Evaluation of Properties and Mechanisms of Waste Plastic/Rubber-Modified Asphalt

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Abstract: Waste plastic, such as polyethylene (PE), and waste rubber tires, are pollutants that adversely affect the environment. Thus, the ways these materials are used are important in realizing the goals of reduced CO₂ emissions and carbon neutrality. This paper investigates the fundamental properties, compatibility, and interaction mechanism of waste plastic/rubber-modified asphalt (WPRMA). Dynamic shear rheology, fluorescence microscopy, a differential scanning calorimeter, and molecular dynamic simulation software were used to evaluate the properties and mechanisms of WPRMA. The results show that the anti-rutting temperature of WPRMA with different waste plastic contents is higher than 60 °C and the optimal dosage of waste PE in WPRMA is 8%, which can enhance the high-temperature properties and compatibility of rubber-modified asphalt. The temperature can directly promote the melting and decomposition of the functional groups in WPRMA and thus must be strictly controlled during the mix production process. The interaction mechanism suggests that waste plastic can form networks and package the rubber particles in rubber-modified asphalt. The main force between waste plastic and rubber is Van der Waals force, which rarely occurs in chemical reactions.

Keywords: rubber-modified asphalt; waste plastic; polyethylene; molecular simulation; interaction mechanism

1. Introduction

Waste plastic has caused great concern because it pollutes the soil, sea, and atmosphere, especially when it is in the form of micro-nano plastic [1]. Micro-nano plastic is emitted into the atmosphere and thus directly and negatively affects human health, leading to diseases such as pneumoconiosis and cancer [2]. The total quantity of emitted waste plastic is 8 million tons each year around the world [3]. Therefore, positive ways to reuse waste plastic have become a major research focus. For example, researchers have found that waste plastic can act as a modifier to improve the properties of asphalt, which is an effective way to reuse waste plastic [4].

The various types of waste plastic and their compositions are complex in terms of their properties and structure [5]. Polyethylene (PE) is the most common waste plastic and is versatile [6]. PE can be categorized as high-density PE, medium-density PE, and low-density PE, but only medium-density PE and low-density PE can be used in modified asphalt [7]. The different structures of PE provide different modification effects for asphalt. For example, medium-density PE-modified asphalt offers the best high-temperature properties and low-density PE-modified asphalt offers the best low-temperature properties [8]; that is, the inclusion of PE or waste PE into the mix can control the high-temperature or low-temperature properties of asphalt. Therefore, if waste PE is used to modify rubber-modified asphalt, then the properties of the asphalt and the interaction between the waste plastic and rubber-modified asphalt need to be investigated.
In recent years, many research efforts have focused on rubber-modified asphalt. For example, Jitsangiam et al. [9] investigated the effects of rubber latex on rubber-modified asphalt based on macro and micro-observations and found that the performance of rubber-modified asphalt depends not only on the rubber latex content but also on the various internal interactions between the rubber latex and asphalt binder. Wang et al. [10] evaluated the properties of rubber asphalt that had been modified by bio-oil and found that swine manure bio-oil can reduce the viscosity of the binder and improve its workability. In other work, Wang et al. [11] studied the fundamental properties and mechanisms of rubber asphalt that had been modified by tetraethyl orthosilicate and showed that the physical and rheological properties of the tetraethyl orthosilicate and rubber-modified asphalt could be improved due to the formation of a network structure between the silicon-oxygen bonds (Si–O–Si) and rubber powder. Cao et al. [12] investigated the flue gas composition of waste rubber-modified asphalt and found that the main hazardous flue gases are sulfur-containing organics, benzene homologues, and hydrogen sulfide (H$_2$S). Ren et al. [13] examined the effects of the swelling-degradation degree of crumb rubber on crumb rubber styrene-butadiene-styrene (CR/SBS)-modified porous asphalt binder and mixture. Ren et al.’s results showed that CR/SBS-modified bitumen could be used as a high-viscosity binder in porous asphalt. Yin et al. [14] studied the mechanical properties and reaction mechanism of microwave-activated crumb rubber-modified asphalt and found that the microwave activation of the crumb rubber improved the anti-aging properties of the asphalt. Gui et al. [15] evaluated the rheological characteristics of warm-mix crumb rubber-modified asphalt and found that the combination of crumb rubber and warm-mix crumb rubber-modified asphalt exhibits a positive synergistic effect on the asphalt’s rheological properties. The aforementioned studies suggest that adding rubber particles alone cannot achieve the excellent properties of rubber-modified asphalt, because rubber-modified asphalt has poor compatibility and low-temperature properties. Therefore, the key to the effective use of rubber-modified asphalt is to improve its low-temperature and compatibility properties.

