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Research Article

Infrared Spectrum Analysis of Aged Asphalt Regenerated by Recycled Engine Oil Bottom

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In order to analyze the feasibility of recycled engine oil bottom (REOB) as a regenerant for old asphalt and to determine its improvement effect on the performance of recycled asphalt, this paper analyzes the functional groups of two kinds of old asphalt and matrix asphalt through infrared spectrum test, such as carbonyl group, butadiene double bond, styrene double bond, methyl group, methylene group, and aromatic ring in the molecules of REOB. Find out the compatibility characteristics between them from the microscopic point of view. The recycled asphalt with 4%, 6%, and 8% content was prepared, and the physical property test was conducted to analyze the regeneration effect of different contents of REOB, and the optimum amount of REOB for 10-year-old asphalt was determined to be 6%. The study found that REOB contains aromatic hydrocarbons and colloids, which can supplement the reduction of aromatic hydrocarbons and colloids caused by asphalt aging. Therefore, REOB helps to adjust the composition of aging asphalt, improve its performance, and achieve good regeneration.

1. Introduction

Both the motor oil and the asphalt used in road construction is the production of petroleum produced by different refining processes. Although they come from the same raw material, they have different chemical components. The motor oil is mainly composed of base oil and additive. Base oil is generally refined from crude oil, and its composition generally consists of alkane, cycolalkane, aromatic hydrocarbon, cycloalkyl aromatic hydrocarbon, and some organic compounds containing oxygen, nitrogen, and sulfur. Asphalt is a complex macromolecular mixture of linear alkanes with different length side chains, cycolalkanes with multiple substituents, and aromatic fused rings. Although the composition of the motor oil and asphalt is not exactly the same, the components have a good compatibility [1]. At present, the regeneration of aged asphalt is fundamentally to reconcile its composition by adding new asphalt or a regenerant containing suitable chemical components to restore the performance of the aged asphalt [2, 3]. As early as the 1990s, in Europe and the United States, large quantities of residues from the re-refining used waste oil have been used as fillers in industrial and roofing asphalts. "the re-refining used waste oil" is a Technical Term. However, the use of waste oil residues in road science was not widespread, at least not widely reported in the literature, until 1992, when Herrington published a paper given a preliminary discussion on the use of re-refined waste oil residues as additives for road asphalt [4, 5]. In fact, recycled engine oil bottom have been studied abroad as asphalt material modifiers. DeDene [6] used waste oil to modify the aged asphalt and carried out rutting test and water stability evaluation test of the modified asphalt mixture, and the results showed that the waste oil can be used as a softening regenerant to restore the performance of aged asphalt.

In recent years, the main research direction of asphalt regeneration in China has begun to focus on finding materials that are both environmentally friendly and have good regeneration effects. The use of bio-binders, bio-diesel residues, etc. as rejuvenating agents for aging asphalt has received much attention in the field of domestic road research [7]. However, there are few research studies and applications on the application of industrial wastes to the regeneration of aged asphalt in China. Most scholars mainly focus on the application of bio-asphalt in road engineering. The research of REOB used as the aging of asphalt regenerant is rarely in China, most of the research is aimed at the performance of waste engine oil regeneration asphalt and its mixture. However, the composition of waste oil and REOB is different, in particular, the components that affect the regeneration improvement effect, such as soft oil content and metal particle impurity content, are very different. In this study, the characteristics of function groups, such as the carbonyl, butadiene double bond, styrene double bond, methyl group, methylene, aromatic ring in matrix asphalt, REOB, and aged asphalt, are analyzed by using Fourier transform infrared spectroscopy (FTIR) to find out the compatibility characteristics among them and analyze the theoretical feasibility of REOB as a regenerant for recycling asphalt. The basic physical properties of REOB recycled asphalt were evaluated to verify its regeneration effect and determine the optimum dosage of REOB as regenerating agent.

