

# Functionalization of reclaimed polyethylene with maleic anhydride and its application in improving the high temperature stability of asphalt mixtures

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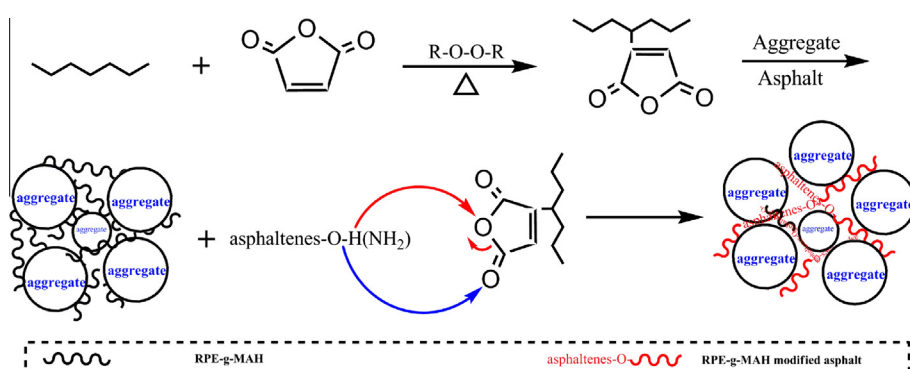
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## HIGHLIGHTS

- RPE-g-MAH was successfully prepared by a melt reactive process.
- DCP and MAH contents played the key role in DS and RD.
- High temperature performance of RPE-g-MAH modified asphalt mixture were investigated.
- RPE-g-MAH had superior rutting resistance towards the asphalt mixture.

## GRAPHICAL ABSTRACT



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## ABSTRACT

A process to improve the properties of reclaimed polyethylene (RPE) polymer modified asphalt mixture by grafting of maleic anhydride (MAH) onto RPE (RPE-g-MAH) in the presence of dicumyl peroxide (DCP) as initiator was proposed. Fourier transform infrared spectra (FTIR) was employed to verify the grafting of MAH onto RPE. Modified asphalt mixture under different conditions were investigated on the RPE-g-MAH samples. To assess the effects of the RPE-g-MAH on the high-temperature stability of modified asphalt mixture, we discussed dynamic stability (DS) and rutting depth (RD) of modified asphalt mixture by studying the influences of various experimental parameters such as MAH content, DCP content, mixing temperature, wet mixing times and gradation type of mixture. Experimental results indicated that the RPE-g-MAH is feasible to improve high-temperature stability performance and low-temperature performance of asphalt mixture, and it could dramatically improve resistance to rutting of the asphalt mixture. Possible mechanism was proposed for RPE-g-MAH modification of asphalt mixture.

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## 1. Introduction

Asphalt mixtures have been exposed to greater stresses due to the increase of traffic volumes, truck traffic and higher tire

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pressures. One of the most common forms of distress of asphalt pavements is rutting. Rutting is the defined as the progressive accumulation of permanent deformation of each layer of the pavement structure under repetitive loading [1–4]. Different additives materials such as polymers, fibers, rubber powder, and sulfur have been employed to improve the rutting resistance of asphalt mixtures [5–10]. Polymer materials were found to be effective with

beneficial effects on the anti-rutting behavior of asphalt mixtures. They have been used to modify asphalt cement and to improve the properties of asphalt mixtures as an asphalt modifier in order to improve the performance and extend the lifetime of asphalt pavement mixtures [11–14]. Recently, various researchers have shown that the addition of polymers decreases the pavement rate of deterioration and enhances its performance [15–18]. Tayfur et al. [19] studied the rutting performance of conventional and five modified asphalt mixtures containing polymer modifiers, the results show that additives performed more resistance to permanent deformation according to the conventional mixtures. Khattak et al. [20] reported the horizontal plastic deformation rates of polymer modified mixtures were lower than the neat ones and were related to the lap-shear strength and toughness energy. Othman [21] reported a significant increase in both indirect tensile and compressive strength of the LDPE-modified asphalt concrete mixtures.

The functionalized polyethylene via grafting reaction with polar unsaturated monomers in solution [22–24], melt [25–27], solid state [28] and radiation [29] have attracted much attention due to its specifically application [30–33]. Among these unsaturated monomers, MAH is widely used. In order to improve its performance, many efforts have been made to graft MAH onto PE. However, little information has been reported on the effects of grafting polymerization on the properties of modified asphalt mixtures. Thus, further studies are still necessary to establish.

Here, we reported a melt reactive process to improve the properties of RPE polymer modified asphalt mixture by grafting of MAH onto RPE in the presence of DCP as initiator. To assess the effects of the RPE-g-MAH on the high-temperature stability of modified asphalt mixture, we discussed DS and RD of modified asphalt mixture by studying the influences of various experimental parameters such as MAH content, DCP content, mixing temperature, wet mixing times and gradation type of mixture. Experimental results indicated that the RPE-g-MAH is feasible to improve high-temperature stability performance of asphalt mixture and it could dramatically improve resistance to rutting of the asphalt mixture. The pavement performance of the modified mixtures were improved with the GD of the RPE-g-MAH increasing.

## 2. Experimental section

### 2.1. Materials

Asphalt was provided by China National Petroleum Corporation. Softening point temperature was 45.3 °C, penetration at 25 °C was 82.1 (0.1 mm).

