

## Article

# Preparation and Properties of Waterborne Epoxy-Resin-Emulsified Asphalt Modified by Oxidized Extraction Oil

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**Abstract:** Waterborne epoxy-resin-emulsified asphalt (WEREA) has excellent adhesion and can be used as a good waterproofing tack coat; however, there are some problems such as the poor compatibility between the waterborne epoxy and the emulsified asphalt, and the brittleness of the cured material. In the present work, oxidized furfural extract oil was used as a compatibilizer to prepare the waterborne epoxy emulsion and waterborne epoxy-resin-emulsified asphalt, and their modification effects were studied. The extraction oil was oxidized with potassium permanganate. The effects of oxidized extraction oil on the waterborne epoxy-resin-emulsified asphalt performance were investigated through experiments on viscosity, mechanical properties, and aging resistance. Combined with infrared spectroscopy and fluorescence microscopy, the compatibility and microstructure of the oxidized extraction oil modified WEREA were observed and analyzed. The result showed that the carboxyl group was introduced into the chemical structure of the extraction oil after oxidation. Oxidized extraction oil (OEO) and waterborne epoxy resin (WER) had good compatibility. When the content of OEO in the WER is 21%, the elongation at break of the WER can reach up to a maximum of 91.5%, and has a significant increase of 33.2%. OEO can significantly improve the elongation at the break and aging resistance of WEREA, especially when the mix ratio of oxidized extraction oil and epoxy resin was 6:5, when the breaking elongation of WEREA can be increased by 69%, and the compatibility between the epoxy resin and emulsified asphalt was the best. Moreover, the loss in elongation at the break of aged WEREA decreased from 13.7% to 4.9%.

**Keywords:** furfural extraction oil; mechanical property; compatibility; toughening; emulsified asphalt



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## 1. Introduction

Emulsified asphalt has the advantages of simple construction, energy-saving and environmental protection, being less limited by construction season, etc. Therefore, it has been widely used in the tack coat oil of asphalt pavement and cold repair asphalt mixture [1–3]. However, in the application of road pavement materials, the characteristics of asphalt flowing at high temperatures and becoming brittle at low temperatures remain unchanged [4–6]. Emulsified asphalt still has the shortcomings of low early strength, weak adhesion, poor temperature stability, etc., which severely restricts its further application [7–9]. Emulsified asphalt modified with polymer is the most common and effective method to improve its performance [10–12]. Waterborne epoxy resin takes water as the dispersion medium, which is safe, pollution-free, and has good fluidity [13–15]. Epoxy resin is a thermosetting polymer with a three-dimensional network structure after reacting with the curing agent; the cured product has excellent mechanical properties, strong adhesion, and good heat resistance [16–18]. Waterborne epoxy-resin (WER)-modified emulsified asphalt not only retains the excellent properties of emulsified asphalt with high viscoelasticity and convenient construction but also endows it with the characteristics of high adhesion and marvelous high-temperature performance [19–21]. Relevant researches show that waterborne epoxy resin has a remarkable effect on the modification of emulsified asphalt, which is mainly reflected in cutting down emulsified asphalt's temperature sensitivity, increasing fatigue resistance and durability, and

providing superior substrate adhesion [21–23]. For example, Fu H. et al. [3] prepared waterborne epoxy-modified emulsified asphalt with excellent adhesion, mechanical strength, and fatigue life by using a self-synthesized waterborne curing agent. The addition of WER reduces the brittleness of the binder. After aging, the tensile strength retention rate of WEREA is above 88%, indicating sufficient durability and fatigue life. When the content of WER increased from 15% to 20%, the effect of improving the adhesion of WEREA to aggregate is obvious, and the shedding rate of binder decreased by about 45–55%. The WEREA can be used as the binder for cold cycle and cold mix asphalt pavement. Yin Y. et al. [23] introduced a new micro-surfacing binder of WEREA. The results showed that WEREA can significantly enhance the adhesion between asphalt binder and aggregates. The content of WER has the most significant effect on improving the adhesion between asphalt binder and acid aggregate. The addition of 20% WER apparently reduces the spalling of asphalt film on the surface of aggregates. Nevertheless, the cured epoxy resin with a high crosslinking density is brittle and prone to crack [24]. A high dosage of epoxy resin makes epoxy-emulsified asphalt hard and brittle, causing performance degradation [15,25]. Researchers have carried out many studies on the toughening modification of epoxy asphalt. The toughness of epoxy asphalt is mainly improved by adding hyperbranched polyester, rubber, fiber, and nanoparticles [26–28]. Xu P. et al. [28] introduced hyperbranched polyester (HBP) with low viscosity, high branching, and many functional end groups to improve the toughness of epoxy resin. The results showed that HBP-modified epoxy asphalt can effectively improve the elongation of epoxy asphalt and maintain sufficient tensile strength. Su W. et al. [27] toughened epoxy asphalt by adding 100–200 nm nano core-shell rubber (CSR). The microstructure showed that the interaction between glass shell and asphalt led to the fracture of the glass shell of CSR nanoparticles; the rubber shell of CSR nanoparticles expanded in microscale in epoxy asphalt, and the newly expanded CSR particles were evenly dispersed in epoxy resin. Mechanical studies showed that CSR nanoparticles greatly improved the mechanical properties of pure epoxy resin. The epoxy asphalt mixture with 1% CSR nanoparticles had the maximum fracture elongation and toughness, which were increased by 60% and 200%, respectively, compared with pure epoxy asphalt. Zhang J. et al. [29] studied the softening and toughening effect of basalt fiber on epoxy asphalt. The results showed that the reinforcement and toughening effect of basalt fiber can prevent or delay the generation and extension of cracks in epoxy asphalt materials, thus improving the fatigue resistance and low-temperature crack resistance of epoxy asphalt mixtures.