2. Test Sample and Mechanism

2.1. Preparation of Aged Asphalt and Recycled Engine Oil Bottom

(1) The matrix asphalt used in this test is NO.70 matrix asphalt. The aged asphalt is extracted by the chemical solvent method from ten-year-old and fifteen-year-old asphalt mixture used on pavement. The asphalt and the aggregate of the mixture are separated according to the T0722-1993 centrifugal separation method in <Test Procedure for Highway Engineering Asphalt and Asphalt Mixture >(E20-2011). Bitumen in the extracted bitumen solution was recovered from the trichloroethylene solvent by the Abson method (T0726-2011).

At present, the commonly used method for removing mineral powder is to use a high-speed centrifuge to settle the mineral powder in the extract. Many researchers have found that the centrifugal extraction method cannot extract completely the mineral powder in the RAP material according to the centrifugal force and centrifugation time specified in the specification. After several extraction and recovery experiments, it was found that the sedimentation velocity of the mineral powder decreased with the increase of the asphalt concentration, and the increase of the relative centrifugal force could significantly increase the sedimentation velocity. The asphalt concentration in the extract obtained in this test was generally 20% to 30%. In order to completely remove the mineral powder in the asphalt solution, a high-speed centrifuge is used to apply a relative centrifugal force of $4000 \times g$, and the centrifugation time is 30 minutes, so as to completely remove the mineral powder in the solution.

(2) The aging deterioration of motor oil is caused by various factors, such as moisture, unburned fuel, soot, carbonaceous material mixed in, dust inhaled, metal scraps generated by excessive wear, and insoluble and soluble oxides generated by the oxidation of motor oil [8]. The recovery residue of motor vehicle maintenance replaced oil selected in this study is a typical representative domestic REOB, which is obtained from the motor oil accessed from the automobile 4S shop randomly and then recovered by ultrafiltration and centrifugal separation. In order to reduce the aging during storage, store the REOB in opaque containers and keep them in a cool place. When taking them, they should be fully stirred to avoid uneven composition caused by stratification caused by standing for too long. The REOB was tested according to the performance index of asphalt rejuvenating agent required by the specification, and the results are shown in Table 1.

It can be seen that the viscosity of REOB can meet the needs of construction and mixing, and the flash point is high, which meets the requirements of construction safety. After the film oven aging test, the viscosity change and quality loss are relatively small, indicating that the anti-aging performance of REOB meets the requirements of the regenerant requirements.

2.2. Infrared Spectroscopy Test. When infrared light illuminates the sample, the radiation is not enough to cause the transition of the electron energy level in the molecule, but it can be absorbed by the molecule to cause the transition of the vibration level. In the case of vibration, infrared photons are absorbed by those with dipole moment changes, thereby forming infrared absorption spectrum [9]. Different substances have different absorption frequencies for infrared radiation, and the positions of the bands formed are also different. The intensity and shape of the bands formed are also different depending on the amount of material. For the functional group, it has a characteristic infrared absorption peak. According to the parameters such as the position, number, relative intensity, and shape of the infrared characteristic absorption peak of various substances, it can be inferred which groups exist in the sample substance, and the molecular structure thereof is determined [10, 11].

Fourier transform infrared spectroscopy (FTIR) is a commonly used method for qualitative or semi-quantitative analysis of organic compounds. Compared with other microstructure analysis tests, infrared spectroscopy has a wide range of applicable materials, high resolution, and short test time, requires less samples, and can quickly provide the infrared characteristic absorption peaks of various functional groups in the constituent molecules of the test sample. The chemical functional group species of the sample can be qualitatively analyzed by infrared spectroscopy, and its functional group composition can also be analyzed semi-quantitatively. In this experiment, the infrared spectroscopy technique was used to evaluate and analyze the recycled aged asphalt and waste oil residues of different service life. The microscopic principle of using REOB to regenerate the aged

Table 1: Performance index of recycled engine oil bottom (REOB).