RPE was produced by Longda plastic Co. All the other chemicals including dicumyl peroxide (DCP, the initiator), MAH acetone, xylene, and ethanol were reagent-grade products and used without further purification.

### 2.2. Preparation of RPE-g-MAH

The melt-grafting reactions were carried out in a twin-screw extruder ( $\Phi = 30$  mm;  $L/D = 28$ ). RPE pellets, monomer(s) (0–3 wt%), and initiator (0–2.0 wt %) were premixing before being charged into the extruder. The temperature of the cylinder was set at 160–195 °C. The screw speed was fixed at 200–400 r/min.

In a typical preparation procedure, 1000 g of RPE pellets, 7.5 g of MAH and 5 g of DCP were mixed at room temperature in a high speed mixing machine under vigorously stirring for 5 min. After that, the mixed materials were transferred into the twin-screw extruder with the temperature of the cylinder at 170–175 °C. The screw speed was fixed at 250 r/min. The strips from the extruder were cut into granules about 2–4 mm long after cooling in a water bath, thus RPE-g-MAH were obtained.

### 2.3. Characterization of RPE-g-MAH

The RPE-g-MAH samples were dissolved in refluxing xylene at a concentration of 1% (wt/vol.), and excess acetone was then added to precipitate them. By this procedure the copolymer of MAH was separated, and the actual graft degree could be obtained. The precipitated samples were filtered, washed and dried under vacuum at 80 °C for 24 h. The purified products were then hot-pressed into a piece of thin films for fourier transform infrared ray (FT-IR) characterization.

### 2.4. Wheel tracking

In a typical test, when mineral aggregates are heated up to 185 °C, the amount of RPE-g-MAH with 0.3% (weight ratio of RPE-g-MAH to aggregate) is directly entered into the heated mineral in an asphalt mixer at a mixing temperature of 180 °C and mixed continuously for 40 s till all RPE-g-MAH is completely dispersed in mineral aggregates. Then, 4.5% of asphalt (heated up to 160 °C) by optimum asphalt-aggregate ratio was added to the mixtures and stirred for 90 s. Finally the filler was added into the mixtures, and stirring continued for another 90 s. After the mixing completed, the mixtures were prepared in a  $300 \times 300 \times 50$  mm steel mould (three specimens per parallel control test). Slab of the compactor may pass on 24 times to and fro and 9 kN load pressures during compacting. And all wheel tracking tests were conducted at 60 °C (0.5 °C accuracy) and 0.7 MPa (0.05 MPa accuracy) to measure DS and RD.

## 3. Results and discussion

### 3.1. FTIR spectroscopy

IR spectra of RPE and RPE-g-MAH are shown in Fig. 1. Compared with the spectrum of RPE, the spectrum of RPE-g-MAH had some new peaks (1868, 1790, and 1715  $\text{cm}^{-1}$ ) which are the characteristic absorption peaks of the grafted MAH since the MAH monomer had been removed from the samples in the purification process. The two characteristic absorption peaks at 1868 and 1790  $\text{cm}^{-1}$  are due to symmetrical and asymmetrical stretching of carbonyl ( $\text{C}=\text{O}$ ), respectively. The bands at 1715  $\text{cm}^{-1}$  belong to the carbonyl groups ( $\text{C}=\text{O}$ ) of MAH [34].

### 3.2. Effect of temperature

In the process of grafting by melt extrusion, materials are in the semi-closed state in the process of melt reaction in the twin-screw extruder, which effectively reduces losses of MAH caused by sublimation. Extrusion temperature has immediate impact on decomposition rate of initiator, meanwhile, it also has obvious impact on materials plasticizing and melt viscosity. When the reaction temperature is relatively low, the decomposition rate of initiator DCP is low in the whole process of grafting, and the melt viscosity of reaction mass is comparatively high, which results in insufficiency of grafting reaction. On the contrary, when the reaction temperature is high, the increase of decomposition rate of DCP can easily causes consumption of initiator ahead of time, and the concentration of generated free radical is also relatively high, it's also beneficial to the occurrence of side reaction.

The melt index and grafting ratio data of RPE blend under different reaction temperature is shown in Table 1. Table 1 shows that when the temperature is at 155–195 °C, there is relatively big difference in melt index of grafting blend, but no big change in

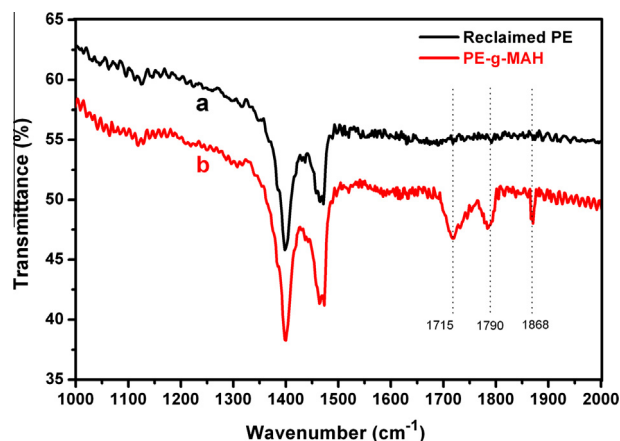


Fig. 1. FTIR spectra of RPE and RPE-g-MAH copolymers: (a) RPE and (b) RPE-g-MAH.

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