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FINAL SUMMARY REPORT

RESEARCH PROJECT REF. 558065

"SELF-HEALING ASPHALT USING EMBEDDED CAPSULES"

University of Nottingham

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TABLE OF CONTENTS

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1. SUMMARY

In this report, calcium-alginate capsules with sunflower oil as the encapsulated rejuvenator were manufactured and added to asphalt mixtures (dense and stone mastic asphalt) 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 of asphalt mixture and break inside the mixture releasing the encapsulated oil. In addition, it was observed that the addition of the capsules used in the study reduced 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, rutting, and skidding resistance of asphalt mixture. Capsules showed a good spatial distribution inside the SMA samples. The capsules content in asphalt mixture has a significant influence on the healing level, where a higher capsule content led to obtaining higher healing levels. Likewise, asphalt with, and without, capsules presents an increase of the healing level when the temperature increases. Finally, it was proven that the healing temperature has higher influence on the healing levels of the asphalt below 40ºC. Overall, the results proved that the capsules with improved asphalt self-healing properties can be safely used to improve the durability of asphalt roads but it is still unclear at which moment they would release their content in the road and, for that reason, additional tests are still required until they can be safely implemented in a road.

2. BACKGROUND

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 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 example, 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. Details of another type of polymeric capsule were published by Micaelo et al. [12] and Al-Mansoori et al. [[13], [14]]. These capsules were made by the ionotropic gelation of sodium alginate in the presence of calcium chloride solution. In these capsules, the encapsulated material was also sunflower oil and their size was a few millimetres. Broadly, these studies proved that the capsules are resistant to asphalt fabrication and release the rejuvenators only when broken due to external loading, and they have a positive effect on the durability and safety of asphalt mixtures [12]. Additionally, these capsules proved the self-healing ability of aged bitumen and rejuvenated aged asphalt mixture; although these studies did not evaluate 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 mixtures with capsules.

In this work, the mixing methodology of asphalt containing capsules will be investigated. Furthermore, the effect of the amount of capsules on the mechanical and self-healing properties of asphalt mixture will be analysed by means of experimental tests. Finally, in the report, dense and Stone Mastic Asphalt (SMA) have been used.

3. MATERIALS AND METHODS

3.1 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 49.8ºC, and graded Tunstead limestone aggregate with a density of 2.700 g/cm^3. Furthermore, a Stone Mastic Asphalt mixture SMA 14 surf 40/60 and calcium-alginate capsules were used. The SMA mixture consists of virgin bitumen 40/60 pen with a density of 1.030 g/cm^3 and graded Tunstead limestone aggregate with a density of 2.700 g/cm^3.

Table 1: Aggregate gradations and design properties of the mixtures used.

	Dense asphalt	Stone mastic asphalt
Aggregates gradation	% passing	% passing
20 mm	100.0	100.0
14 mm	--	98.6
10 mm	80.2	96.15
6.3 mm	60.3	60.3
4 mm	45.3	--
2 mm	29.7	29.7
0.5 mm	15.5	--
0.125 mm	9.9	--
0.063 mm	8.0	8.0
Properties	Value	Value
Binder content (%$_M$)	4.50	5.90
Bulk density (kg/m^3)	2,384	2,243
Air voids (%)	4.5	5.5
Voids in mineral aggregate (%)	14.9	14. 0
Voids filled with bitumen (%)	69.8	75.0

In addition, 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/cm3, 227ºC and 315ºC, 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 [15]. 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.

3.2 Encapsulation procedure of sunflower oil

Calcium-alginate capsules with a water and oil ratio of 0.1 were prepared at 20ºC by ionotropic gelation of alginate in the presence of calcium [*13*]. A scheme of the manufacturing process of the capsules is illustrated in Figure 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 minute 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 minutes, see Figure 1. The amount of the sodium alginate used to produce the polymer capsules (see Figure 2(a)) was defined so as to obtain a structure of calcium-alginate (see Figure 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].