Index	REOB	Method
Appearance	Black viscous liquid	Visual test
Density (20°C, g/cm³)	0.9605	GB/T1884
Viscosity (60°C, Pa.s)	0.235	GB/T11137
Flash point (COC) (°C)	295	GB/T3536
Pour point (°C)	6	GB/T3535
Viscosity ratio before and after film oven test	2.4	GB/T11137
Quality loss before and after film oven test	3.0	GB/T0609

asphalt was explored, which provided the theoretical basis for REOB as the asphalt regenerant.

In this test, we used the Nicolet iS10 attenuated total reflection infrared spectrometer from American Thermoelectric Corporation, with a spectral resolution better than 10px-1 and a wavenumber accuracy better than 0.125px-1. During the test, the asphalt sample and a certain amount of KBr powder were simultaneously ground into a fine powder of micron grade and pressed into a transparent sheet by a special tableting device and then analyzed.

2.3. Test Mechanism. Current research suggests that the aging of asphalt is an irreversible phenomenon mainly caused by changes in asphalt composition under load and temperature; the aromatic component will be oxidized and converted into colloid during the asphalt aging, and colloid oxidized to form asphaltenes. With load and temperature persisting, asphaltene is converted to coke and hydrocarbon finally, and then changes the performance of asphalt [12, 13]. The study found that the viscosity of asphalt is proportional to the content of asphaltenes and gums and inversely proportional to the contents of aromatics and saturates; the ductility of asphalt is proportional to the content of gums; asphaltenes can be dissolved in aromatics, but insoluble in saturated point. The composition analysis of asphalt before and after aging shows that the content of aromatic components decreases and the content of asphaltene increases after aging. From this perspective, asphalt regeneration is to use a rejuvenating agent containing a large amount of aromatic components to reconcile the components of the aging asphalt, so that the content of each component meets the component content relationship required by the performance requirements, so as to achieve the effect of asphalt regeneration. The use of REOB as a regenerant for aging asphalt is to consider the regeneration of asphalt from the perspective of component reconciliation. REOB contains a lot of aromatic components. In theory, adding it to the aging asphalt can make the content of each component of the asphalt meet the performance requirements and achieve the effect of asphalt regeneration.

The main mechanism of this test is to add REOB sample to the aged asphalt by certain methods, reconciles component of aged asphalt with REOB ingredients, and then completes the regeneration of asphalt. When the aged asphalt is regenerated, it is required that the aged asphalt can be fully integrated with the rejuvenating agent, and the main factors affecting the dispersion degree of the rejuvenating agent in the aged asphalt are the regeneration temperature and time. The high-speed shearing method is used for the preparation of recycled asphalt. When the shearing temperature is too high and the time is too long, it will cause secondary aging of the asphalt. If the temperature is too low and the shearing time is too short, the asphalt and REOB cannot be fully mixed, and the properties of the prepared recycled asphalt are not uniform. Therefore, referring to the preparation process of polymer-modified asphalt, this paper chose to control the shear temperature at 150°C, the shear time as 30 min, and the shear rate as 4000 r/min. The prepared regenerated asphalt was allowed to stand at room temperature for 24 hours, so that REOB was fully mixed with the aged asphalt to achieve a state of stable performance.

3. Research Results and Discussion

3.1. Infrared Spectrum Analysis of Asphalt and REOB. The asphalt mixture with natural aging for 10 years and 15 years was obtained from the maintenance site of the BeiYuan elevated road in Jinan. The infrared spectrum of the two recycled asphalt was tested, $70^{\#}$ matrix asphalt and REOB samples. The infrared spectrum obtained is shown in Figures 1 and 2.

Looking at the infrared spectrum, it can be seen that the frequency region of the spectral absorption peak can be divided into the following five regions:

(1) 400~900 cm^{-1}: the absorption peak in this region can be used to confirm the cis-trans configuration of the compound, and the absorption peaks at 840 and 730 cm^{-1} represent alkenyl or benzene ring C=C bond.

(2) 900~1300 cm^{-1}: this area is the absorption area of C-O, C-N, C-P, and other single-bond stretching vibrations and C=S, S=O, and other double-bond stretching vibrations.

