Viscoelasticity Evaluation of Regenerated Asphalt Containing Waste Engine Oil Based on Rheological Analysis

YAJUN Xiao 1, WENTONG Wang2,*, WENWEN Wang3, DONGDONG, Yuan2

1Shaanxi Railway Institute, Weinan Shaanxi 714000, China;
2School of Highway, Chang'an University, Xian 710064, China;
3Department of Information Engineering, Shandong Labor Vocational and Technical College, Jinan, 250022, China

YAJUN Xiao e-mail address: 2015121251@chd.edu.cn
*Corresponding author. WENTONG Wang e-mail address: wentongwang@chd.edu.cn
WENWEN Wang e-mail address: wangwenweny@sdlvtc.cn
DONGDONG Yuan e-mail address: ddy@chd.edu.cn

Abstract. In order to explore the viscoelastic properties of waste engine oil (WEO) on mixed asphalt, mixed asphalt was prepared by fusing base asphalt and aged asphalt. The effect of waste engine oil content on the physical and viscoelastic properties of asphalt mixtures was studied through experiments. The test results show that the viscosity and softening point of asphalt mixture is reduced by adding waste engine oil. The complex modulus (G*) of asphalt binder decreases with the addition of waste oil at different test frequencies. With the increase of waste oil content, the phase angle (δ) of asphalt mixtures increases at specific frequencies. When the content of waste oil exceeds 6%, G* and δ will not change significantly. The stiffness increase caused by ageing asphalt in mixed asphalt will be offset by adding 2-4% waste oil. This indicates that waste oil can be used as regenerant to provide elasticity and flexibility for asphalt binder.

1. Introduction
It is widely known that, the transportation sector consumes a lot of motor oil. With the continuous operation of the engine, the performance of the engine oil gradually decreases and is eventually replaced, and the replacement becomes waste engine oil (WEO)[1]. As the molecular structure of WEO and asphalt is similar, based on the principle of similar compatibility, it is possible to make modified or regenerated asphalt compatible with WEO and asphalt binder[2]. According to statistics, my country's transportation industry alone generates 25-30 million tons of WEO every year[3], so the full use of waste engine oil will play a role in resource conservation and environmental protection[4].

In the process of using asphalt pavement, aging will inevitably occur, which will increase the brittleness, hardness and elasticity of the asphalt binder[5]. The decrease of asphalt performance will cause fatigue cracking, temperature cracking and pothole damage during the use of asphalt mixture[6]. In addition, the workability loss will also occur during the mixing construction process[7]. Therefore, it is quite challenging to use aged asphalt to replace part of the base asphalt in road construction. Before
adding aged asphalt into the asphalt mixture, the performance of the latter can be improved in a targeted manner, which can make its utilization efficiency higher and its performance more perfect.

Since WEO has the ability to reduce the viscosity of asphalt and soften asphalt, it can act as a regeneration agent [8]. Villanueva et al. (2008) pointed out that the addition of WEO in asphalt would reduce the softening point of modified asphalt [9]. Borhan et al. (2009) showed that WEO reduces the ductility and specific gravity of modified asphalt [10]. Recently, Paliukaite et al. studied the addition of WEO to direct distillation and polymer modified asphalt. The test results showed that with the increase of WEO content, the high and low temperature grades of modified asphalt increased to varying degrees [11]. Zhao Peisong et al. studied the regeneration and aging asphalt of waste engine oil. The results showed that adding WEO can improve the fatigue resistance of asphalt, and the addition of WEO can improve the grade of asphalt, so it has higher economic benefits.

The purpose of this research is to use WEO as the regeneration agent of asphalt mixture to analysis the effect of WEO on the viscoelastic properties of medium and high temperature asphalt binder, so as to seek the optimal WEO content of recycled aging asphalt, which is helpful to environmental protection, reduce energy consumption and reduce construction cost.

2. Experiment Part

2.1 Materials and Instruments

2.1.1 Asphalt. The asphalt is Jingbo 70# matrix asphalt, its technical indicators are shown in Table 1. Asphalt mixture old material (RAP) from Jiqing highway reconstruction Jinan section road milling recycling old material. According to the requirements of “《Highway engineering asphalt and asphalt mixture test procedures》” [13], trichloroethylene was used as a solvent, and the aged asphalt was recovered and extracted from RAP material by centrifugal extraction technology, and the physical properties of the asphalt were tested. Different indexes are shown in Table 1. Stored the recycled aged asphalt in a container and sealed.