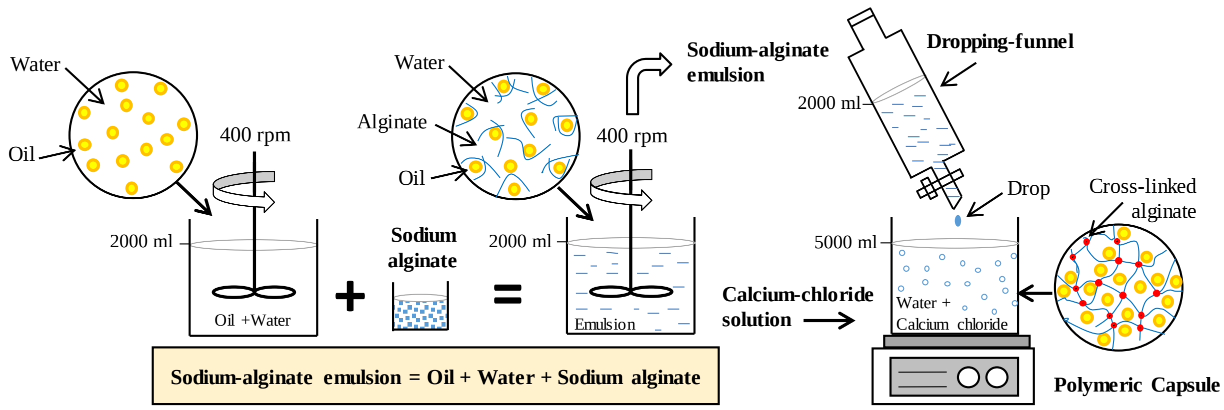

Figure 1. Schematic diagram of the manufacturing process of capsules by ionotropic gelation of alginate.

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 Figure 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 [16]. 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 hours, approximately. After this time, the capsules were decanted and washed with deionised water and then dried in an electric dryer at 40ºC for 36 hours. Then, the capsules were stored in a freezer at -18ºC 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.

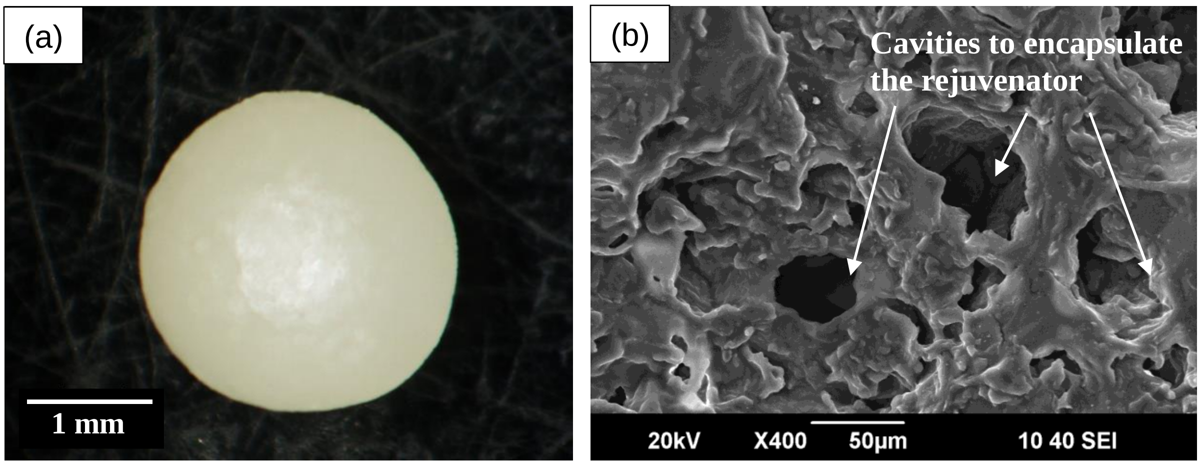

Figure 2. (a) Individual polymer capsule, and (b) SEM image of calcium-alginate internal structure of capsules.

3.3 Manufacturing of asphalt mixture test samples

Prismatic and cylindrical asphalt mixture samples with and without capsules were manufactured in this study. A capsule content of 0.5% by total mass of the mixture was added in the test samples. This amount provided an approximate oil-to-bitumen content of 6.97% by mass in bitumen for the dense asphalt mixture, and of 5.23% by mass of bitumen for the stone mastic asphalt, which is approximately set with the oil-to-bitumen content (6% to 8%) recommended by Ji et al. [17]. Furthermore, asphalt mixtures were produced in the laboratory in batches of approximately 14 kg, using a lab mixer equipped with a helical horizontal mixing shaft. Capsules were pre-heated and mixed in four different conditions with the aim of evaluating the effect of the mixing order on the mechanical and self-healing properties of asphalt mixtures.