(3) 1300~1900 cm^{-1}: this region is the single-bond bending vibration region and the double-bond stretching vibration region, mainly including three kinds of stretching vibration—carbonyl C=O double between 1650 and 1900 cm cm^{-1} The bond stretching vibration absorption peak is the basis for judging the existence of high polymers such as ketones, esters, and aldehydes; the C=C double bond stretching vibration of olefins between 1500 and 1700 $cm^{-1}$is generally at 1500 cm^{-1} and 1600 cm^{-1}annexes appear two absorption peaks, which can be used as the basis

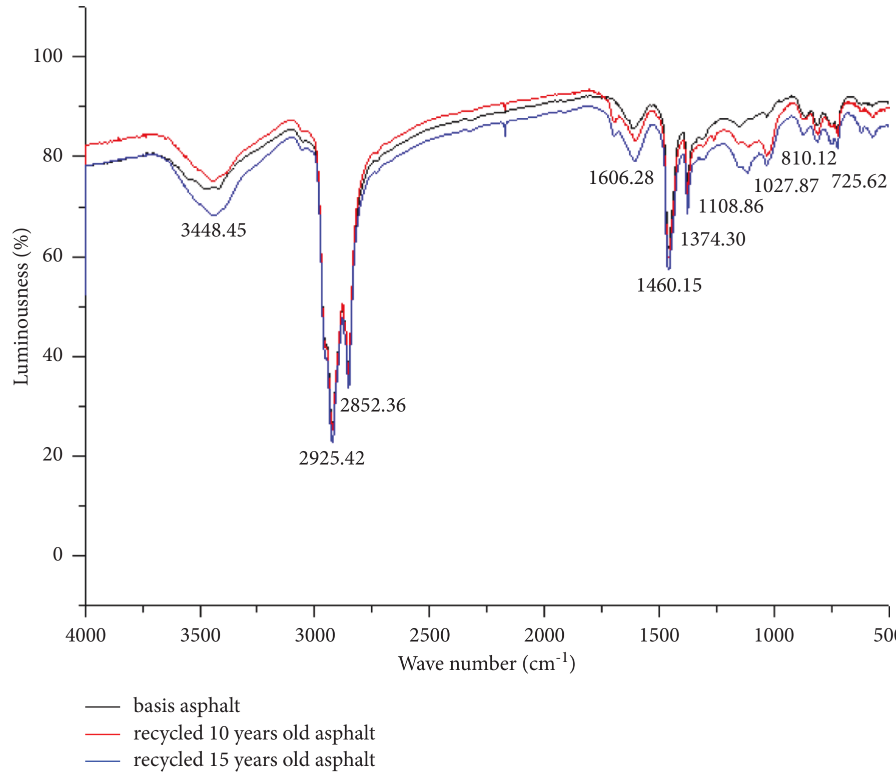

Figure 1: Infrared spectrum of matrix asphalt and two kinds of recycled asphalt.