Compatibility has been another reason for the unsatisfactory performance of epoxy asphalt; the problem of poor compatibility mainly comes from the difference in physical and chemical properties such as polarity and solubility parameters between epoxy resin and asphalt [30–32]. Currently, the methods of improving the compatibility of epoxy asphalt mainly include the modification of asphalt or curing agents and the addition of compatibilizers. Yang K. et al. [33] grafted maleic anhydride onto asphalt to improve the polarity of asphalt and promote the compatibility between asphalt and epoxy resin. The results showed that the modified asphalt has good compatibility with epoxy resin. Si L. et al. [34] improved the compatibility of asphalt and epoxy resin by introducing epoxidized soybean oil (ESO), and the dynamic mechanical analysis results showed that with the increase of ESO content, the glass transition temperature  $T_g$  of cold-mixed epoxy asphalt gradually changed from two to one, indicating that the addition of epoxy soybean oil makes asphalt and epoxy resin have good compatibility.

Furfural extraction oil is an industrial by-product separated from lubricating oil production and contains a large number of monocyclic, bicyclic, and polycyclic aromatic hydrocarbons, and a small number of saturated hydrocarbons, colloids and asphaltenes [35]. Its aromatic content is as high as 50–80%, belonging to the typical high aromatic oil. In the comprehensive utilization and research of furfural extraction oil, it was found that the high aromatic hydrocarbons in the extraction oil can greatly improve the dispersion degree of asphaltene and ductility of asphalt [36,37]. Therefore, the extraction oil is widely used in blending road asphalt and has been proved to be an ideal softening component which can

effectively improve the ductility and other properties of asphalt; however, the extraction oil is incompatible with polar epoxy resin, and it cannot be directly used as a compatibilizer of waterborne epoxy-resin-emulsified asphalt.

In this article, the oxidation-treated extract oil is used to improve its compatibility with epoxy resin. The aim is to study the toughening effect of oxidized extraction oil on waterborne epoxy-resin-emulsified asphalt and the relationship between microstructure and mechanical properties of the blend system of waterborne epoxy-resin-emulsified asphalt modified by oxidized extraction oil. Part of the chemical structure of the furfural extract oil is oxidized to improve the chemical polarity of the extraction oil so that it can be compatible with water-based epoxy resin but also has excellent solubility with asphalt. As a compatibilizer of water-based epoxy resin and emulsified asphalt, it can not only be used to prepare a stable water-based epoxy resin emulsion but also be used to prepare a good toughness-modified waterborne epoxy-resin-emulsified asphalt with excellent aging resistance.

## 2. Materials and Methods

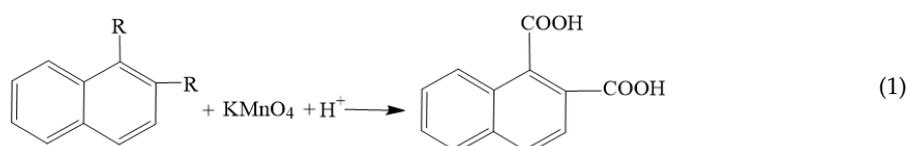
### 2.1. Materials

The emulsified asphalt used in the experiment is cationic emulsified asphalt from Xin Ji Yong Xing Emulsion Asphalt factory. Solid content of the emulsified asphalt is 60%. E51 epoxy resin and waterborne curing agent are respectively from Shan Dong Pin Shang New Material Company and Wuxi Da He Polymer Materials Co., LTD. (Yixing, China). The extraction oil is supplied by Hebei Jiale Petroleum Technology Co., LTD.

### 2.2. Preparation Process

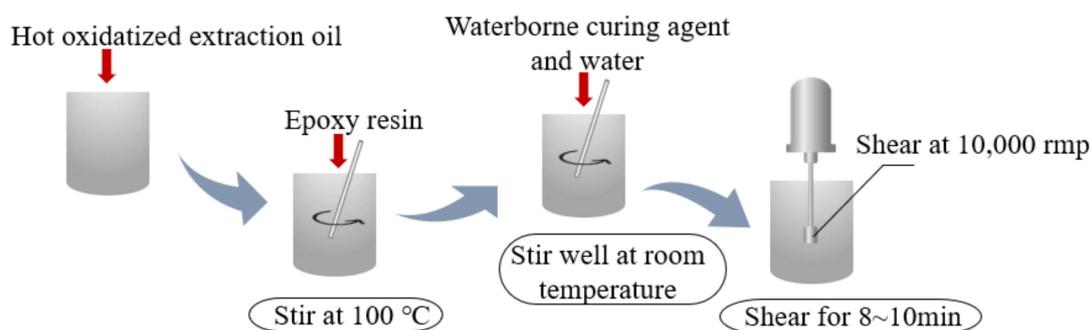
#### 2.2.1. Extraction Oil Oxidation Pretreatment

Briefly, 0.5% catalyst (Dodecyl trimethyl ammonium chloride, from Bo Xing Hao Long Chemical Co., Ltd. (Binzhou, China)), 1% phosphoric acid (Nanjing Chemical Reagent Factory No. 1 (Nanjing, China)), and hot-melt extraction oil were added into a three-mouth flask and placed in an intelligent magnetic mixer. The prepared 0.5 mol/L potassium permanganate (Guangdong Xilong Chemical Factory (Shantou, China)) solution was put into the separating funnel and added into the mixture gradually. A stirrer and condensate reflux device were installed in a three-mouth flask. The mixture was stirred and reacted at 110 °C for 6 h to obtain oxidized extraction oil (OEO). The oxidation mechanism of extraction oil is shown in Formula 1.



