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Experimental observation of the self-healing microcapsules containing rejuvenator states in asphalt binder

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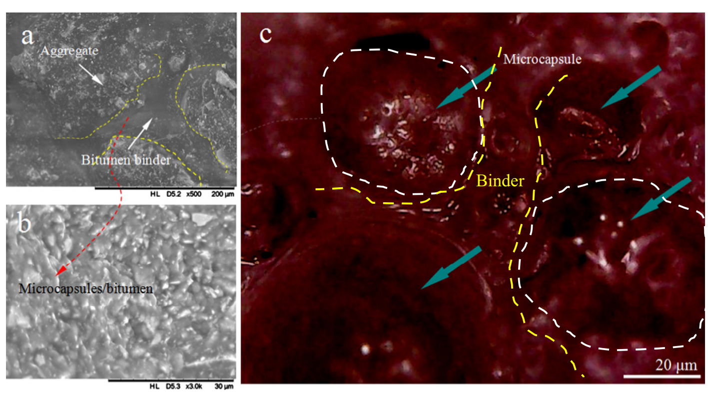
HIGHLIGHTS

- Self-healing microcapsules containing rejuvenator survived in asphalt binders.
- XCT results proved that microcapsules dispersed in asphalt binders homogeneously.
- Microcracks in asphalt binders triggered the break of microcapsules.

GRAPHICAL ABSTRACT

Microstructure morphologies of microcapsules in asphalt sample (MB-7) at room temperature state, (a) ESEM morphology of asphalt with aggregate and bitumen binder, (b) ESEM morphology of microcapsules dispersing in bitumen, and (c) a fluorescence microscope morphology of microcapsules in bitumen binders.



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ABSTRACT

Microcapsules containing rejuvenator are particles used to increase the self-healing capability of asphalt. To date, no reports focus on studying their behaviors in asphalt binders between the aggregates. The purpose of this work was to directly observe the states of self-healing microcapsules in asphalt binders. Asphalt samples were prepared by mixing bitumen and various weight contents of microcapsules. Experimental tests were carried out to observe the morphology, integrity, distribution, thermal stability, interface bonding and triggered rupture of microcapsules in asphalt binders. Fluorescence microscope morphologies and X-ray computed tomography images showed that microcapsules survived in asphalt resisting high temperature and strong agitation without premature damage. At the same time, microcapsules were homogenously dispersed in asphalt binders avoiding particles aggregation and adhesion. A circular heating-cooling process was used to simulate temperature changes in the natural environment. It was found that microcapsules still kept a stable state after an extreme temperature change. In addition, interface debonding phenomenon did not appear. Microscopic observation results reflected that

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microcapsules could be pierced by microcracks in the asphalt binder and the encapsulated rejuvenator flowed out under the force of capillary action. All the above conclusions indicate that microcapsules containing rejuvenator meet the application conditions and play the role of self-healing material in asphalt binders.

1. Introduction

Asphalt is a widely used composite material for pavement. Because of the effect of natural environment, loading and other factors, asphalt becomes brittle and microcracks will generate [1]. The aging problem of bitumen will damage the asphalt original properties, especially its self-healing capability. This capability of self-healing is associated with temperature, healing time and aging degree of itself. Comparing with the deterioration process, the self-healing capability of bitumen is not enough to repair the damage, as surface raveling and reflective cracking, which are caused by aging. Therefore, many researchers have paid attentions to enhance the self-healing capability of bitumen. Several methods have been used including rejuvenation addition, polymer blend, heating induction, and nanoparticles mixture [2]. It has been found that rejuvenator addition is one of the most effective methods [3]. However, oily rejuvenators nearly could not penetrate into the asphalt pavement more than 2 cm [4]. Interestingly, encapsulation rejuvenator mixed in bitumen is an alternative method not only to promote repair capacity but also overcome the disadvantages of oily rejuvenator [5]. When micro-cracks encounter capsules in propagation, capsules triggers to rapture and the rejuvenator fills the cracks under the help of capillarity; at the same time the released rejuvenator reconstitutes the asphalt binder's chemical composition caused by aging [5].