| Performance                           | 70 # asphalt | recycling ageing asphalt | specification value |
|---------------------------------------|--------------|--------------------------|---------------------|
| Softening point, °C                   | 46.9         | 67.0                     | >46.0               |
| Penetration (25°C, 5s, 100g) 0.1mm    | 72.5         | 80                       | 60.0-80.0           |
| Brookfield Viscosity (135°C) mPa.s    | 497          | 7120                     | -                   |
| Flash point, °C                       | 275          | 300                      | ≥260.0              |

WEO was taken from a 4S repair shop of a car in Tianjin, which was the engine oil after the replacement of the car with a mileage of 250 million kilometers. WEO was brown and black, and the hand twist was not granular.25°C dynamic viscosity is 54 mPa·s, flash point and specific gravity are 186 °C and 0.86 respectively.

2.2 Preparation of Modified Asphalt
Aged asphalt (250g) was added to base asphalt (1000g) (JB-70) (25%) to make mixed asphalt. At 160°C, WEO was slowly added to the asphalt mixture according to different dosages, and continuously stirred for 30 min until the mixture was evenly mixed. Reference to previous research experience, determine the WEO content range of 0% to 8%, incremental 2% The recycled asphalt mixed with waste oil of 0%, 2%, 4%, 6% and 8% is denoted as JB-25-0, JB-25-2, JB-25-4, JB-25-6 and JB-25-8. The matrix asphalt without ageing asphalt and waste oil is denoted as JB-0-0.
2.3 Performance Testing
Brookfield viscometer was used to test the viscosity of recycled asphalt before and after ageing at different temperatures (110°C, 135°C, 150°C and 180°C). The softening point of recycled asphalt with different WEO content was tested by global softening point test. Rotating film ageing (RTFOT) was used for short-term ageing of recycled asphalt, and the test temperature was set to 163°C.

The changes of the chemical bond and chemical structure of asphalt before and after regeneration were qualitatively and quantitatively analyzed by using Fourier transform infrared spectroscopy (FTIR) test and measuring the infrared absorption value of the material. When the sample is irradiated by infrared rays, the molecular bonds in the material vibrate or rotate at discrete frequencies. When the molecular bond vibrates/rotates, part of the radiation is absorbed, and the other part is transmitted through the material. After measurement by the detector, the corresponding absorption spectrum is given to determine the function wavelength. Because the spectrum of each molecular structure is unique, it is convenient and accurate to identify the functional groups in the material. In this study, Bruker-Alpha-FTIR spectrometer was used with the wavelength range of 500 - 4000 cm⁻¹.

The complex modulus and phase angle of recycled asphalt before and after aging were measured by Anton Paar MCR 102 dynamic shear rheometer (DSR). The steel plate with a diameter of 25 mm and a thickness of 1 mm was used in the experiment. The strain amplitude was 0.1% and the frequency scanning range was 0.01–10Hz. In order to explore the viscoelastic properties of recycled asphalt at medium and high temperature, the test temperature is set to 36°C -60°C, 6°C.

3. Analysis of test results

3.1 Chemical composition
As mentioned above, FTIR spectroscopy is used to study the changes of functional groups of WEO and recycled asphalt before and after aging. Infrared spectra of WEO recycled asphalt with different contents after non-aging and short-term aging are shown in Figs 1. (b) and (c). By comparing the absorption spectra of asphalt with different WEO contents, it is found that there are significant differences among the peaks. For a particular asphalt, changes in absorption peak values can also be observed before and after short-term aging. This indicated that the functional groups changed with WEO incorporation and short-term aging. By comparing the spectra of asphalt before and after aging, it was found that the absorption spectra of asphalt with wavelength less than 1400 cm⁻¹ decreased due to the addition of WEO, indicating the change of olefins. For a particular asphalt, a small peak (corresponding to sulfoxide group) appears at 1010cm⁻¹ wavenumber after short-term aging compared with before aging. The absorbance of 3000 - 4000cm⁻¹ wavenumber decreased during short-term aging.