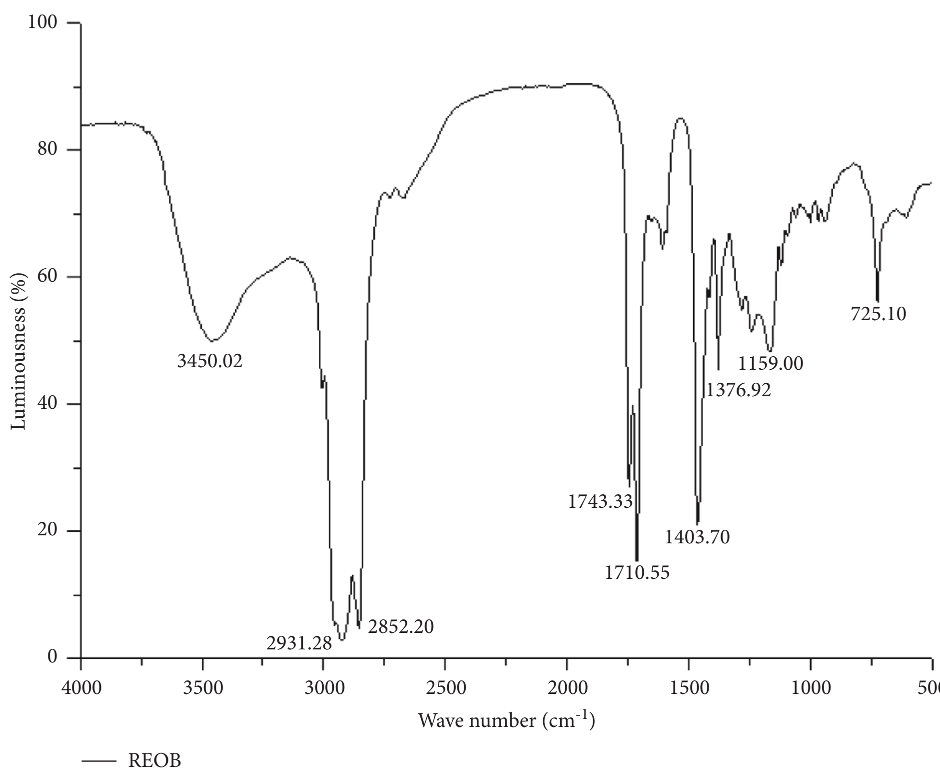

Figure 2: Infrared spectrum of REOB.

for judging the existence of aromatic rings; C=C and C-H absorption peaks between 1600 and 2000 $cm^{-1}$are the basis for judging the type of aromatic ring substitution.

(4) 1900~2500 cm^{-1}: this region is the absorption region of triple bond stretching vibration such as C≡C and C≡N and cumulative double-bond asymmetric stretching vibration such as C=C=C and C=C=O.

(5) 2500~4000 cm^{-1}: this region is the single-bond stretching vibration region such as C-H, O-H, H-H. Among them, the O-H absorption peaks between 3200 and 3700 cm^{-1} can be used as the basis for judging the existence of alcohols, phenols, and organic acids; 2800~3000 cm^{-1} is the stretching vibration absorption of saturated C-H group peak; above 3000 cm^{-1} is the stretching vibration absorption peak of unsaturated C-H group.

The following conclusions can be drawn from the analysis of the test data.

(1) As can be seen in Figure 1, there are six distinct absorption peaks in the infrared spectrum of the three kinds of asphalt, in the vicinity of 3448 cm^{-1}, 2925 cm^{-1}, 2852 cm^{-1}, 1606 cm^{-1}, 1460 cm^{-1}, 1374 cm^{-1}, it is known from the infrared frequency characteristic frequency table of common functional groups, 2925 cm^{-1} and 2852 $cm^{-1}$are absorption peaks of two functional groups of alkyl-C H_2 and-C H_3,3448 $cm^{-1}$is the absorption peak of heteroaromatic ring pyrrole, 1606 cm^{-1} is the absorption peak of benzene ring and fused aromatic ring C–, 1460cm^{-1}is the absorption peak of –C H_2-bending vibration and –C H_3asymmetric bending vibration, 1374 cm^{-1} is the absorption peak of symmetrical bending vibration of –C H_3,the presence of these functional groups shows that there are many kinds of saturated long-chain alkanes, aromatic hydrocarbons and long-chain hydrocarbons in asphalt [14, 15]. Comparing the absorption spectra of three kinds of asphalt, we can find, as the aging time increases, the light transmittance decreases and the absorption increases, it shows that the polymerization of short-chain hydrocarbons during the aging process of asphalt produces –C H_2-and –C H_3, enhance the absorption of these two functional groups, as the fragrance and gel reduce, causing the reduction of the asphalt penetration.

