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Microcapsules for self-healing of asphalt mixture without compromising mechanical performance

T. Al-Mansoori [a], R. Micaelo [a,b,c], I. Artamendi [d], J. Norambuena-Contreras [a,e], A. Garcia [a,*]

[a] Nottingham Transportation Engineering Centre, School of Civil Engineering, University of Nottingham, University Park, Nottingham, UK
[b] DEC, FCT, Universidade NOVA de Lisboa, 2829-516 Caparica, Portugal
[c] CERIS, CESUR, Universidade de Lisboa, 1049-001 Lisboa, Portugal
[d] Aggregate Industries UK Limited, Derbyshire DE6 3ET, UK
[e] LabMAT, Department of Civil and Environmental Engineering, University of Bío-Bío, Concepción, Chile

HIGHLIGHTS

- Calcium-alginate microcapsules containing sunflower oil were manufactured.
- Microcapsules did not affect the mechanical properties of asphalt mixture.
- A new test for asphalt self-healing by the action of microcapsules was proposed.
- Microcapsules caused a significant improvement on asphalt self-healing.

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ABSTRACT

In this research calcium-alginate capsules containing vegetable oil that can release their content due to a mechanical trigger have been made and mixed in asphalt mixture to improve its natural self-healing properties. The physical, mechanical and self-healing properties of asphalt mixture containing these capsules have been evaluated for the first time. Three different capsule contents were used, with oil-to-bitumen ratio 1.1, 2.8 and 5.5, respectively. Capsules were strongly bonded to the asphalt mixture and results showed similar mechanical performance to that of asphalt with and without capsules in the water sensitivity, particle loss and permanent deformation tests. This shows that capsules for asphalt self-healing can be safely used in the road, without affecting its quality. Asphalt containing capsules had slightly lower stiffness, which can be easily solved by reducing the size of the capsules in the future. Furthermore, a new method for testing asphalt containing capsules was designed and tested. It was found that cracked asphalt mixture with capsules recovered 52.9% of initial strength at 20 °C versus 14.0% of asphalt mixture without capsules.

1. Introduction

Asphalt mixture, which consists of mineral aggregates (90% v/v) with size ranging from a few microns to 2–3 cm and bitumen (10% v/v), is the most widely used road surfacing material. Aggregates give structural stability to asphalt, while bitumen, a fluid with temperature dependant viscosity, binds the aggregates together. To produce asphalt mixture, aggregates and bitumen have to be mixed together at approximately 180 °C to allow coating the aggregates with bitumen. After, the material is mechanically compacted to reach maximum packing of the aggregates at approximately 140 °C [1].

After a number of years, the effects of traffic load, oxygen, UV radiation, thermal cycling and ambient moisture result in oxidation of the bitumen and microcracking in the mastic (bitumen with fine aggregate inclusions) [2]. Moisture impregnates bitumen by Fickian diffusion [3] and reduces adhesion to the aggregates [4]. Mechanical loads then create interfacial cracks, which accelerates the water penetration until maintenance is needed. However, it has been demonstrated that draining bitumen into the microcracks can cause them to heal [5]. The draining flow is Newtonian above 20–30 °C, depending on the bitumen type, and can be predicted using the Navier-Stokes equations as a function of viscosity, gravity and surface energy [5]. Since bitumen is a very high viscosity liquid

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* Corresponding author.
E-mail address: alvaro.garcia@nottinghmam.ac.uk (A. Garcia).

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at the ambient temperature, asphalt self-healing occurs in a period of days [6]. However, due to the recurrent flow of traffic, the rate of crack growth may be greater than that of healing and hence macro-cracks may still form [7].

Asphalt self-healing can be accelerated from days to seconds by the addition of capsules containing rejuvenators during the mixing process. The idea is that these capsules will remain inactive in the asphalt road for several years until an external factor, e.g. an excessive loading, triggers the release of rejuvenators that dissolve the bitumen around the microcapsules [8]. As a consequence, the binder can easily flow into the cracks and self-healing is accelerated. Previous researchers have prepared capsules for asphalt self-healing. For example, by encapsulating rejuvenators using (1) in-situ polymerization of urea-formaldehyde [9], methanol-melamine-formaldehyde [10] or phenol-formaldehyde [11], (2) ionotropic gelation of sodium alginate in calcium chloride solution [12] and (3) saturating porous aggregates with rejuvenators and sealing them with a shell made of epoxy resin and cement [8]. In general, capsules resist the mixing and compaction operations and release the rejuvenator due to loading [13]. Moreover, crack healing induced by capsules has been shown in bitumen [14,15] and in asphalt mixture [12]. However, the development of capsules containing rejuvenators is still at an early stage and their effect on the durability of asphalt mixture is unclear.

There is a strong suspicion among the international research community that capsules may affect the durability and performance of asphalt roads. For example, the release of rejuvenator in asphalt mixture could compromise the load bearing capacity and resistance to permanent deformation, ravelling, bleeding, etc. In addition, as it was reported by Gilabert et al. [16], who studied numerically the effect of the interface bonding strength and the stress concentration around capsules embedded in a polymeric matrix, capsules could influence the stress concentration patterns and the initiation and propagation of cracks in the road. Furthermore, based on the experience of the authors, it is difficult to test the self-healing properties of asphalt containing capsules with the existing testing methods.

