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Effect of mixing and ageing on the mechanical and self-healing properties of asphalt mixtures containing polymeric capsules

J. Norambuena-Contreras [a,b,*], E. Yalcin [a,c], A. Garcia [a], T. Al-Mansoori [a], M. Yilmaz [c], R. Hudson-Griffiths [d]

[a] Nottingham Transportation Engineering Centre, School of Civil Engineering, University of Nottingham, University Park, Nottingham, UK
[b] LabMAT, Department of Civil and Environmental Engineering, University of Bío-Bío, Concepción, Chile
[c] Firat University, Faculty of Engineering, Department of Civil Engineering, Elazig, Turkey
[d] Highways England, Safety, Engineering and Standards – Pavement Materials, 199 Wharfside Street, Birmingham, UK

HIGHLIGHTS

- Calcium-alginate capsules for self-healing of asphalt mixtures were manufactured.
- Physical, thermal and mechanical properties of the capsules were evaluated.
- Mixing and ageing effect on the mechanical and healing properties of mixtures were studied.
- Capsules addition did not improve the mechanical properties of mixtures.
- Self-healing levels obtained by the mixtures varied depending on the capsules addition order.

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ABSTRACT

In this paper, polymeric capsules with sunflower oil as the encapsulated rejuvenator were manufactured and added to asphalt mixtures to improve their self-healing properties. A capsule content of 0.5% by total mass of mixture was added to the asphalt samples. Physical, thermal and mechanical properties of the capsules were evaluated. Additionally, the effect of the mixing order and the ageing time on the mechanical stability and self-healing properties of asphalt mixtures with, and without, capsules were evaluated through stiffness modulus and flexural strength tests. Self-healing properties of asphalt mixtures were evaluated through three-point bending tests on cracked asphalt beams with, and without, capsules. In addition, capsules' distribution and their integrity inside the asphalt mixtures were analysed using X-ray computed tomography. The main results proved that the capsules can resist the mixing and compaction conditions and break inside the mixture releasing the encapsulated oil in small volumes. In addition, it was observed that the addition of capsules did not improve the stiffness modulus of asphalt mixtures compared to mixtures without capsules, and that the mixing order and the ageing time did not have a significant influence on the flexural strength of the mixtures. Moreover, the healing levels obtained by the mixtures varied depending on the order of addition of capsules, and mixtures with capsules showed higher healing levels than mixtures without capsules. Finally, the levels of healing for the mixtures without ageing were greater than those of mixtures after the ageing process.

1. Introduction

An asphalt mixture is composed of aggregates and bitumen, and it is the most used material to build road pavements worldwide. Aggregates usually give strength and structural stability, while bitumen works as a binder. The capacity of pavements to carry loads depends on the bond between aggregate particles provided by the bitumen [1]. However, this bond degrades over time due to the most significant issue that bitumen faces, which is the damage by ageing [2]. The damage by ageing results from oxidation and loss of the volatiles from bitumen composition, which causes stiffness and an increase in viscosity. This leads to the appearance of microcracks which evolve to form cracks and the detachment or ravelling of aggregates in asphalt pavements, reducing their mechanical performance over time [3].

Hence, to maintain pavements in good condition during their lifetime, external maintenance is usually required by the road

* Corresponding author at: LabMAT, Department of Civil and Environmental Engineering, University of Bío-Bío, Concepción, Chile.
E-mail address: jnorambuena@ubiobio.cl (J. Norambuena-Contreras).

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agencies of each country [4]. The most common solution to restore the original properties of aged pavements and reconstitute the bitumen chemical composition is to use rejuvenators, which are low viscosity oils with high maltenes content, on the surface of asphalt roads [5]. However, spreading rejuvenators over the surface of roads has side effects, such as reducing the skid resistance of the pavement surface because they penetrate only for the first few centimetres. Additionally, roads must be closed for some time after the application of rejuvenators [6].

To overcome the disadvantages of using rejuvenators on the pavement surface, researchers have proposed the use of encapsulated rejuvenators to restore the original properties of the bitumen via self-healing processes[6–14]. This technique is supported by the fact that bitumen is a self-healing material with the ability to close microcracks by itself [10]. The principle behind this approach is that these capsules containing rejuvenators will remain inactive in the asphalt road for several years until external damage happens to the asphalt pavement [13]. Consequently, cracks will break the shell of the capsules in a timely manner, leading them to release the rejuvenator into the asphalt medium, which will diffuse and reduce the bitumen viscosity, so it can easily flow into the cracks [11].

Different methods have been used to manufacture microcapsules, or capsules, with encapsulated rejuvenators for asphalt self-healing purposes. For instance, Su et al. [9] prepared microcapsules containing rejuvenator droplets by in-situ polymerisation of urea–formaldehyde, making a Methanol-Melamine-Formaldehyde (MMF) prepolymer as a shell. Garcia et al. [11] prepared capsules of a larger size by saturating porous sand with sunflower oil as rejuvenator material, protected by a hard shell of cement and epoxy resin. Additionally, another types of calcium-alginate capsules have recently been published by Micaelo et al. [12], Al-Mansoori et al. [13–15] and Xu et al. [16]. These capsules were made by the ionotropic gelation of sodium alginate in the presence of calcium chloride solution. In these capsules, the encapsulated healing agents were sunflower oil or an industrial rejuvenator and their sizes were a few millimetres.