Several approaches have been reported on exploring novel microcapsules through chemical method. García et al. [6,7] reported a work to fabricate capsules containing oily rejuvenator using epoxy resin as a coating material and porous sand as a skeleton material. However, the rejuvenator could not be easily released from the porous sands while the capsules were broken owing to extreme viscidity of healing agents. Sun et al. [8] reported a method to fabricate microcapsules using melamine-formaldehyde (MF) resin as shell material. It was also reported by Pei et al. [9] that the core/shell structure of microcapsules could be fabricated by healing agents and melamine-formaldehyde resin. In previous works, microcapsules were fabricated with MF resin shells by an in-situ polymerization [10–12]. The mechanism of fabrication of capsules is named as a one-step polymerization (OSP) [13]. This structure of microcapsules can not satisfy with non-penetration in the asphalt pavement. In order to obtain higher compact shells, Su [14] fabricated self-healing microcapsules with a methanol modified melamine-formaldehyde (MMF) resin as shell material which was synthesized through a two-step polymerization (TSP). The formation of shells was a two-step coacervation due to the interfacial equilibrium. The rigidity and toughness of shells was promoted by using MMF resin.

Bitumen is a binder material of aggregates, which has a melting point of 180 °C. It can be imaged that self-healing microcapsules need excellent thermal stability and mechanical properties to keep their integrity in asphalt. It was showed that the size of microcapsules, the shell thickness and the core/shell ratio are main factors determining the stability of microcapsules [15]. Inorganic/organic composite shell structure can be formed to improve the thermal and mechanical properties of microcapsules [16]. For example, microcapsules had been fabricated with a nano-$CaCO_3$/polymer shell structure [17]. The size of microcapsules did not greatly affected by the structure of microcapsules with nano-inorganic/organic shells. On the contrary, it was varied that the shell thickness increased due to the addition of nano-$CaCO_3$. Moreover, microcapsule shells could resist a higher temperature and protect microencapsulated rejuvenator. It was noted that the addition of inorganic particles enhanced interaction between the asphalt binders and the microcapsules [18]. The self-healing mechanism of asphalt using microcapsules was investigated by mechanical tests; self-healing process contained four steps: the crack generation, microcapsules broken, rejuvenator release and rejuvenator capillarity-diffusion [19]. Oily rejuvenator flowed along microcracks with the help of capillarity and diffused into the aging bitumen [5].

Although many efforts have been carried out to investigate microcapsules containing rejuvenator in bitumen, the states of microcapsules in asphalt binders have not been explored. Asphalt is composed of bitumen, aggregate particles and air voids. Self-healing process in asphalt normally occurs in the binders. After several years of use, bituminous material loses part of its visco-elastic capability. At the same time, microcracks occur and develop at the interface between binders and aggregates. Microcapsules in asphalt binders are expected to keep ideal states in a self-healing process. In view of the above, the purpose of this work was to analyze the morphology, distribution, thermal stability of microcapsules in asphalt binders. In addition, microcracks were generated to determine the trigger rapture state of microcapsules in asphalt binders. Based on these observations, it can be deduced a conclusion about the application possibility of the self-healing microcapsules in asphalt binders.

2. Experimental method

2.1. Materials

Bitumen was supplied by Qilu Petrochemical of China. The aged bitumen sample (40/50 penetration grade) was artificially produced with 80/100 penetration grade bitumen using a thin film oven method [5]. Rejuvenator (0.905 g/cm^3, 4.24 Pa·s) was obtained from Shanghai Chem. Co., Ltd. of China. Methanol melamine-formaldehyde (MMF) was supplied by Aonisite Chemical Trade Co., Ltd. (Tianjin, China). Styrene maleic anhydride (SMA) was applied as dispersant copolymer (Scripset® 520, Hercules, USA) [14]. Nano-$CaCO_3$ powder (mean size 20 nm) was purchased from Tianjin Sinago Technology Co., Ltd. of China.

2.2. Microcapsules fabrication process

The method of fabrication microcapsules containing rejuvenator was divided into three steps [20]: (1) SMA powder was added into 50 °C water mixing for 2 h, and then the solution was adjusted pH value to 10 by a 0.1 mol L NaOH solution. Oily rejuvenator was added with a stirring rate stirring speed of 500 r min^{-1} for 10 min. (2) The above emulsion was transferred into a three-neck bottomed flask. MMF resin was added with a stirring speed of 300 r min^{-1}. At the same time, temperature was elevated to 80 °C with a speed of 2 °C min^{-1}. (3) The polymerization was kept for 2 h, and then the temperature was decreased to room temper-