#### 2.2.2. Preparation of Extraction Oil/Waterborne Epoxy Resin System

Firstly, the hot-melt oxidized extraction oil and epoxy resin were mixed evenly; then waterborne curing agent and water were added and sheared at the speed of 10,000 revolutions per minute (rpm) for 8~10 min after mixing. The ratio of epoxy resin and curing agent is 1:1. The solid content was 60%. Finally, uniform waterborne epoxy resin emulsion modified by oxidized extraction oil was obtained. Waterborne epoxy resin emulsion modified by oxidized extraction oil is marked as WEO. The mass ratio of epoxy resin to oxidized extraction oil is 5:2, 5:3, 5:4, and 5:5, marked as WEO-2, WEO-3, WEO-4, and WEO-5, respectively. The preparation process is shown in Figure 1.



**Figure 1.** Preparation process of WER modified by oxidized extraction oil.

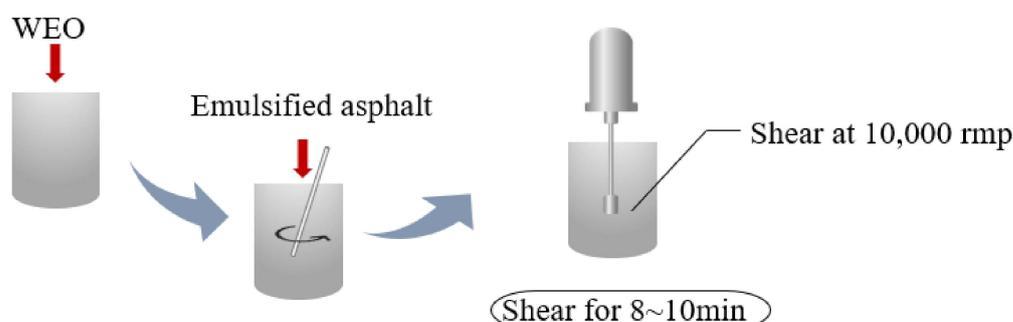
### 2.2.3. Preparation of Extraction Oil/Waterborne Epoxy-Resin-Emulsified Asphalt

Waterborne epoxy-resin-emulsified asphalt (WEREA) was prepared according to the mass ratio in Table 1. When the mass ratio of epoxy resin/oxidized extraction oil is 5:4, 5:5, 5:6, or 5:7, the waterborne epoxy-resin-emulsified asphalt modified by oxidized extraction oil is marked as WEAO-4, WEAO-5, WEAO-6, and WEAO-7 in the order given.

**Table 1.** Mass ratio of oxidized extraction oil/waterborne epoxy-resin-emulsified asphalt.

Samples	Epoxy Resin	Curing Agent	Emulsified Asphalt	Oxidized Extraction Oil	Deionized Water
WEREA	5	5	5	0	6.67
WEAO-4	5	5	5	4	9.33
WEAO-5	5	5	5	5	10
WEAO-6	5	5	5	6	10.67
WEAO-7	5	5	5	7	11.33

The preparation process is shown in Figure 2.



**Figure 2.** Preparation process of WEREA modified by oxidized extraction oil.

The preparation process of WEREA modified by oxidized extraction oil is as follows: first, the WEO was prepared as shown in Figure 1, and then a certain amount of emulsified asphalt was added into WEO and sheared at 10,000 rpm for 8~10 min.

### 2.3. Characterizations

The viscosity of WEO or WEREA modified by oxidized extraction oil was measured using a Brookfield viscometer. The tensile strength and fracture elongation were measured using a universal testing machine at a load rate of 10 mm/min. According to the International Standard EN ISO 4624:2002, the pull-off tests were carried out to assess the adhesive strength between the coating and the aluminum plate. The aging resistance of samples was evaluated by studying the change of tensile properties after exposure to ultraviolet (UV) at 60 °C for 24 h. The molecular structure and functional groups of materials were characterized by

Fourier transform infrared spectroscopy (FTIR). The compatibility was investigated using a fluorescence microscope.

### 3. Results and Discussion

#### 3.1. Performance Analysis of Oxidized Extraction Oil

##### 3.1.1. FTIR

The change of the chemical structure of extracted oil after potassium permanganate oxidation treatment was verified by infrared spectroscopy. The infrared spectrum of extraction oil (EO) and oxidized extraction oil (OEO) is shown in Figure 3. There are many sharp aromatic hydrocarbon absorption bands between  $650\text{ cm}^{-1}$  and  $1000\text{ cm}^{-1}$ . Comparing the characteristic absorption peaks of EO and OEO, there is almost no change in the CH characteristic absorption peaks of the C-H bonds. The result shows that the main chemical structure of the extracted oil has not changed significantly after oxidation; however, there is an obvious characteristic absorption peak of the O-H bond on the carboxyl group. This demonstrates that part of the carbon on the naphthalene ring in the extraction oil is oxidized to carboxyl groups by potassium permanganate, and the hydrophilic polar groups are introduced into the chemical structure of the extraction oil. This situation is also consistent with the chemical fact that potassium permanganate can oxidize the hydrocarbon group on the benzene ring to the carboxyl group.