The current literature states that the capsules with encapsulated rejuvenators can resist to asphalt manufacture and release the rejuvenators only when broken due to external loading. Furthermore, capsules addition had a positive effect on the durability and safety of asphalt mixtures [12], and proved the self-healing ability of aged bitumen and rejuvenated aged asphalt mixture [13–16]. Nevertheless, there are still many open research questions that need to be addressed with the aim of understanding the phenomena associated to the development of new self-healing asphalt mixtures by the action of capsules. For example, the effect of the mixing method of the capsules and ageing conditions of the asphalt on the mechanical stability and self-healing properties of asphalt mixture with capsules is not clear yet. Another little researched areas concern the influence of mixing temperature on the physical, thermal and mechanical properties of the polymeric capsules, as well as the distribution and integrity of the capsules inside the asphalt mixtures.

This paper aims to evaluate the effect of mixing and ageing conditions on the mechanical stability and self-healing properties of asphalt mixtures with, and without, capsules. With this purpose, polymeric calcium-alginate capsules for asphalt self-healing have been designed and experimentally tested in asphalt mixture specimens prepared using different mixing orders and ageing conditions. The manufacturing process of the capsules and their physical, thermal and mechanical properties at different temperatures are presented in the paper. The mechanical tests applied in this study to evaluate asphalt mixtures were stiffness modulus and flexural strength. Self-healing properties of the studied asphalt mixtures were quantified through three-point bending tests on cracked asphalt beams. Finally, an X-ray computed tomography analysis was carried out to evaluate the capsules' distribution and their integrity inside the asphalt mixture.

2. Materials and methods

2.1. Materials

A standard, dense asphalt mixture AC 20 base (according to EN 13108-1) and calcium-alginate capsules for asphalt self-healing were used. Asphalt mixture consists of virgin bitumen 40/60 pen with a density of 1.030 g/cm^3 and a softening point of 49.8 °C, and graded Tunstead limestone aggregate with a density of 2.700 g/cm^3. Table 1 shows the aggregate gradation and design properties of the mixture used. Polymeric capsules with a density of 1.116 g/cm^3 were made of a calcium-alginate polymer that encapsulated the rejuvenator. The rejuvenator used in the capsules was sunflower oil, with a density, smoke point and flash point of 0.92 g/cm^3, 227 °C and 315 °C, respectively [12]. Sunflower oil was selected because of its low cost, thermal stability, and the fact that extra health and safety procedures are not required in the laboratory [17]. Furthermore, the polymer structure of the capsules was made of sodium alginate ($C_6H_7O_6Na$) and a calcium chloride ($CaCl_2$) source, provided in granular pellets of 7 mm diameter and 93% purity.

2.2. Encapsulation procedure of capsules

Calcium-alginate capsules with a water and oil ratio of 0.1 were prepared at 20 °C by ionotropic gelation of alginate in the presence of calcium [13]. A scheme of the manufacturing process of the capsules is illustrated in Fig. 1. To prepare the capsules, first, 150 ml of sunflower oil and 1500 ml of deionised water were introduced into a 2000 ml glass container and stirred to produce a stable emulsion. Sunflower oil and water were mixed using a laboratory gear drive mixer for 1 min at 400 rpm. Then, 55 g of sodium alginate were added to the glass container and stirred until complete solution at 400 rpm for 10 min, see Fig. 1. The amount of the sodium alginate used to produce the polymer capsules (see Fig. 2(a)) was defined so as to obtain a structure of calcium-alginate (see Fig. 2(b)) strong enough to hold the oil and hence with a strength higher than the capsules developed by Micaelo et al. [12] and Al-Mansoori et al. [13].

Simultaneously, a calcium chloride solution at 2%$_w$ was prepared by mixing 1800 ml of deionised water with 36 g of calcium chloride in a 5000 ml glass container, see Fig. 1. Capsules were formed by letting the oil-in-water emulsion drop into the calcium chloride solution from a 2000 ml pressure-equalising dropping funnel with 3 mm socket size. The process is called a cross-link of calcium-alginate via ionotropic gelation of sodium alginate in the presence of calcium ions [18]. During the capsule formation process, the calcium-chloride solution was gently agitated using a magnetic stirrer at 60 rpm. Capsules were allowed to stay in the solution until the end of the encapsulation process. In this study, the capsules' manufacturing process was of 2–3 h, approximately. After this time, the capsules were decanted and washed with deionised water and then dried in an electric dryer at 40 °C for 36 h. Then, the capsules were stored in a freezer at -18 °C to avoid the release and oxidation of the oil at room temperature. The encapsulated procedure allowed the manufacture of capsules with an average diameter of 2.5 mm and composed of a 75% vol. of sunflower oil and a 25% vol. of calcium-alginate polymer. A total of 3 kg of capsules were manufactured in this study.

Table 1
Gradation and design properties of asphalt mixture AC 20 base 40/60.

Sieve size (mm)	% passing
20	100.0
10	80.2
6.3	60.3
4	45.3
2	29.7
0.5	15.5
0.125	9.9
0.063	8.0
Property	**Value**
Binder content (%$_M$)	4.5
Bulk density (g/cm^3)	2.384
Air voids (%)	4.5
Voids in mineral aggregate (%)	14.9
Voids filled with bitumen (%)	69.8