Previous research has shown that low-density PE can improve the high-temperature rheological properties and low-temperature crack resistance of rubber-modified asphalt, as well as the compatibility between the rubber particles and asphalt binder [16]. Liang et al. [17] investigated the utilization of wax residue as a compatibilizer for rubber/recycled PE-modified asphalt and found that wax residue can enhance the storage stability of asphalt with a composite modifier and that the proper amount of wax residue improves the mixture’s low-temperature behavior. Yan et al. [18] found that the inclusion of low-density waste PE can enhance the high-temperature properties of rubber-modified asphalt but diminishes its low-temperature properties. Gibreil and Feng [19] found that high-density PE can improve rubber-modified asphalt’s resistance to moisture, while Alghrafy et al. [20] studied the rheological properties and aging performance of sulfur-extended asphalt modified with waste PE and found that the waste PE made the asphalt stiffer and thus better resistant to rutting/fatigue cracking. The aforementioned studies confirm that the inclusion of waste PE in the mix can improve the rheological properties and compatibility of rubber-modified asphalt. However, the precise effects of waste PE on the rheological properties and interaction mechanism of rubber-modified asphalt remain unclear.

In this study, the waste PE were added into rubber-modified asphalt to improve the properties of rubber-modified asphalt. The properties of waste plastic/rubber-modified asphalt (WPRMA) such as rheological property and storage stability are evaluated by using dynamic shear and softening point test. And also the interaction mechanism of WPRMA mechanisms are analyzed by using fluorescence microscopy, a differential scanning calorimeter (DSC), and molecular dynamic simulation software.

2. Materials and Methods

The materials used in the study, as well as the laboratory tests conducted, are discussed in this section. This includes the raw materials, test methods, and preparation of WPRMA.
2.1. Raw Materials

The following raw materials were purchased for this study: waste PE from Taiyuan, Shanxi Province, China, rubber powder (80 mesh) from Changzhou Rongao Chemical New Material Co., Ltd., Changzhou, China. Solubilizer was bought from Sinopharm Chemical Co., Ltd., Shanghai, China, and the base asphalt SK-70 from Panjin Northern Asphalt Co., LTD, Panjin, China. The fundamental properties of the base asphalt are the softening point of 46 °C, penetration of 62 dmm, and ductility (15 °C) of 108 cm.

2.2. WPRMA Preparation

The WPRMA was prepared as follows. Firstly, the base asphalt SK-70 was heated to 135 °C in a stainless-steel asphalt binder extractor. Then, the rubber powder was added into the base asphalt slowly and stirred at 600 rpm for 1 h. Next, the mix was heated to 175 °C and put into the waste PE. The waste PE was then sheared for 1.5 h at a high-speed 5000 rpm shear for 1 h until the waste PE was completely dissolved in the asphalt binder. The waste rubber powder content was 20% and the dosages of the waste PE were 0%, 2%, 4%, 6%, 8%, and 10%, respectively.

