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Investigation the self-healing mechanism of aged bitumen using microcapsules containing rejuvenator

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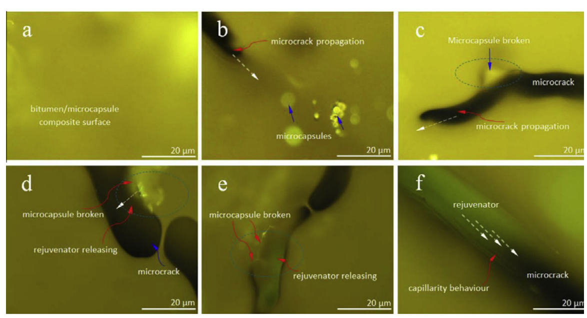
HIGHLIGHTS

- The microcapsules containing rejuvenator can be broken by microcracks.
- With the help of capillarity, the rejuvenator can fill the microcracks.
- With the help of the penetration of rejuvenator, the aged bitumen has a trance to recovery its virgin properties.

GRAPHICAL ABSTRACT

The capillarity behaviors of rejuvenator in self-healing bitumen by microcapsules, (a–c) a microcrack was generated by liquid N_2 with the width about 10–15 μm, the microcrack propagates and pierce the shells of microcapsules, (d and e) the liquid of rejuvenator leaked out from microcapsules and flowed into the microcapsules, and (f) movement trace and direction of rejuvenator during the capillarity.



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ABSTRACT

The aim of this paper is to investigate the self-healing mechanism of aged bitumen using microcapsules containing rejuvenator. Various microcapsule samples were successfully fabricated and the mean size and shell thickness were adjusted. It was found that the shell thickness was not greatly affected by the core/shell ratios and the emulsion stirring rates. These microcapsules can survive in melting bitumen with a good thermal stability. Based on these data, a typical microcapsule sample, fabricated by 3000 r min^{-1} with core/shell ratio of 2/1, was selected to investigate the self-healing mechanism. The whole self-healing process was observed including the crack generation, capillarity and the diffusion behaviors. The results showed that microcapsules broke by microcracks and leaked the oily–liquid rejuvenator into microcracks. With the help of capillarity, the rejuvenator filled the cracks with a movement speed mainly determined by the volume of microcapsules in bitumen. A diffusion phenomenon was also observed by using a fluorescence microscope. Properties of virgin and rejuvenated bitumen measurements confirmed that the aged bitumen can be partly recovered by the microcapsules containing rejuvenator.

1. Introduction

Every year about 95% of the almost 100 million tonnes of bitumen is applied in the paving industry where they essentially act

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as a binder for mineral aggregates to form asphalt [1]. One important issue need to be considered is the ageing of bitumen from climate and traffic in service life [2]. After years of usage, the stiffness of asphalt concrete increases while its relaxation capacity decreases, the binder becomes more brittle causing development of microcracks and ultimately cracking of the interface between aggregates and binder occurs [3]. The ageing problem of bitumen leads to pavement failure including surface raveling and reflective cracking. It will increase the expense of maintaining bituminous pavements [4]. Although part of aged asphalt can be recycled as an economic and environmentally sound solution, it is still a big problem to deal with the hazardous material of aged asphalt in large quantities. Therefore, preservation and renovation bitumen of pavement is a low-cost and effective approach.

An increase in the application of a higher percentage of the preservation and renovation of asphalt pavement is achievable using a rejuvenator. It is the only one method that can restore the original properties of the pavements [5]. The most important goal of utilizing rejuvenator products is to restore the asphaltenes/maltenes ratio. Rejuvenating agents have the capability of reconstituting the binder's chemical composition and consist of lubricating and extender oils containing a high proportion of maltene constituents [6]. However, a rejuvenator is not successfully applied because it is hard to penetrate the pavement surface. Shen et al. [7] reported the results of using three rejuvenators. It was found that none could penetrate into the asphalt concrete more than 2 cm. When applying these materials, the road must be closed for some time after their application. The rejuvenator, at the same time, will cause a high reduction of surface friction of pavement for vehicles. Moreover, an important aspect of these rejuvenators is that they may be dangerous to the environment. Encapsulation rejuvenator inside-usage in asphalt may be an alternative approach. García et al. [8] reported a method to prepare capsules containing rejuvenator by using epoxy resin as coating and pours sand as skeleton. We reported a method to fabricate microcapsules containing rejuvenator utilizing methanol modified melamine–formaldehyde (MMF) resin as shell material [9]. These microcapsules had satisfactory thermal stability in bitumen and reliable mechanical properties resisting the mixing process and temperature changes [10]. It had been proved that this product was environmental-friendly powder encapsulating suitable size rejuvenator for chemical engineering and construction engineering [11].