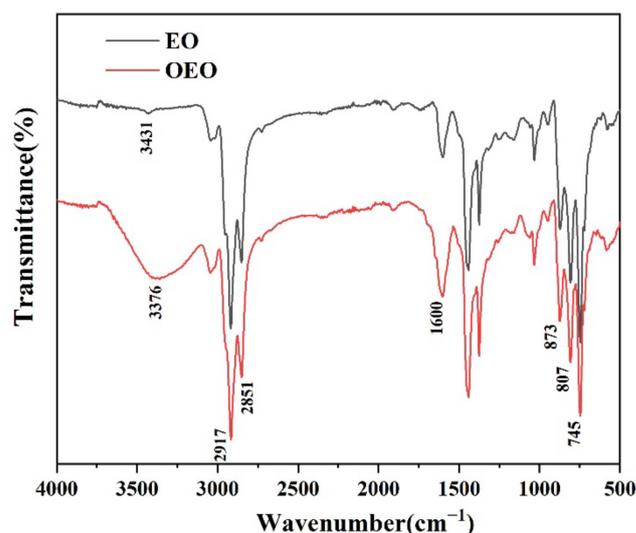


Figure 3. FTIR spectra of extraction oil (EO) and oxidized extraction oil (OEO).

##### 3.1.2. Emulsion Properties Comparison of Extraction Oil before and after Oxidation

The properties of the emulsion after mixing the extraction oil and oxidized extraction oil with waterborne epoxy resin were studied. The experimental results are shown in Table 2. The oxidized extraction oil introduced a hydrophilic group carboxyl group, which has good compatibility with water-based epoxy resin, so it is easy to shear and the prepared emulsion is uniform and stable, without color difference, and with no agglomeration. Therefore, oxidized extraction oil with good performance was adopted to modify the epoxy-emulsified asphalt.

Table 2. Properties of extraction oil/waterborne epoxy emulsion before and after oxidation.

Properties	EO/WER Emulsion	OEO/WER Emulsion
Shear difficulty	Difficult	Easy
Emulsion uniformity	Homogeneous	Non-homogeneous
Stability	Layered, separated	Stable

### 3.2. Extraction Oil/Waterborne Epoxy Resin System

#### 3.2.1. Emulsion Properties

Table 3 shows the influence of oxidized extraction oil content on the emulsification process, stability, drying time, and film formation of the WEO system. It can be seen that the drying time of WEO (surface drying and solid drying) is extended with the increase in OEO content. The higher the oxidized extraction oil content, the slower the drying rate. The emulsifying effect, stability and film homogeneity of WEO-5 are not as good as WEO-4, and a slight segregation phenomenon occurs.

**Table 3.** Performance of WEO system with different contents of extraction oil.

Samples	Shear Difficulty	Appearance	Drying Time/h		Film Forming Property
			Surface Dry Time	Real Drying Time	
WER	Easy	Homogeneous, no-stratification	4	22	Good
WEO-2	Easy	Homogeneous, no-stratification	4.2	22.3	Good
WEO-3	Easy	Homogeneous, no-stratification	4.8	23	Good
WEO-4	Easy	Homogeneous, no-stratification	5.3	23.5	Good
WEO-5	Relatively easy	Relatively non-homogeneous slight segregation	6	24.2	Fairly good

#### 3.2.2. Viscosity

An irreversible curing reaction begins to occur when epoxy resin and curing agent are mixed. It is gradually transformed from a viscous state to a solid polymer with certain strength. Therefore, the viscosity of WEO increases with time. The change in viscosity over time can evaluate the initial cure rate of WEO. To ensure that the coating has certain operability and workability during the construction process, the viscosity of the system should maintain an appropriate viscosity during the operation time. The operating time is for the viscosity to reach 3000 mpa·s time spent [38]. In addition, the curing speed should not be too slow. If the coating is cured too slowly, it may cause problems such as low early strength and prolonged open time.

Figure 4 shows the viscosity curve of WEO with different OEO content with time. The viscosity of WEO increased steadily with time, which indicates that the emulsion is uniform and stable. Overall, the curing rate of WEO is negatively correlated with the content of OEO. With the increase of OEO content, the curing speed of the system is reduced and the operable time is prolonged. When the ratio of oxidation extraction oil and epoxy resin is 0:5, 2:5, 3:5, 4:5, and 5:5, the operating time is 68 min, 72 min, 80 min, 95 min, and 100 min, respectively. It can ensure certain construction operability. All in all, the epoxy resin system can be diluted by the addition of oxidized extraction oil. The viscosity and curing rate of emulsion decrease with the increase in the content of oxidized extraction oil.

#### 3.2.3. Mechanical Properties

Figure 5 shows the tensile strength and elongation at break of the cured WEOs and WER. The tensile strength of the WER is the highest, which is up to 5.06 MPa. However, the tensile strength decreases with an increasing mass ratio of OEO in WEO, maybe because OEO is filled with epoxy resin and curing agent as a dispersed phase. It reduces the curing crosslinking degree of the epoxy resin.

The elongation at the break of WEO first increases and then decreases with an increasing mass ratio of OEO in WEO. The elongation at break of the WEO-4 is the largest, reaching 91.5%, with a significant increase of 33.2%. It indicates that OEO plays a toughening effect in the WEO system. When the amount of oxidized extraction oil continues to increase, the elongation of WEO-5 decreases. This phenomenon can be explained by the results in Table 3 above; that is, when the amount of oxidized extracted oil increases to WEO-5, the compatibility between the oxidized extracted oil and waterborne epoxy resin system becomes poor. The non-uniformity of the material results in a decrease in elongation. Obviously, the content of OEO has the opposite effect on the tensile strength and elongation

at the break of the material. This is related to the crosslinking rate of epoxy resin and curing agent. If the epoxy resin and curing agent in the system have a high degree of crosslinking, the permanent deformation of brittle failure is small, which means lower elongation and higher strength. In short, the toughness of the epoxy resin is improved by the addition of oxidized extraction oil.