(a) Waste Engine Oil
In order to further explain the results of infrared spectrum changes, different functional group indexes were used for quantitative analysis[14]. Different exponential expressions are listed in Formulas (1) - (4). Generally speaking, aromatics, carbonyl, sulfoxide index has a similar trend. In the absence of aging, the above three indicators are increased with the increase of WEO content. However, after short-term aging, with the increase of WEO content, each index decreased. This indicates that asphalt aging leads to the increase of aromatic index and the decrease of aliphatic compounds, as shown in Fig 2. For a particular asphalt, short-term aging can lead to increased carbonyl and sulfoxide content. Therefore, the addition of WEOB can increase the aromatic, carbonyl and sulfoxide index. Other researchers have similar results[15].

\[
I_{AI} = \frac{A_{2757-3035}}{\sum A}
\]

\[
I_{Ar} = \frac{A_{1394-1518}}{\sum A}
\]

\[
I_{C=O} = \frac{A_{1607-1705}}{\sum A}
\]

\[
I_{S=O} = \frac{A_{974-1064}}{\sum A}
\]

In the formula:  \( I_{AI} \) represents the aliphatic index, \( I_{Ar} \) represents the aromatic index, \( I_{C=O} \) represents the hydroxyl index, \( I_{S=O} \) represents the sulfoxide index, \( I_{a-b} \) is the peak area of wave number \( a \) cm\(^{-1}\) to wave number \( b \) cm\(^{-1}\), \( \sum A \) is the sum of peak area of wave number 500-4000 cm\(^{-1}\).
3.2 Viscosity

Viscosity of recycled asphalt before and after aging was measured by Brookfield viscometer. Two repeated tests should be carried out at a certain measuring temperature. Under different temperature conditions, different asphalt viscosity values before and after short-term aging are shown in Figs 3. (a) and (b).

The relationship between viscosity and temperature of asphalt can be described by exponential function, as shown in Formula (5) Regression analysis is used to determine the regression coefficient, so as to minimize the sum of squares of error between measured and predicted values. Table 2 lists the regression coefficient (C) and correlation coefficient (R^2) values of different asphalt. For all WEO modified asphalt, the addition of WEO makes C1 (intercept) decrease, which shows that the addition of WEO will lead to the viscosity of asphalt at different temperatures. Comparing all recycled asphalts, it can be found that with the increase of WEO content, the absolute value of C2 decreases, indicating that WEO can reduce the temperature sensitivity of asphalt binder. The results show that the negative exponential equation can well describe the viscosity-temperature relationship of asphalt.

\[ \omega = c_1 \times e^{c_2 \times t} \]  

In the formula: \( \omega \) represents the viscosity of asphalt at a certain temperature (t), \( c_1 \) represents the intercept of the regression equation, \( c_2 \) represents the slope of the regression equation.

| Ageing Conditioning | Unaged | Aging |
|--------------------|--------|-------|
| Kind of Asphalt    | Unaged | Aging |
| JB-0-0             | 472.163 | -0.048 | 9875 | 1154.821 | -0.054 | 9875 |
| JB-25-0            | 464.557 | -0.045 | 9847 | 1356.724 | -0.052 | 9821 |
| JB-25-2            | 448.351 | -0.043 | 9833 | 895.346 | -0.05 | 9814 |
| JB-25-4            | 94.768  | -0.042 | 9769 | 546.287 | -0.049 | 9804 |
| JB-25-6            | 78.432  | -0.04 | 9765 | 502.467 | -0.047 | 9789 |
| JB-25-8            | 35.158  | -0.037 | 9485 | 268.157 | -0.046 | 9789 |

Figure 2. Functional Group Index Under Different WEO Content and Aging Conditions
As shown in Fig3, for a particular asphalt, the viscosity value of the asphalt decreases with the increase of temperature. This is also the same as the trend of the absolute value of coefficient C2 in Table 2. It can be seen from the figure that before and after asphalt aging, the viscosity value of JB-25-0 asphalt binder is the highest under different temperature conditions. The reason may be due to the hardening effect of aged asphalt extracted from RAP. At all given temperature conditions, adding WEO will reduce the viscosity of asphalt. The test results show that the recycled asphalt of JB-25-2 has similar viscosity to that of matrix asphalt (JB-0-0). Further comparison shows that before and after the aging of recycled asphalt, when the content of WEO exceeds 2%, the viscosity of asphalt will be reduced. The possible reasons for this phenomenon are: 1) WEO has regeneration performance; 2) Physical and/or chemical reactions occur between base asphalt, WEO and aged asphalt.

