International Journal of Pavement Engineering*, vol. 11, no. 6, pp. 541–553, 2010.
- [10] S. Rubab, K. Burke, L. Wright, S. A. M. Hesp, P. Marks, and C. Raymond, "Effect of engine oil residues on asphalt cement quality," in *Proceedings of the Fifty-Sixth Annual Conference of*

- the Canadian Technical Asphalt Association*, pp. 1–12, Québec City, Canada, November 2011.
- [11] H.-B. Ding, Y.-J. Qiu, W.-Q. Wang et al., “Adverse effect of waste engine oil bottom on asphalt performance and its mechanism,” *Journal of Building Materials*, vol. 20, no. 4, pp. 646–650, 2017.
- [12] J.-A. Shen, *Road Performance of Asphalt and Asphalt Mixture*, People’s Communications Press, Beijing, China, 2001.
- [13] Y. Zhang, M.-Z. Chen, S.-P. Wu et al., “Study on physical properties and structure of recycled asphalt with different waste oil,” *Journal of Wuhan University of Technology (Transportation Science and Engineering Edition)*, vol. 41, no. 1, pp. 104–108, 2017.
- [14] J.-G. Geng, *Study on Asphalt Aging Mechanism and Regeneration Technology*, Chang’an University, Xi’an, China, 2009.
- [15] Y.-H. Nie, P. Liu, Z.-G. Zhou et al., “Study on the effects of regenerator on the mechanism of asphalt regeneration,” *Highway Engineering*, vol. 37, no. 6, pp. 65–68, 2012.
- [16] Z.-H. Wang, J.-H. Fang, and M.-F. He, “Discussion on the aging process and regeneration mechanism of asphalt,” *Qinghai Transportation Technology*, no. 4, pp. 45–48, 2017.
- [17] H.-X. Chen, M.-S. He, X.-H. Ji et al., “Grey relationship analysis of asphalt properties and asphalt composition,” *Journal of Chang’an University (Nature Science Edition)*, vol. 34, no. 3, pp. 1–6, 2014.
- [18] J.-L. Deng, *Basic Methods of Grey System*, East China University of Technology Press, Shanghai, China, 1987.
- [19] Y.-G. Wang, K.-J. Liao, F. Yan et al., “Reclaimed waste asphalt by blending method,” *Chemical Technology*, no. 2, pp. 22–24, 2003.



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