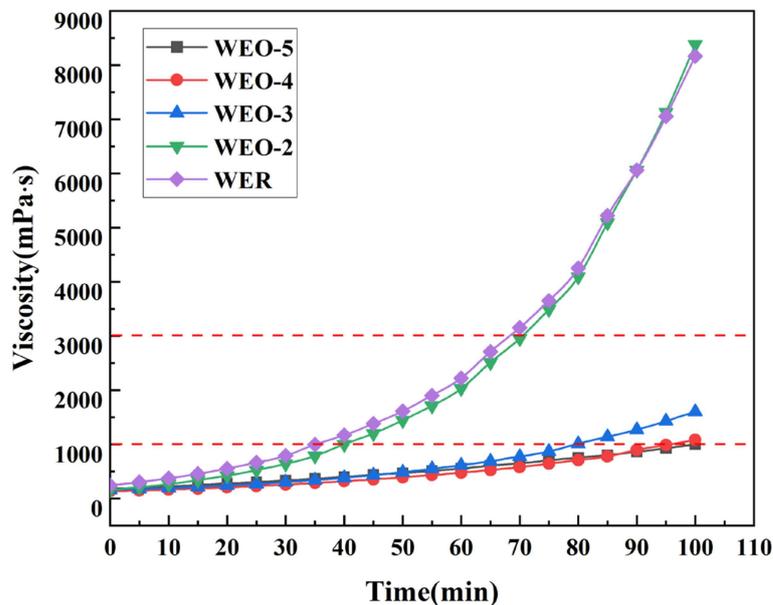


Figure 4. Viscosity of WER and WEO.

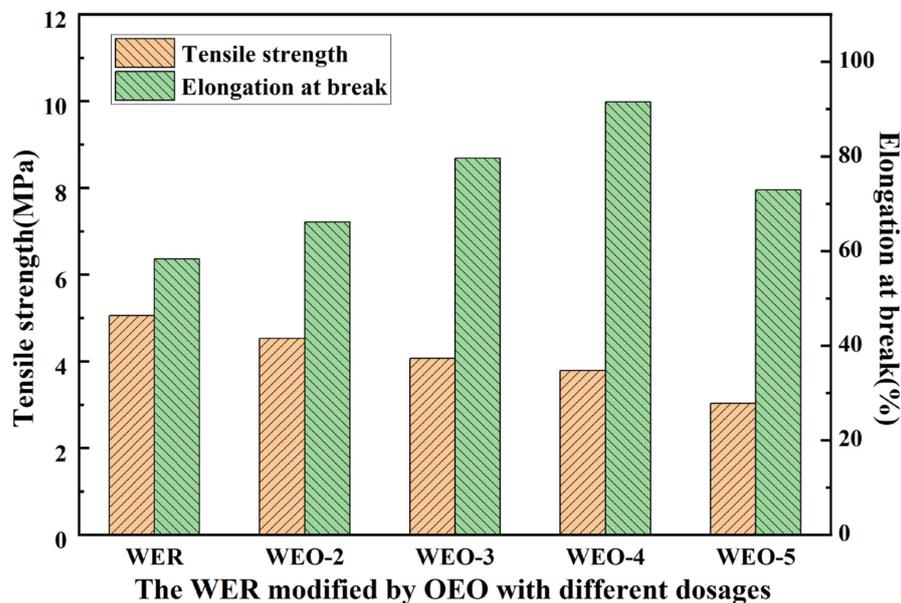
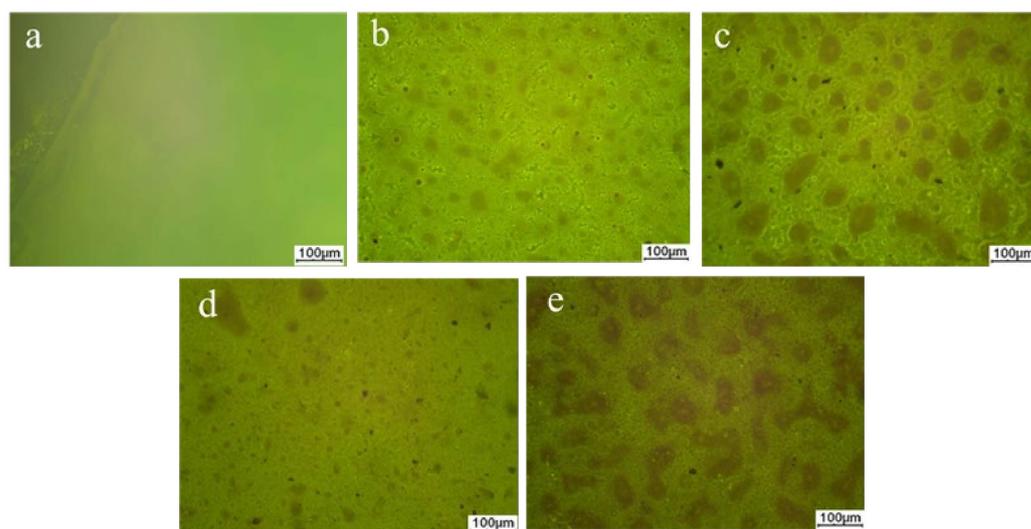


Figure 5. Mechanical properties of the cured WER and WEO.

### 3.2.4. Compatibility

Figure 6 shows the fluorescence micrograph of pure WER and WEO, wherein Figure 6a illustrates the image of the condition of the WER, which is yellow–green, smooth, and uniform. Figure 6b–e illustrate the image of the epoxy resin system blended with the extraction oil, wherein the yellow–green epoxy resin is the continuous phase; the black oxidized extraction oil is the dispersed phase. The oxidized extraction oil is evenly dispersed in the epoxy resin. With the increase of OEO content, the black phase gradually increases, the

color deepens, and the size of the phase domain becomes more immense. The average particle size of the black phase of WEO-4 is the smallest. It illustrates that the oxidized extraction oil has good dispersion and compatibility with epoxy resin. The oxidized extraction oil and epoxy resin are crosslinked into a stable three-dimensional network structure, which has excellent mechanical properties in macro. When OEO continues to increase, the phase domain size of OEO becomes more immense, mainly caused by the insufficient shear of the mixture.