### 3.3 Softening point

The softening point of recycled asphalt before and after aging was determined in this research, as shown in Table 3. Under the conditions of non-aging and short-term aging, the changes of softening points of different asphalts are shown in Fig 4. It can be seen from the figure that the softening point of asphalt after short-term aging is significantly increased compared with the non-aging asphalt. This can be attributed to the volatilization loss of light components in asphalt due to aging.

| Kind of Asphalt | JB-0-0 | JB-25-0 | JB-25-2 | JB-25-4 | JB-25-6 | JB-25-8 |
|----------------|--------|---------|---------|---------|---------|---------|
| Unaged          | 46.9   | 54.5    | 46.2    | 44.3    | 41.1    | 38.5    |
| Short-term Aging| 55.2   | 59.8    | 57.1    | 55.4    | 53.4    | 51.5    |
As the softening point decreased with the increase of WEO content, the relationship between softening point and WEO content was analyzed by regression analysis, so as to find the best fitting function. It can be seen from the figure that when the regression coefficient $R^2$ is used as the evaluation standard, the relationship between softening point and WEO content can be well fitted under two aging conditions. Formulas (6) and (7) respectively list the final fitting negative exponential function of asphalt after non-aging and short-term aging. The regression coefficients of non-aging and short-term aging conditions were 0.9687 and 0.9971, respectively. The negative exponential function curve shows that the softening point can be significantly reduced by adding WEO at low doses, while it can be ignored at high doses due to the small effect.

\[
SP_{una} = 53.782e^{-0.045x} \tag{6}
\]

\[
SP_a = 59.685e^{-0.016x} \tag{7}
\]

In the formula: $SP_{una}$ represents the softening point of non-aging, $SP_a$ represents the softening point after aging, $x$ represents the percentage of WEO content in asphalt.

### 3.4 Complex Modulus

Figs. 5. (a) and 5 (b) show the main curves of complex modulus of different asphalts without and after short-term aging at 48°C. It can be seen from the figure that before and after aging, with the increase of reduction frequency, the complex modulus values of all asphalts show an upward trend, and the complex modulus of JB-25-0 recycled asphalt is the highest at different reduction frequencies. With the increase of WEO content, the complex modulus of asphalt gradually decreases at different reduction frequencies. This shows that the incorporation of WEO makes the hardness of asphalt mixture containing aging asphalt decrease. The main curves of JB-0-0 are close to JB-25-4 and JB-25-2, respectively, under the condition of non-aging and short-term aging. Before and after aging, the complex modulus values of JB-25-6 and JB-25-8 are very close at all frequencies. This shows that the addition of WEO beyond a certain level may not have obvious regeneration effect.

### 3.5 Phase Angle

Figs 5. (a) and (b) show the variation of asphalt phase angle ($\delta$) under different WEO contents under the conditions of non-aging and short-term aging, respectively. It can be seen from the figure that the reduction frequency is negatively correlated with $\delta$, and it reaches the 90° extremum of asphalt $\delta$ at the minimum reduction frequency. However, the limit $\delta$ at the highest frequency depends on the WEO content in recycled asphalt. For a specific asphalt, under all frequency conditions, the $\delta$ of unaged asphalt is higher than that after short-term aging. The maximum $\delta$ was observed in JB-25-6 and JB-25-8 before and after aging. Under different reduction frequencies, JB-25-0 has the smallest $\delta$. In addition, the $\delta$
principal curves of JB-25-6 and JB-25-2 recycled asphalt and JB-0-0 matrix asphalt are the closest under the conditions of no aging and short-term aging.

![Graphs showing complex modulus and phase angle principal curves for different asphalts](image)

**Figure 5. Complex Modulus and Phase Angle Principal Curve of Different Asphalts**

### 4. Conclusion

The influence of WEO on asphalt mixture (including base asphalt and RAP extraction recycled asphalt) was studied through laboratory test. Therefore, according to different aging conditions, the recycled asphalt with different proportion of matrix asphalt, recycled asphalt and WEO content was prepared. Frequency-temperature scanning tests were carried out on all prepared recycled asphalts, and the test results of composite modulus and phase angle were obtained. The main curves of composite modulus and phase angle were established, and the viscoelastic properties of composite asphalt materials were studied. The conclusions of this study are as follows:

(a). The addition of WEO reduces the viscosity and softening point of asphalt mixture. The negative exponential function can reasonably describe the viscosity-temperature relationship. At the same time, this function can also well fit the relationship between softening point and WEO content.

(b). At any given temperature, the addition of WEO results in (i) a decrease in the complex modulus and (ii) an increase in the phase angle at all reduced frequencies.

(c). Under short-term aging conditions, the main curves of complex modulus and phase angle obtained by adding 2 % WEO are similar to those of the control group. This shows that at the dosage of 2%, WEO can weaken the stiffness increase caused by adding aging asphalt.

(d). In view of the short-term aging conditions used in this experiment, the optimum dosage of WEO was 2-4%. With the increase of WEO dose, the regeneration effect will gradually decrease.

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