Therefore, the objective of this paper is to provide the first overview of the effect of calcium-alginate capsules on the mechanical performance and self-healing properties of asphalt mixture. With this purpose, an extensive experimental programme has been performed in asphalt mixture containing capsules, based on standardised tests commonly used by industry. Standard testing protocols were used to evaluate the most important properties, such as water sensitivity, particle loss, stiffness, rutting resistance and fatigue. In addition, a new test to measure the self-healing properties of asphalt mixture containing capsules was designed and used to produce the self-healing results in this paper.

2. Materials and testing methods

This section describes the experimental programme followed in this study, see Fig. 1.

2.1. Capsules

Calcium-alginate capsules with average diameter 2.9 mm (see Fig. 2) were produced by the authors, with the materials and procedures described in a previous research [12]. The rejuvenator was sunflower oil, which has been used in previous research as a healing agent [17–19]. The materials used to make the capsules were (1) sodium alginate ($C_6H_7O_6Na$) (Sigma-Aldrich), (2) calcium chloride ($CaCl_2$) (Sigma-Aldrich) and (3) sunflower oil (East End, UK). First, an emulsion was prepared by mixing 60 g of oil and 15 g of sodium alginate in 600 ml of water at a high shear rate, 3,000 rpm. After, the emulsion was introduced in a dropping funnel with a 3 mm socket and let to drop in a calcium chloride solution prepared with 600 ml of water and 12 g of calcium chloride under continuous agitation using a magnetic stirrer. Capsules were allowed to stay in the solution until the end of the encapsulation process. Finally, the microcapsules were washed with deionized water and dried by introduce them in an electric dryer at 40 °C during 12 h. Table 1 presents the dimensional and strength properties of capsules, obtained in [12], where it was shown that they are able to survive asphalt mixing and compaction.

2.2. Asphalt mixture

Dense asphalt concrete mixture, AC20 base 40/60, see EN 13108-1, was selected for this study. The bitumen selected was paving grade 40/60. The aggregates were Tunstead limestone and Table 2 presents the aggregate gradation and volumetric properties of the asphalt mixture. Three different capsule contents, in terms of mass proportion into the total mixture, were tested, 0.10%; 0.25% and 0.50%. These values correspond to an approximate oil-to-bitumen content by mass in bitumen of 1.1%, 2.8% and 5.5%, respectively. Moreover, in reference [12] it was concluded that these capsules could release oil to the mixture during mixing and compaction. Therefore, an equivalent amount of sunflower oil, as it is detailed in Section 3.1, was added to the asphalt mixture without capsules to differentiate the self-healing caused by the capsules from that caused by the oil released during the fabrication process. It was assumed that the oil released during mixing and compaction was uniformly dispersed in asphalt mixture.

Asphalt mixture was produced in the laboratory in batches of approximately 14 kg, using a lab mixer equipped with a helical horizontal mixing shaft. The aggregates and binder were preheated at 160 °C for 12 h and 4 h, respectively, while capsules were not pre-heated. The materials were mixed for 2 min at 125 rev/min at 160 °C, for adequate dispersion. Then, the mixture was transferred to the moulds for subsequent compaction.

Cylindrical and prismatic test specimens were used for the research. Prismatic specimens, approximately $150 \times 100 \times 60\ mm^3$, were cut from $306 \times 306 \times 60\ mm^3$ slabs compacted using a lab roller compactor to reach air voids content 5%. Cylindrical test specimens, with 100 mm diameter and approximately 50 mm height, were compacted using a gyratory compactor, with an inclination angle of 2.0° and 650 kPa of static pressure. Not more than 250 gyrations were applied during compaction.

2.3. Amount of oil released by the capsules

Attenuated Total Reflection – Fourier Transform Infrared spectroscopy (ATR-FTIR), measured with a Bruker Tensor 27 spectrometer was used to determine the amount of oil released by capsules during asphalt mixing and compaction. The sunflower oil had a distinct absorption peak at $\sim 1745\ cm^{-1}$ while bitumen did not have it, see Fig. 3. The test was set in the absorption mode in the range of 400 to $4000\ cm^{-1}$ and with a resolution of $4\ cm^{-1}$. Oil release measurements were calculated by comparing the percentage of oil released from damaged capsules versus all the oil introduced in the capsules. The normalised area under the FTIR was used to give an indication of the oil in the mixture [30]. Three oil-to-bitumen ratios were used to simulate full release of oil after breaking all the capsules, 1.1%, 2.8% and 5.5%, that corresponded to asphalt mixtures with capsules content 0.10%, 0.25% and 0.50% by total mass in the mixture, respectively. For sampling mastic, compacted asphalt specimens were heated to 100 °C, broken by hand and mastic samples were collected by using a hot knife from the surface of coarse aggregates. To calculate the amount of oil released by the capsules, the normalised area under the FTIR curve,