**Figure 6.** Microstructure of extraction oil/waterborne epoxy resin system with different contents of oxidized extraction oil. ((a) WER, (b) WEO-2, (c) WEO-3, (d) WEO-4, (e) WEO-5).

### 3.3. Oxidized Extraction Oil/Waterborne Epoxy-Resin-Emulsified Asphalt System

#### 3.3.1. Viscosity

Viscosity is a vital index to characterize the curing speed and workability of the emulsion. Figure 7 shows the variation curve of viscosity of oxidized extraction oil/WEREA with time. It can be seen that all the viscosity of WEREA modified by oxidized extraction oil changed faster than that of WEO-4 because emulsified asphalt contains more asphaltene, whose viscosity is higher than that of oxidized extraction oil. The more oxidation extraction oil that is added, the smaller the viscosity of the WEREA. With the extension of curing time, the viscosity curve of WEO becomes mild and the curing speed of WEO becomes slow. This demonstrates that the addition of oxidized extraction oil can dilute the WEREA, reduce its viscosity and maintain certain processability and operability.

#### 3.3.2. Mechanical Properties

It can be seen from Figure 8 that with the increase of the content of oxidized extraction oil, the elongation at the break of the WEREA gradually increases while the tensile strength decreases. When the content of OEO is more than 5 parts, the elongation of WEREA is more than 100%. The results of the elongation show that the oxidized extraction oil can improve the dispersion of asphaltenes, thereby improving the toughness of the epoxy-emulsified asphalt.

The results of tests demonstrate that the tensile strength of the pure WEREA is the highest and up to 3.1 MPa. It is well known that the mechanical property of WEREA mainly depends on the content of the epoxy resin and its reaction crosslinking degree. As the content of extracted oil increases, the relative content of the epoxy resin in WEREA decreases gradually, so the tensile strength of WEREA modified by oxidized extraction oil naturally declines. On the other hand, the carboxyl group in the oxidized extraction oil interferes with the curing and crosslinking degree of the water-based epoxy resin, resulting in the weakening of the cohesion of the epoxy resin. The bonding force of WEREA

increases first and then decreases with increasing OEO content, which, because of the effective compatibility, gradually forms. This demonstrates that the appropriate amount of extraction oil can improve the adhesion of the material.

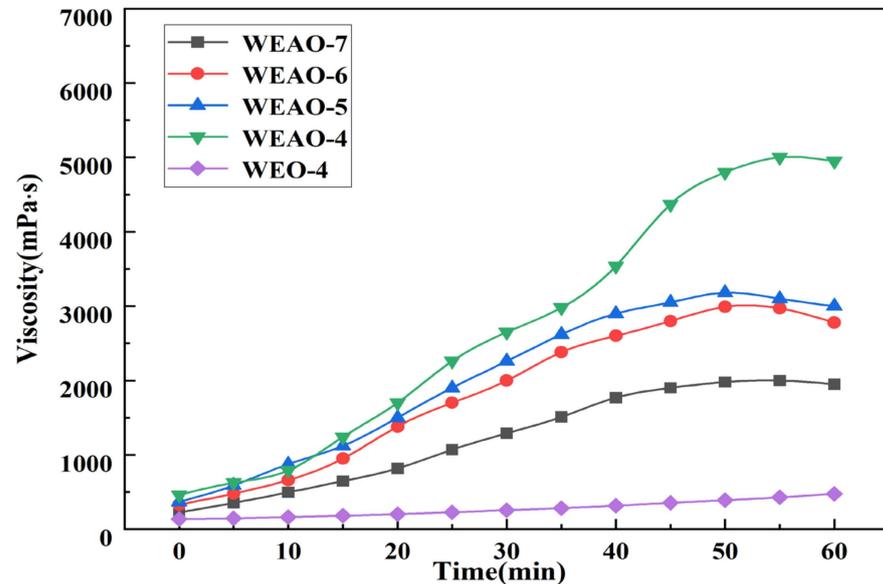


Figure 7. Viscosity of WEO and WERA modified by oxidized extraction oil.

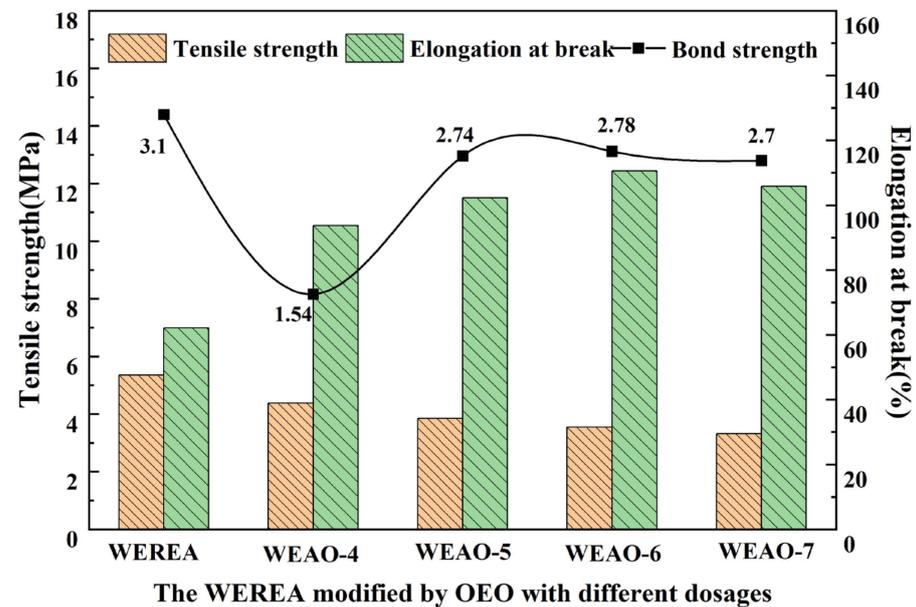


Figure 8. Mechanical properties of cured WEA and WEAOs.