2.3. Laboratory Experimentation and Testing

The fundamental properties of the WPRMA were tested as follows. The penetration of the WPRMA was measured at 25 °C according to ASTM D5/D5M-20. The softening point and ductility of the WPRMA were tested according to ASTM D36/D36M-20 and ASTM D113, respectively. The dynamic viscosity of the WPRMA was measured at 135 °C using a Brookfield viscometer in accordance with ASTM D4402. The rheological properties of the WPRMA were tested using an Anton Paar dynamic shear rheometer in accordance with ASTM D7175-15. The high-temperature rheological properties were tested using a 25-mm diameter parallel plate with a gap of 1 mm, and the low-temperature rheological properties were tested using an 8 mm diameter parallel plate with a gap of 2 mm. The tests were performed under controlled strain conditions. Temperature sweeps were applied with a fixed frequency of 10 rad/s, and the temperature ranged from 10 to 30 °C and 30 to 80 °C. The storage stability of the WPRMA was measured using a NETZSCH differential scanning calorimeter (DSC). The test temperature ranged from 50 to 240 °C and the heating rate was 10 °C/min. The softening point difference also was used to evaluate the storage stability of the WPRMA. The micro-morphology of the WPRMA was measured using an optical microscope (Shangguang, XSP-63, Shanghai, China) at 100× magnification.

2.4. Molecular Dynamic Modeling

For the molecular simulations of the WPRMA, PE (C_1600H_3208) was used as the waste plastic and butyl rubber (C_9H_16) represented the rubber powder. The asphalt was selected using the four components model which had been used in previous research [21]. Molecular models of WPRMA were built using layered modules. Figure 1 presents the molecular models of the WPRMA with 0%, 2%, 4%, 6%, 8%, and 10% waste PE.
Figure 1. Molecular models of waste plastic/rubber-modified asphalt at various percentages of waste polyethylene: (a) 0% waste PE; (b) 2% waste PE; (c) 4% waste PE; (d) 6% waste PE; (e) 8% waste PE; (f) 10% waste PE.
During the molecular simulation process, the molecular models of the WPRMA were first treated using geometry optimization. Then, the cohesive energy density formula was used to calculate the cohesive energy density between the waste PE and rubber-modified asphalt and to analyze the compatibility of the WPRMA. Molecular dynamic simulations were performed to evaluate the diffusion coefficients, interaction force, and structural changes of the WPRMA using the Forcite module with NVT and NPT ensembles. The total simulation time was 100 ps and the time step was 1 fs.

3. Results and Discussion

3.1. Assessment of Reliability of Molecular Simulation Results

The energy and density results were used to assess the reliability of the molecular simulations. Figure 2a shows that the energy of the WPRMA decreases with an increase in the simulated time and that the energy fluctuation of the WPRMA trends to zero when the simulated time is five steps. The results show that the total time of 100 steps is acceptable because only the energy fluctuation trends to zero, which indicates that the system is stable and the results for this time interval are reasonable. Figure 2b shows that the density of the WPRMA decreases, then increases sharply, then decreases slightly, and finally trends to a constant (1.02 g/cm³). Moreover, the constant density values are equal to the real density values of asphalt, which suggests that the molecular models can be used to simulate the properties of WPRMA.

![Figure 2](image)

**Figure 2.** Reliability assessment of energy and density results obtained from molecular simulations: (a) energy curves, and, (b) density curves.

3.2. Assessment of WPRMA Properties via Molecular Simulations

Molecular simulations also can be used to evaluate the properties of WPRMA, i.e., the diffusion coefficient, viscosity, glass transition temperature, free volume, and cohesive energy density, as discussed in the following paragraphs.