The resilience of capsules against different mixing methodologies was evaluated in dense asphalt. The pre-heating and mixture types for asphalt with, and without, capsules were as follows:

- Mixture 1: the aggregates were pre-heated at 160ºC for 12 h, while the bitumen and the capsules were pre-heated at 160ºC for 4 h. The capsules were mixed at the beginning of the mixing process with the aggregates, see Figure 3(a). After, all the components were mixed for 2 minutes at 125 rpm at 160ºC, ensuring an adequate dispersion.

- Mixture 2: the aggregates were pre-heated at 160ºC for 12 h, while the bitumen and the capsules were pre-heated at 160ºC for 4 h. Aggregates and bitumen were mixed for 2 minutes at 125 rpm at 160ºC, ensuring an adequate dispersion. The capsules were mixed 20s before the end of the mixing process with the asphalt mixture.

- Mixture 3: the aggregates and bitumen were pre-heated at 160ºC for 12 h and 4 h, respectively, while the capsules were left to defrost for two hours at 20ºC before mixing. The capsules were mixed at the beginning of the mixing process with the aggregates, as in Mixture 1.

- Mixture 4: the aggregates and bitumen were pre-heated at 160ºC for 12 h and 4 h, respectively, while the capsules were left to defrost for two hours at 20ºC before mixing. The capsules were mixed 20 s before the end of the mixing process with the asphalt mixture, as in Mixture 2, see Figure 3(b). This mixing mode will be used to measure the mechanical properties of asphalt in the successive.

- Reference mixture (WO/C): the aggregates and bitumen were pre-heated at 160ºC for 12 h and 4 h, respectively. Then, the raw materials were mixed for 2 minutes at 125 rpm at 160ºC, ensuring an adequate dispersion.

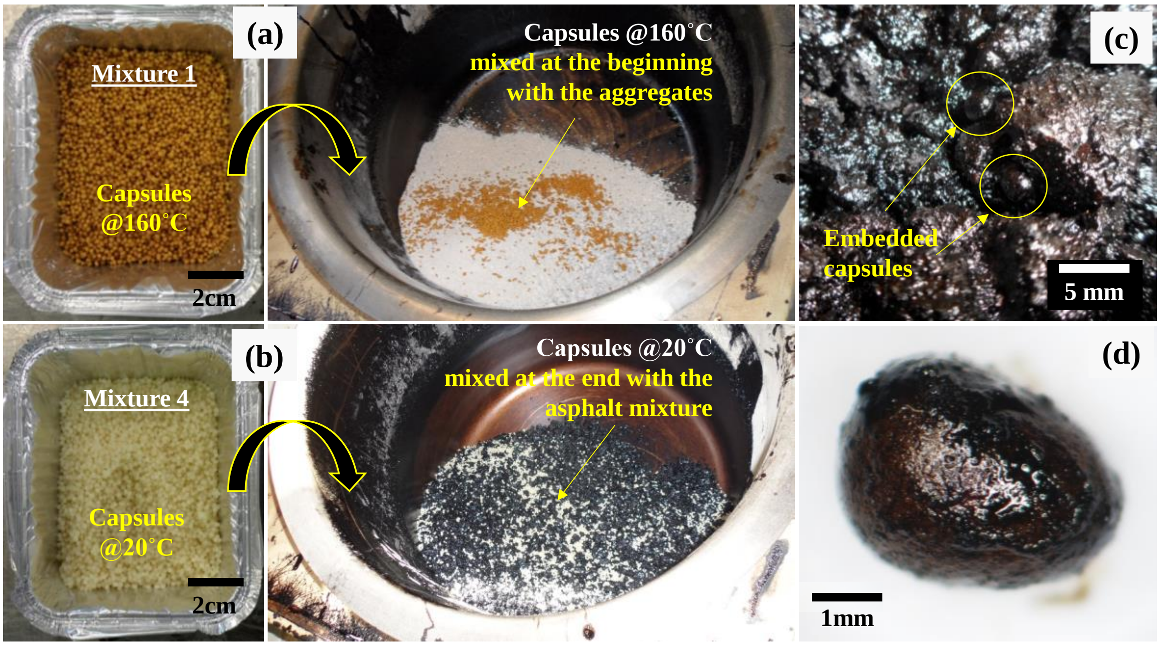

Figure 3. Images of (a-b) pre-heating and mixing process for the asphalt mixture types 1 and 4, (c) embedded capsules in mixture, and (d) individual capsule extracted after mixing process.