According to the results of tensile properties and bond strength, the WEAO-6 shows high bond strength, breaking elongation, and tensile strength. To sum up, when the mix ratio of oxidized extraction oil and epoxy resin was 6:5, the breaking elongation of waterborne epoxy-resin-emulsified asphalt can be increased by 69%. The WEAO-6 is the ratio with the best comprehensive performance.

The previous study [3] showed that when the WER content was 25%, the tensile strength of WERA reached the maximum value of 1.75 MPa. During the test, it was found that when the content of waterborne epoxy resin increased to 30%, the stability of WERA became poor, which led to a decline in its performance. At the same time, with the increase of the content of WER, the elongation at the break of WERA decreased significantly. Compared with

the previously published work, oxidized-extraction-oil-modified WEREA has better tensile strength and elongation at break. Moreover, when the content of waterborne epoxy resin varied from 30% to 40%, the oxidized-extraction-oil-modified WEREA can still maintain a stable emulsion. With the increase in the content of WER, from WEAO-7 to WEAO-4, the elongation at break of WEREA only decreased by 11%.

### 3.3.3. Aging Performance

Table 4 shows the experimental results of tensile strength and elongation at break of extraction oil/epoxy-emulsified asphalt before and after UV aging. The tensile strength and elongation at break of WEREA decreased slightly under the aging conditions. It may be attributed to the fracture of long-chain alkyl side chains in asphalt molecules and partial oxidative dehydrogenation of aromatic hydrocarbons and colloids caused by aging, which has an adverse impact on the curing and crosslinking process of epoxy resin, resulting in the decrease of crosslinking density. With the addition of oxidized extraction oil, the loss rate of tensile strength and elongation at break of the WEREA before and after aging are gradually reduced. Oxidized extraction oil had an obvious effect on the elongation at the break of the WEREA after aging. The elongation at break loss of WEREA before and after aging is 13.7%, while the loss rate of elongation at break of the WEREA after adding oxidized extraction oil is about 5~7%. It indicates that the oxidized extraction oil can improve the aging performance of WEREA.

**Table 4.** Tensile strength and elongation of oxidized-extracted-oil-modified WEREA before and after thermal aging.

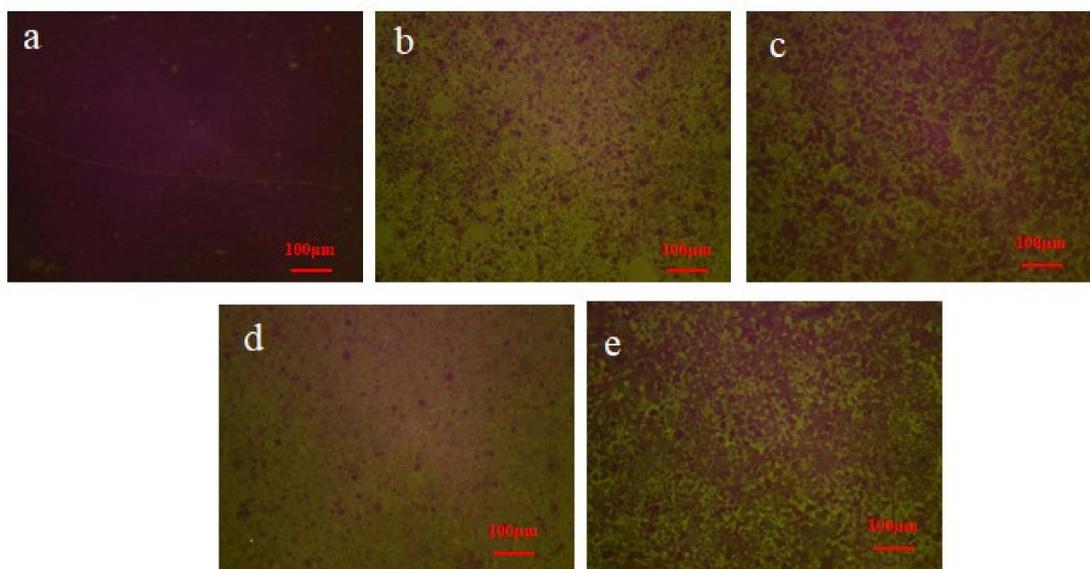
Samples	Tensile Strength (MPa)			Elongation at Break (%)		
	Before Aging	After Aging	Loss Ratio	Before Aging	After Aging	Loss Ratio
WEREA	5.36	4.91	8.4%	62.12	53.61	13.7%
WEAO-4	4.34	4.04	7.76%	93.72	86.94	7.23%
WEAO-5	3.85	3.57	7.27%	102.25	96.24	5.9%
WEAO-6	3.55	3.31	6.76%	110.6	105.1	4.97%
WEAO-7	3.32	3.12	6.02%	105.88	100.86	4.74%