The diffusion coefficient can be used to analyze the movement of molecules and evaluate the self-healing properties of the mix. For the molecular simulations, the diffusion coefficient can be calculated by Equation (1).

\[
D = \frac{1}{6T} \left( r(t) - r(0) \right)^2
\]

where \( D \) is the diffusion coefficient, \( T \) is the total time of the molecular movement, \( r(t) \) is the displacement at time \( t \), and \( r(0) \) is the displacement at time 0.
Figure 3 shows that the diffusion coefficient of WPRMA decreases with an increase in the dosage of the waste PE and that the rate of decrease of the diffusion coefficient slows when the waste PE dosage is above 8%. The reason for this outcome is that, owing to the increase in its dosage, the waste PE curls up and becomes entwined with the rubber-modified asphalt, which leads to the slow molecular movement of the waste PE. Thus, these results show that waste PE can inhibit the movement of WPRMA and diminish the self-healing properties of WPRMA.

\[ \eta = \frac{\rho R T R_g^2}{6 M D} \]

where \( \eta \) is viscosity, \( \rho \) is density, \( R \) is constant (8.314), \( T \) is the total time, \( R_g^2 \) is average radius of gyration, \( M \) is the molecular mass, and \( D \) is the diffusion coefficient.

Figure 4 shows that the simulated viscosity of WPRMA increases with an increase in the dosage of waste PE. In this research, it selected the optimal dosage of waste PE as 8% because, as shown in the figure, the viscosity of the WPRMA increases slowly when the dosage is above 8%. These results indicate that waste PE can increase the viscosity and enhance the adhesive properties of WPRMA. Previous research found that viscosity is related to the high-temperature properties of asphalt, whereby the greater the viscosity, the better the high-temperature properties of the asphalt [22]. Similarly, it can be concluded that waste PE can promote improvement in the high-temperature properties of WPRMA. Previous research also found that the viscosity of rubber-modified asphalt ranges from 300 to 400 cp. Compared with these earlier results, it was found that the simulated viscosity of rubber-modified asphalt is 320 cp, which indicates that the simulated results are reliable.

Our previous research found that density-temperature curves can be used to indicate the real glass transition temperature via molecular simulations [23]. Similarly, in this study, it can be obtained the glass transition temperature (\( T_g \)) from density-temperature curves using molecular simulations. Figure 5 shows that the glass transition temperature of WPRMA increases with an increase in the dosage of waste PE, while the glass transition temperature of WPRMA is lower than that of rubber-modified asphalt. This finding indicates that waste PE can increase the glass transition temperature of WPRMA and improve its low-temperature properties. Previous research also found that low-density PE could improve the low-temperature properties of asphalt [24]. This finding verifies the reliability of the simulation results for the glass transition temperature of WPRMA.
Our previous research found that density-temperature curves can be used to indicate the real glass transition temperature via molecular simulations [23]. Similarly, in this study, it can be obtained the glass transition temperature ($T_g$) from density-temperature curves using molecular simulations. Figure 5 shows that the glass transition temperature of WPRMA increases with an increase in the dosage of waste PE, while the glass transition temperature of WPRMA is lower than that of rubber-modified asphalt. This finding indicates that waste PE can increase the glass transition temperature of WPRMA and improve its low-temperature properties. Previous research also found that low-density PE could improve the low-temperature properties of asphalt [24]. This finding verifies the reliability of the simulation results for the glass transition temperature of WPRMA.

Free volume can be used to evaluate the mobility properties of WPRMA. Figure 6 presents the free volume results of WPRMA with different dosages of waste PE. As shown, the Van der Waal forces with free volume percentages of 0%, 2%, 4%, 6%, 8%, and 10% WPRMA are 158,490.24 Å$^3$, 171,425.57 Å$^3$, 197,199.04 Å$^3$, 220,463.42 Å$^3$, 247,293.81 Å$^3$, and 270,809.26 Å$^3$, respectively. The free volume of the WPRMA is shown to increase with the increase in the waste PE dosage, which indicates that waste PE can increase the free volume of WPRMA and promote the mobility properties of WPRMA.
the increase in the waste PE dosage, which indicates that waste PE can increase the free volume of WPRMA and promote the mobility properties of WPRMA.