Self-healing materials based on microcapsules have been studied widely which have structurally incorporated ability to repair damage caused by mechanical usage over time. Monomer is encapsulated and embedded within the matrix materials. When the crack gets to the microcapsule, the capsule breaks and the monomer bleeds into the crack, where it can polymerize and mend the crack [12]. Besides the similar structure of microcapsules self-healing materials for bitumen/microcapsules materials, we still need to consider their special self-healing mechanism. Microcapsules in bitumen can be broken by microcracks, then the released rejuvenator seals the microcracks and permeate surrounding bitumen. The capillaries and penetration behavior will determine the self-healing efficiency of aged bitumen [13]. With the help of capillarity, rejuvenator flows into narrow microcracks without the assistance of, and in opposition to, external forces. Firstly, the core–shell structure of microcapsules is taken into account for the specific requirement such as size distribution, encapsulation ratios and non-biodegradable property, since this influences their service performance. As bitumen acts as thin layers between aggregates which are usually less than 50 μm, size of microcapsules containing rejuvenators should be smaller than 50 μm to avoid being squeezed or pulverized during asphalt forming. Secondly, the shell thickness must be controlled to make sure that the microcapsules have excellent thermal stability. It was reported have found that thicker shell will enhance the mechanical properties of microcapsules, the microcracks may not able to break these microcapsules [14]. It must be prevented that the microcracks propagation will go round the shells.

In view of the above, the aim of this paper is to investigate the self-healing mechanism of aged bitumen using microcapsules containing rejuvenator. Various microcapsule samples were fabricated to optimize the core–shell structures. In order to observe the self-healing behaviors, liquid nitrogen was used to generate microcracks in aged bitumen. The effects of mean size and shell thickness of microcapsules were evaluated to understand the self-healing behaviors. At the same time, the capillaring and penetration behaviors were analyzed to evaluate the rejuvenator movement. Properties of virgin and rejuvenated bitumen were compared to evaluate of recovery for aged bitumen.

2. Experimental

2.1. Materials

The shell material was commercial prepolymer of melamine–formaldehyde modified by methanol (solid content was 78.0%) purchased from Aonisite Chemical Trade Co., Ltd. (Tianjin, China). The core material used as rejuvenator is dense, aromatic oil (density is 0.922 g/cm^3, viscosity is 4.33 Pa s) obtained from Petro plus Refining Antwerp (800DLA, Belgium). Styrene maleic anhydride (SMA) copolymer (Scripset® 520, Hercules, USA) was applied as dispersant. A small percentage of the anhydride groups have been established with a low molecular weight alcohol and it is fine, off-white, free flowing powder with a faint, aromatic odor [15]. The bitumen used in this study was 80/100 penetration grade obtained from Kuwait Petroleum. The aged bitumen 40/50 penetration grade was artificially produced by thin film oven test. The 40/50 aged bitumen was then blended with microcapsules using a propeller mixer for 30 min at 160 °C with a constant speed of 200 r min^{-1}.

2.2. Microencapsulation procedure

The method of fabrication microcapsules containing rejuvenator by coacervation proceed can be divided into three steps [16]: (1) SMA (10.0 g) was added to 100 ml water at 50 °C and allowed mix for 2 h. Then a solution of NaOH (10%) was added dropwise adjusting its pH value to 10. The above surfactant solution and rejuvenator were emulsified mechanically under a vigorous stirring rate for 10 min using a high-speed disperse machine. (2) The encapsulation was carried out in a 500 ml three-neck round-bottomed flask equipped with a condensator and a tetrafluoroethylene mechanical stirrer. The above emulsion was transferred in the bottle, which was dipped in a steady temperature flume (room temperature). Half of MMF prepolymer (16 g) was added dropwise with a stirring speed of 500 r min^{-1}. After 1 h, the temperature was increased to 60 °C with a rate of 2 °C min^{-1}. Then another half of prepolymer (16 g) was dropped in a bottle at the same dropping rate. (3) The temperature was increased to 75 °C. After polymerization for 1 h, the temperature was decreased slowly at a rate of 2 °C min^{-1} to ambient temperature. At last, the resultant microcapsules were filtered and washed with pure water and dried in a vacuum oven.

2.3. Morphologies observation

An optical microscope (BX41-12P02, OLYMPUS) was used to check the fabrication process of microcapsules in emulsion. About 1 ml of the colloidal solution was extracted and spread on a clean glass slide (1 × 3 cm). The dried microcapsules were adhered on a double-side adhesive tape without cracking the shells. The surface morphologies were observed by using an Environmental Scan Electron Microscopy (ESEM, Philips XL30) at an accelerated voltage of 20 kV.

2.4. Mean size and shell thickness of microcapsules

The mean size of microcapsules was measured by a laser particle size analyzer (HELOS-GRADIS, SYMPATEC GMBH, Germany). About 2 g microcapsules was mixed in 5 g epoxy resin. After the composite was dried at room temperature, it was carefully cut to obtain the cross-section. The thickness of shells can be measured from the ESEM images of cross-section of microcapsules [17]. At least 20 shells of each microcapsule sample were measured and the average data were calculated.

2.5. Thermogravimetric analysis (TGA)

The thermal stability characterization of microcapsules was performed on a Dupont SDT-2960 Thermogravimetric analysis (TGA) at a scanning rate of 5 °C min^{-1} in a flow of 40 ml min^{-1} nitrogen (N_2).