



## Stability investigation of self-healing microcapsules containing rejuvenator for bitumen

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### ABSTRACT

Preservation and renovation bitumen of pavement is a big problem for the whole world. Traditionally, application rejuvenator is the only one method that can restore the original properties of the pavements. However, some puzzles still restrict its successful usage. Microencapsulation is a promising method to apply rejuvenator in bitumen. These microcapsules can break and leak the oily-liquid rejuvenator into microcracks and self-healing the aged bitumen. Based on our previous work, the objective of this study was to investigate the thermal stability, mechanical stability and interface stability of microcapsules in bitumen. The results showed that these microcapsules containing rejuvenator survived in melting bitumen and in a violent repeated temperature changes. Microcapsules had the elastic–plastic deformation ability resisting the temperature changes and mixing stress. Moreover, the chemical bonds improved the interface stability between shells and bitumen. Microcapsules containing rejuvenator will be a promising product to realize the smart pavements.

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### 1. Introduction

The decline in the use of natural bitumen for road construction can be traced to the 1910s when the advent of vacuum distillation made it possible to obtain artificial bitumen from crude oil. Currently, 95% of the almost 100 million tons of bitumen produced worldwide each year is applied in the paving industry, where the bitumen essentially acts as a binder for mineral aggregates to form asphalt mixes [1]. Other uses of bitumen are as emulsions, waterproof materials, or formed materials, but these account for less than 5% of the total bitumen produced. As a widely applied material in pavements, bitumen must be sufficiently fluid at high temperature (around 160 °C) to be workable and allow for homogenous coating of the aggregates upon mixing. Another important issue that has to be considered is the extent to which it ages from climate and traffic. After years of use the stiffness of asphalt concrete increases while its relaxation capacity decreases. This causes the binder to become more brittle, causing the development of microcracks and ultimately to cracking of the interface between the aggregates and

binder [2]. This occurs mainly as a result of oxidation of the hydrocarbon compounds contained within the bitumen [3]. Bitumen binders are usually categorized into two subdivisions: solids called asphaltenes and liquids called maltenes. Maltenes can be further divided into polar aromatics, naphthalene aromatics, and saturates (paraffins) [4]. The main aging mechanism of bitumen is the loss of volatiles and oxidation, which leads to bitumen with higher viscosity (stiffer) [5]. In other words, the amount of solid component increases and that of the liquid component decreases, thus resulting in an increase in the rigidity of the pavement.

The aging problem of bitumen leads to pavement failure, including surface raveling and reflective cracking. It therefore increases the cost of renovating and preserving bituminous pavements [6]. Several physical and chemical methods are currently employed for bitumen preservation, including the use of rejuvenator emulsions or fog seals, and through additive modification or thin overlay technologies [1,7]. However, of these methods only the first, i.e., the application of rejuvenators, can restore the original properties of the pavement [6]. The most important goal of using rejuvenator product is to restore the asphaltene/maltene ratio to its original balance [8]. Rejuvenating agents have the ability to reconstitute the binder's chemical composition and they consist of lubricating and extender oils that contain a high proportion of maltene constituents [9]. Rejuvenator can soften the aged binder and provide comprehensive rejuvenation that replenishes the

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volatiles and dispersing oils while simultaneously promoting adhesion.

However, for a rejuvenator to be successfully applied the difficulty in penetrating the pavement surface still remains a significant problem. Shen et al. [10] reported on the use of three rejuvenators and found that none could penetrate more than 2 cm into the asphalt concrete. Further issues encountered when applying these materials include the fact that road closures are necessary for some period of time after their application. The rejuvenator may also cause a high reduction in the surface friction of the pavement for vehicles. Moreover, these rejuvenators may also be harmful to the environment.