Figure 6. Free volume of waste plastic/rubber-modified asphalt with different percentages of waste polyethylene: (a) 0% waste PE; (b) 2% waste PE; (c) 4% waste PE; (d) 6% waste PE; (e) 8% waste PE; (f) 10% waste PE.
Cohesive energy density can be used to evaluate the interaction force of molecules and to calculate the solubility parameter of the molecules. The solubility parameter of WPRMA can be calculated by Equation (3).

\[
\delta = k\sqrt{CED}
\]  

where \(\delta\) is the solubility parameter, \(k\) is the constant number, and \(CED\) is the cohesive energy density.

Figure 7a shows that the cohesive energy density values for 0%, 2%, 4%, 6%, 8%, and 10% WPRMA are \(2.09 \times 10^7 \text{ J/m}^3\), \(3.12 \times 10^7 \text{ J/m}^3\), \(3.43 \times 10^7 \text{ J/m}^3\), \(3.45 \times 10^7 \text{ J/m}^3\), \(3.48 \times 10^7 \text{ J/m}^3\), and \(4.84 \times 10^7 \text{ J/m}^3\), respectively. The cohesive energy density of WPRMA first increases, then decreases with an increase in the waste PE dosage, which indicates that waste PE can promote an increase in the interaction force of WPRMA when the dosage is less than 8%. However, waste PE will diminish the interaction force of WPRMA when its dosage is 10%. This outcome indicates that the optimal dosage of waste PE is 8%, which provides the best cohesive energy density. Figure 7b shows that the solubility parameter of WPRMA follows a similar law to that of cohesive energy density. The solubility parameter of WPRMA first increases and then decreases with an increase in the waste PE. These results suggest that waste PE can promote the dissolution of WPRMA to a certain extent. If the waste PE dosage is above 8%, then the waste PE will inhibit the dissolution of the WPRMA.

![Figure 7a](image1.png)

**Figure 7.** (a) Cohesive energy density and (b) solubility parameter of waste plastic/rubber-modified asphalt at various dosages of waste polyethylene.

### 3.3. Assessment of WPRMA Rheological Properties and Storage Stability via Tests

Figure 8a presents the high-temperature scan curves that show a decrease in the complex modulus value of the WPRMA with an increase in temperature and an increase in the complex modulus value of the WPRMA with an increase in the waste PE dosage. The reason for this outcome is that the fill in waste PE can enhance the complex modulus of WPRMA. These results show that waste PE can enhance the high-temperature properties of WPRMA, but high temperatures would diminish the viscoelastic properties of WPRMA. The anti-rutting temperatures at 0%, 2%, 4%, 6%, 8%, and 10% WPRMA are 62 °C, 64 °C, 65 °C, 67 °C, 68 °C, and 69 °C, respectively. These temperatures clearly are higher than 60 °C. Figure 8b presents the low-temperature scan curves that indicate that the complex modulus values of the WPRMA decrease with an increase in temperature and increase with an increase in waste PE. The reason for this outcome is that the PE in waste PE plays an important role by enhancing the viscoelastic properties of WPRMA. These results show that the PE in waste PE can enhance the low-temperature viscoelastic properties and crack resistance of WPRMA.
The softening point difference can be used to evaluate the storage stability of WPRMA. Figure 9 shows that the difference in softening point of the rubber-modified asphalt is greater than 2 °C, which indicates that the storage stability of the rubber-modified asphalt is poor. The softening point difference of WPRMA is less than that of rubber-modified asphalt, that is, lower than 2 °C, except for when the asphalt contains 10% waste PE. The softening point difference of WPRMA increases with an increase in the waste PE dosage. These results suggest that waste PE can improve the storage stability of WPRMA and promote the solubility of rubber powder in asphalt. However, once the waste PE dosage is above 8%, the inclusion of waste PE will diminish the storage stability of WPRMA.

![Figure 8](image1.png)

**Figure 8.** Temperature scan curves of waste plastic/rubber-modified asphalt at: (a) high temperatures and (b) low temperatures.