After mixing, the asphalt was transferred to the moulds for its compaction. Prismatic samples of approximately 150×100×60 mm, see Figure 4(a), were cut from 306×306×60 mm asphalt slabs compacted by using a roller slab compactor to reach the design for air voids, see Table 1. Likewise, to facilitate the creation of a single crack surface on the prismatic samples during crack-healing tests, a transverse notch of 5×5 mm was made at the mid-point on the bottom surface of the beams.

Additionally, cylindrical test specimens, with 100 mm diameter and approximately 50 mm height, were compacted by using a gyratory compactor, with an inclination angle of 2.0º and 650 kPa of static pressure. A maximum of 250 gyrations were applied during compaction.

Furthermore, some semi-circular samples were cut from the cylindrical specimens. On the semi-circular samples, a notch of 5×5 mm was cut at the midpoint in the direction of the loading from the central axis of the sample.

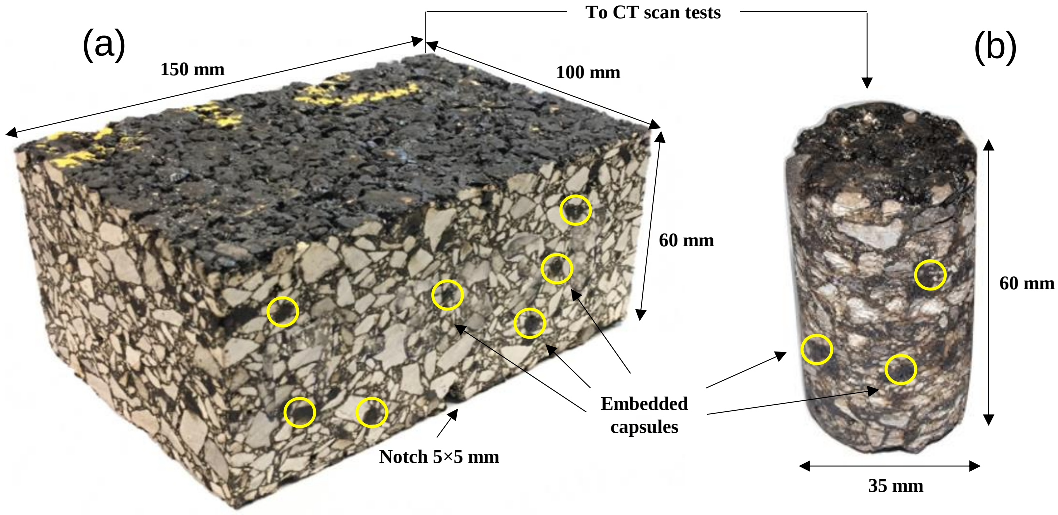

Figure 4. (a) Asphalt beam used in the crack-healing tests, and (b) asphalt core cylinder used in the CT scan tests.

3.4 Programme to evaluate the effect of mixing methodology on the mechanical properties of asphalt containing capsules.

Figure 5 describes the experimental programme. This section of the research was done in dense asphalt mixture. Two different experimental stages were defined depending on their objective, as follows:

- Stage 1: to evaluate the effect of the mixing order and the ageing times on the mechanical and self-healing properties of asphalt mixtures. In this stage, all asphalt mixture types described in Section 2.2 were evaluated using tests of stiffness modulus at 20ºC, flexural strength and crack-healing. In the flexural strength and crack-healing tests the asphalt mixture samples were tested under different ageing times, see Figure 5.

- Stage 2: to evaluate the effect of compaction waiting time and ageing process on the mechanical stability of asphalt mixtures. The waiting time of un-compacted asphalt mixture under in-situ conditions was simulated by pre-conditioning all asphalt mixture types described in section 2.2, as follows: first, batches of un-compacted asphalt mixtures were stored in an oven at 140ºC and 160ºC for 4 h. Then, asphalt mixtures were compacted in asphalt cylinders following the process described in section 2.2. In this stage, pre-conditioned asphalt mixtures at 140ºC and 160ºC were evaluated using tests of stiffness modulus at 20ºC and flexural strength under different ageing times, see Figure 5.