### 3.3.4. Compatibility

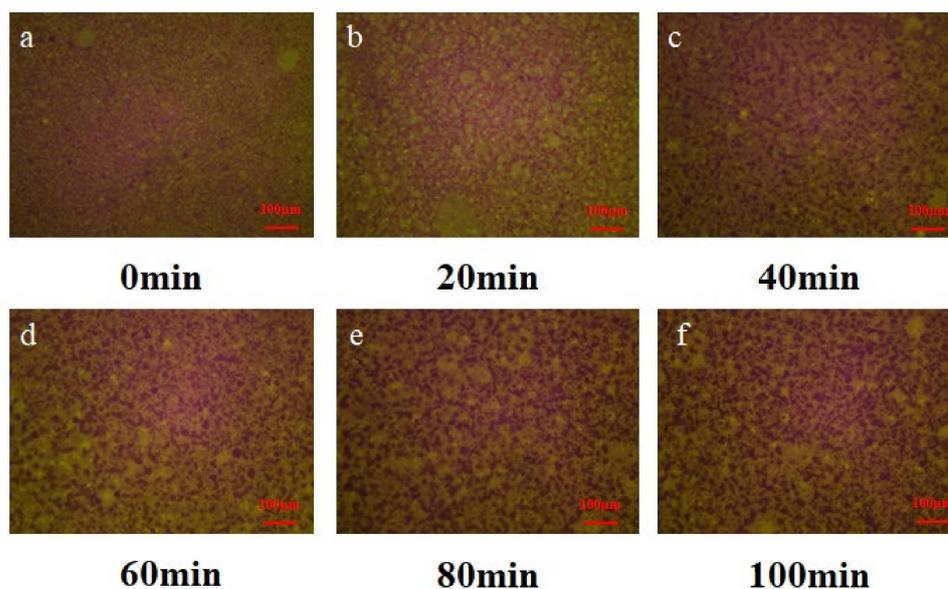
Figure 9 shows the fluorescence images of extraction oil/WEREA. There are no luminescent substances in asphalt, so it appears black (Figure 9a). With increasing oxidized extraction oil content, it can be clearly observed that the emulsified asphalt and oxidized extraction oil were evenly distributed in the epoxy resin (Figure 9b–e). The black phase of WEAO-6 has the smallest domain size and the most uniform mixing of the two phases (Figure 9d). As the content of oxidized extraction oil continues to increase, the compatibility between the emulsified asphalt and the epoxy resin becomes worse.

### 3.3.5. Microstructure of Extracted Oil/WEREA during Curing

Figure 10 shows the microstructure of the OEO modified WEREA system at different curing times. With the progress of the curing reaction, the size of asphalt particles (asphalt and oxidized extraction oil) gradually increases. After curing for 40 min, the particle size gradually increases from a homogeneous state to a phase separation state. Curing for more than 60 min, the particle size remains almost unchanged due to the chemical gel state with fixed microstructure.



**Figure 9.** Pictures of WERA fluorescence microscope with different OEO contents. ((a) Emulsified asphalt, (b) WEAO-4, (c) WEAO-5, (d) WEAO-6, (e) WEAO-7).



**Figure 10.** Microstructure of extraction oil/epoxy emulsified asphalt at different curing times. ((a) 0 min, (b) 20 min, (c) 40 min, (d) 60 min, (e) 80 min, (f) 100 min).

#### 4. Conclusions

To improve the compatibility between the epoxy resin and emulsion asphalt, the effects of oxidized extraction oil (OEO) as compatibilizer on the performance of waterborne epoxy and emulsified asphalt were evaluated through a series of experiments on viscosity, mechanical properties, and aging resistance. The conclusions obtained from the experimental results are as follows:

(1) After being oxidized by potassium permanganate, the chemical polarity of the extracted oil was improved so that the extracted oil became compatible with the epoxy resin. (2) The oxidized-extraction-oil-modified waterborne epoxy resin (WER) emulsion is homogeneous and stable. When the content of OEO in the WER is 21%, the elongation at break of the WER can reach up to a maximum of 91.5%, with a significant increase of 33.2%. It was indicated that OEO played a toughening effect in the WER system. (3) The OEO

can reduce the viscosity and curing speed of waterborne epoxy-resin-emulsified asphalt (WEREA) emulsion. The OEO was a good compatibilizer for WEREA. The results of mechanical properties showed that the OEO can greatly improve the toughness and aging resistance of WEREA. With the addition of OEO, the loss of tensile strength and elongation at the break of aged WEREA gradually reduced; especially when the content of OEO in the WEREA is 21%, the loss in elongation at the break of aged WEREA decreases to 4.7%. (4) The toughening mechanism of the OEO-modified WEREA depended on the improvement of compatibility between emulsified asphalt and epoxy resin. When the mix ratio of oxidized extraction oil and epoxy resin is 6:5, the breaking elongation of WEREA can be increased by 69%. Meanwhile, its tensile strength can still remain larger than 3 MPa, and it has good bonding strength.

## 5. Limitations

The current research only focused on the modification effect of oxidized extract oil on waterborne epoxy-emulsified asphalt. Although oxidized extract oil can significantly improve the toughness and anti-aging performance of waterborne epoxy-emulsified asphalt, it reduces the bonding performance of waterborne epoxy-emulsified asphalt. The optimized process of oxidation treatment of the extracted oil is recommended for future work. Moreover, the chemical reaction conditions between the carboxyl group of the oxidized extracted oil and the epoxy resin have to be further investigated so as to transform the physical modification of the oxidized extracted oil into chemical modification and improve the comprehensive performance of the waterborne epoxy-emulsified asphalt modified by the oxidized extracted oil. The application value of the oxidized extracted oil in the waterborne epoxy-resin-emulsified asphalt may also be systematically evaluated in further studies.

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