![Figure 9](image2.png)

**Figure 9.** Softening point difference of waste plastic/rubber-modified asphalt at various dosages of waste polyethylene (PE).

### 3.4. Interaction Mechanism of WPRMA

Figure 10 presents the DSC curves of the WPRMA and the endothermic melting peaks when the temperature reaches about 90 °C. After the heating process, the melting curve is basically stable, which indicates that most of the functional groups of waste PE undergo a phase transformation at about 90 °C and that this kind of material absorbs heat easily and then melts. The endothermic melting peaks of the rubber-modified asphalt appear at 120 °C and the endothermic melting peaks of 6% WPRMA appear at 160 °C, which indicates that the waste PE degraded first, then the asphalt, and finally, the rubber powder. Therefore,
we recommend a temperature range from 100 °C to 160 °C for WPRMA. WPRMA has different melting peaks, which suggests that the waste PE dosage can significantly affect the degradation of WPRMA and that a synergistic effect is present between waste PE and rubber powder.

![Differential scanning calorimeter curves of waste plastic/rubber-modified asphalt.](image)

Figure 10. Differential scanning calorimeter curves of waste plastic/rubber-modified asphalt.

Figure 11 presents fluorescence microtopographic images of WPRMA that show the significant change in micro-morphology and increase in the movement phase with an increase in the waste PE. These results show that waste PE can improve the micro-morphology and flow of WPRMA. The micro-morphology of 2% or 4% WPRMA still shows an obvious granular shape, indicating that the waste PE dosage is too low and cannot completely melt the compound additive. The movement phase in 6% WPRMA indicates that waste PE and rubber powder can melt completely. Networks appear when the waste PE dosage is 8%, suggesting that 8% WPRMA offers the best compatibility, whereas the separation phase of 10% WPRMA indicates that 10% waste PE can diminish the compatibility of WPRMA.

The micro-mechanisms of WPRMA suggest the presence of (1) a granular shape when the waste PE dosage is low (2–4%), (2) a movement phase when the waste PE dosage is 6%, (3) networks when the waste PE dosage is 8%, and (4) a separation phase when the waste PE dosage is 10%. In short, this micro-morphology change law indicates a granular shape, movement phase, networks, and separation phase. Temperature is the factor that directly promotes the melting and decomposition of the functional groups in WPRMA, and thus, the temperature must be strictly controlled during the mixture production process.

![Fluorescence microtopographic images of WPRMA](image)

Figure 11. Cont.
Figure 11. The micro-morphology of waste plastic/rubber-modified asphalt at various dosages of waste polyethylene: (a) 0% waste PE; (b) 2% waste PE; (c) 4% waste PE; (d) 6% waste PE; (e) 8% waste PE; (f) 10% waste PE.

4. Conclusions

In this study, the properties and mechanisms of WPRMA were evaluated using dynamic shear rheology, fluorescence microscopy, a DSC, and molecular dynamic simulation software. The following conclusions can be drawn from the results.

- The inclusion of waste PE in the mix can inhibit the movement of WPRMA and diminish its self-healing properties. However, the inclusion of waste PE can increase the viscosity and enhance the adhesive properties of WPRMA. Furthermore, waste PE can enhance the high-temperature properties of WPRMA, but high temperatures can diminish the viscoelastic properties of WPRMA due to the presence of fill in waste PE.
- The inclusion of waste PE in the mix can increase the free volume of WPRMA and promote its mobility properties. During the degradation process of WPRMA, first the waste PE degrades, then the asphalt, and finally the rubber powder. The dosage of the waste PE can significantly affect the degradation of WPRMA because waste PE and rubber powder are synergetic.
- The micro-morphology of WPRMA in terms of change regularly indicates a granular shape, movement phase, networks, and a separation phase. Temperature is the factor that directly promotes the melting and decomposition of the functional groups in WPRMA. Therefore, temperature must be strictly controlled during the mixture production process.

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