Finally, the ageing process on the asphalt test samples was simulated in laboratory conditions by using an oven according to Garcia et al. [13]. For flexural strength tests, semi-circular asphalt samples were placed in an oven at 85ºC for 9 different ageing times: 2, 24, 48, 72, 96, 144, 180, 210 and 240 h. While for crack-healing tests, asphalt beam samples were placed in an oven for a single ageing time of 240 h at 85ºC.

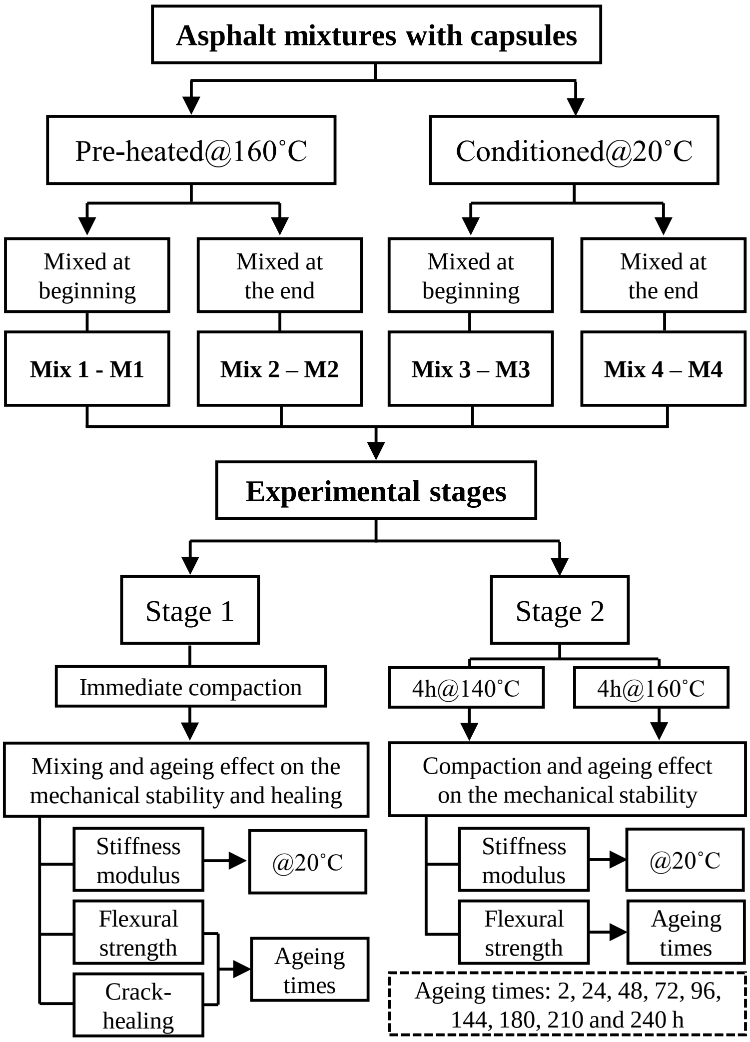

Figure 5. Diagram of the experimental programme followed in this study.

3.5 Physical and mechanical characterisation of the capsules.

The effect of the temperature on the density of the polymeric capsules was measured by non-invasive tests. The tests were performed at temperatures of 20ºC, 140ºC and 160ºC by using a Helium Pycnometer AccuPyc II 1340.

Before testing, capsules were pre-conditioned in an oven for 4 h. The mass of capsules in each sample test was 5 g. The test temperature values were selected with the aim of evaluating the physical behaviour of capsules under environmental, and mixing conditions, respectively.

Additionally, the mechanical strength of capsules was measured through uniaxial compression tests on a total of 20 capsules for each temperature value. Capsules were loaded until failure at a loading rate of 0.2 mm/min, see Figure 6(a). The tests were performed at 20ºC, 140ºC and 160ºC, using an Instron Model 5969 with a 5 kN load cell and an environmental chamber with temperature control. Capsules were pre-conditioned in the chamber for 4 h before each test. Figure 6(b) shows an example of the type of failure that an individual broken capsule presented after test at 20ºC.