To overcome these issues inspiration is provided by the concept of self-healing based on microcapsules. This particular approach involves incorporation of a microencapsulated healing agent and a dispersed catalyst within a polymer matrix [11–13]. Upon damage-induced cracking, the microcapsules are ruptured by the propagating crack fronts resulting in release of the healing agent into the cracks by capillary action [14]. The method of encapsulating rejuvenators inside the bitumen may be an alternative approach worthy of consideration. The application of microcapsules containing rejuvenator to bitumen derives from the success observed for some polymer self-healing materials [15–17]. García et al. [18] reported a method to prepare rejuvenator capsules by using an epoxy resin as a coating and porous sand as a skeleton. The advantages of these capsules include the fact that they are strong enough to resist the mixing process, the high temperature, and all the years in the road until they are required. However, these capsules have some limitations that restrict their application. The primary limitation is that it is hard for the rejuvenator to flow out from the porous sand when the shell is broken because the

rejuvenator has a high viscosity resulting from it consisting of lubricating and extender oils. The capillary action of the porous structure also limits the rejuvenator release. Another limitation is that the capsule size does not fit with the thickness of bitumen between aggregates. To realize the application of these promising chemical products, we have developed a novel method to fabricate microcapsules containing rejuvenator by in situ polymerization using methanol-melamine-formaldehyde (MMF) prepolymer as shells [19]. A two-step coacervation process, with the aid of styrene maleic anhydride (SMA) as a surfactant, was successfully applied to enhance the thermal stability and compactibility of the shells. It has been shown that this product is an environmentally friendly powder that encapsulates a suitably sized rejuvenator for chemical and construction engineering.

To produce microcapsules containing rejuvenator by chemical means, the factors of cost, complexity, and capacity must be considered for the construction industry. These core-shell microcapsule structures need to meet specific requirements in terms of size distribution, encapsulation ratio, and non-biodegradability because these factors will influence their service performance [20]. As bitumen acts as thin layers between aggregates that are usually less than 50  $\mu\text{m}$ , the size of the microcapsules containing rejuvenators should be smaller than 50  $\mu\text{m}$  to avoid being squeezed or pulverized during asphalt forming. Besides the complex fabrication process of microcapsules containing rejuvenator, their survival ability is another important issue that must be addressed. Fig. 1 illustrates the possible states of the microcapsules in bitumen material. First, for practical application the microcapsules must have high thermal stability and high mechanical strength to resist the melting temperature and mixing stress of the asphalt (Fig. 1a–c). The microcapsules must maintain their shape and compatibility at

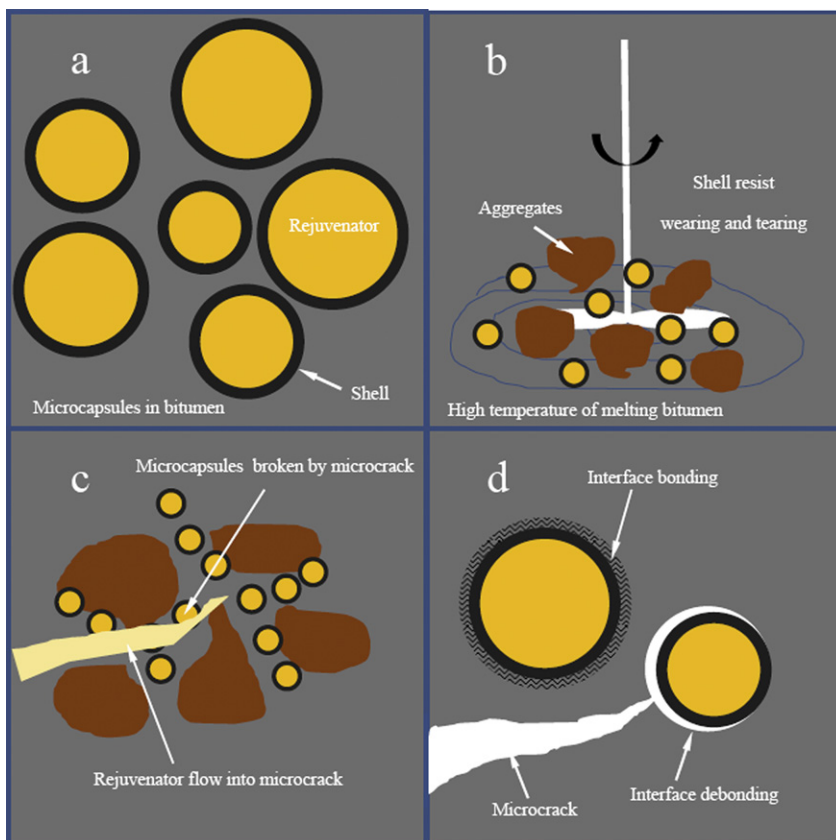


Fig. 1. Illustration of the possible states of microcapsules containing rejuvenator in bitumen.

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