(2) As can be seen in Figure 2, there are obvious absorption peaks at 725 cm^{-1}, 1159 cm^{-1}, 1376 cm^{-1}, 1463 cm^{-1}, 1710 cm^{-1}, 1743 cm^{-1}, 2852 cm^{-1}, 2931 cm^{-1}, and 3450 cm^{-1} in the waste oil residue (REOB). The band of the 3000–2800 cm^{-1} region is caused by C-H stretching vibration of CH_2 or CH; the 2000–1680 cm^{-1} region is a series of weaker bands; they are the absorption of the frequency and group frequency of the corresponding aromatic ring, the position and number of these bands indicate the presence of a monosubstituted aromatic ring in REOB; the band at 725 cm^{-1}, belonging to out-of-plane stretching vibration of protons on five adjacent carbons of the benzene ring, and it is a characteristic band of typical monosubstituted benzene. The two absorption peaks of 1710 cm^{-1} and 1743 $cm^{-1}$are caused by ketones or anhydrides, these three types of substances are oxidation products during the using of oil, and it can balance with the oxidation products in aged asphalt, thereby in hibite the continued aging of the asphalt; these three absorption peaks of 725 cm^{-1}, 1376 cm^{-1}, and 1463 cm^{-1} indicate that REOB contains aromatic hydrocarbons and colloids and can supplement the aromatic hydrocarbons and colloidal reduction caused by asphalt aging, so REOB helps to adjust the composition of aged asphalt, improve the performance of aged asphalt, and achieve good regeneration.

3.2. Performance Comparison between Recycled Asphalt and Aged Asphalt. We add different proportions of waste engine oil residues to recycled natural aged 10 years of asphalt, conduct basic physical index test, compare the three major indicators of aged asphalt and matrix asphalt, and discuss regeneration effect of REOB on aged asphalt. The test results are shown in Table 2.

The three major indexes of asphalt: penetration, ductility, and softening point, are the conventional physical performance indexes of asphalt, which have certain experience. Penetration can be used to measure the degree of softness and hardness of asphalt and its ability to resist shear failure. The larger the value, the softer the asphalt and the lower the viscosity. Ductility represents the plasticity of asphalt and is used to characterize the low-temperature performance of asphalt. The larger the value, the better the plasticity. The higher the softening point, the worse the temperature sensitivity of the asphalt and the better the heat resistance of the asphalt. The regeneration ability of REOB can be evaluated by comparing the changes of physical properties before and after regeneration of aged asphalt.

As can be shown from the above table, adding REOB to aged asphalt can effectively improve the penetration reduction and the decrease of ductility caused by aging, but the improvement effect on the softening point is not significant. With the increase in the amount of REOB added, the penetration of aged asphalt increases, the ductility increases first and then decreases, and the softening point increases. Comparing the test results of the three added amounts, we find that the best amount of REOB recycled aged asphalt is 6%; at this time, the penetration and elongation of the aged asphalt have been moderately improved, but the reducing of the softening point still needs to be adjusted by adding other regenerant. From the above, waste oil residue can be used as a regenerant for aged asphalt to improve its low-temperature

Table 2: Test results of aged asphalt and matrix asphalt after regeneration of different proportions of REOB.

	Penetration / 0.1 mm	Ductility (10°C) / cm	Softening point / °C
70# matrix asphalt	67	20	48.4
Recycling aged 10 years of asphalt	24	4.2	63
REOB added 4%	42	6.4	57.2
REOB added 6%	68	12.8	59.2
REOB added 8%	75	10	62

crack resistance, but it needs to be combined with other regenerants for regulating the softening point.

4. Conclusions and Prospects

Waste oil recovery residue is an environmental pollutant; with the development of China's economy, the process of recycling is becoming more and more mature, and the emissions of waste oil recovery residues are also increasing. Research shows that residues of waste oil contain oxides that inhibit the continued aging of asphalt, and REOB contains a certain amount of aromatic and colloidal, and it can be used as a regenerant for component adjustment of recycled bitumen. This discovery can reduce pollutant emissions, realize the recycling of waste materials, and increase economic efficiency. But, as a regenerant for aged asphalt, REOB can only restore some indicators of asphalt, and there is still a certain gap between the requirements for the use of asphalt. Also, the production technology and construction technology of REOB regenerated aged asphalt are not mature yet. These are the directions that need to be studied and considered in the future.

Data Availability

The data supporting the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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