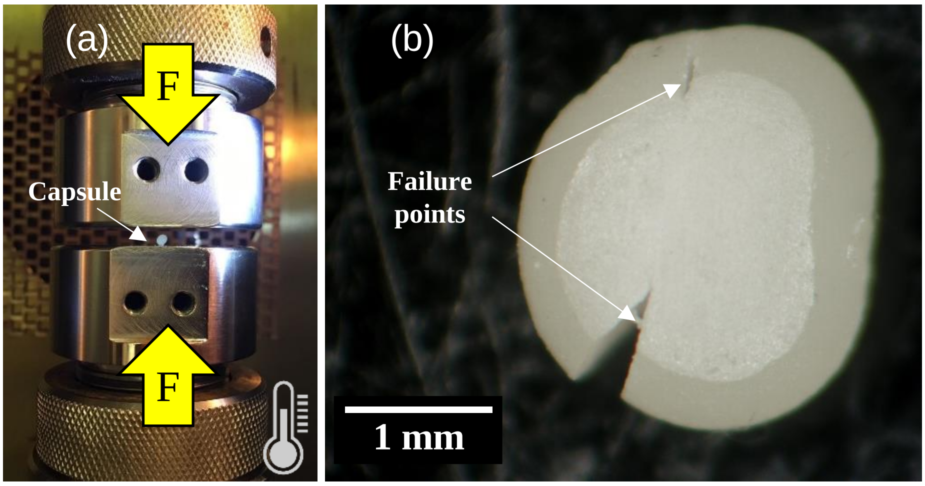

Figure 6. (a) Compressive strength test of capsules, and (b) example of a broken capsule after test at 20˚C.

The main properties of the used polymeric capsules are presented in Table 2.

Table 1. Main properties of the calcium-alginate capsules.

Property	Unit	Value
Diameter	mm	2.5
Rejuvenator content	$\%_{vol}$	75
Polymer content	$\%_{vol}$	25
Bulk density	g/cm^3	1.116
Thermal expansion coefficient	µm/m.ºC	187.10
Mass loss with temperature at:		
160ºC	%	4.2
200ºC	%	6.3
Compressive strength at:		
20ºC	N	45.5
160ºC	N	19.4

3.6 Thermal characterisation of the capsules and their components.

The thermal stability of capsules and their components were conducted by means of Thermogravimetric Analysis, TGA. The test was performed on a NETZSCH, TG 449 F3 Jupiter Thermo-Microbalance, using nitrogen atmosphere and a heating rate of

10ºC/min. Thermogravimetric profiles of mass loss in the temperature range of 0-500°C were recorded.

Additionally, thermal expansion measurements of capsules in the range from 20ºC to 200ºC were developed using a Thermomechanical Analyser (TMA) equipment model Q400. Thermal expansion tests were applied on individual capsule samples to measure the dimension change of the capsule as it is heated, while simultaneously subjected to mechanical loading. The tests were conducted at a heating rate of 5ºC/min.

3.7 Quantification of the oil released from the capsules during asphalt manufacturing.

To quantify the oil release from the capsules into the asphalt mixture beams, several asphalt samples were chemically analysed by using Fourier Transform Infrared Spectroscopy (FTIR) spectrum model Bruker Tensor 27. Samples were randomly taken from the asphalt beams with, and without, capsules after healing tests and were tested by using FTIR spectroscopy tests. Furthermore, samples of virgin and aged bitumen with the same amount of oil contained in the capsules were prepared to simulate the same amount of oil in asphalt if all the capsules had released their content. The oil-bitumen samples group were aged in an oven at 85ºC for 240 h. FTIR tests on the oil-bitumen samples with, and without, ageing process were set in the absorption mode, in the wavenumber band range of 400 to 4000 cm^{-1} at a resolution of 4 cm^{-1}. The oil effect on the bitumen was evaluated from changes in the absorbance peak between the wavenumbers 1700 to 1800 cm^{-1}. This range was adopted based on the methodology proposed by Micaelo et al. [12] and Al-Mansoori et al. [13] to determine the amount of oil released in the asphalt mixture with capsules. These works are based on the fact that vegetable oils have a distinct peak at ~1745 cm^{-1} (C-O stretch), while the bitumen has zero index in this range [25].

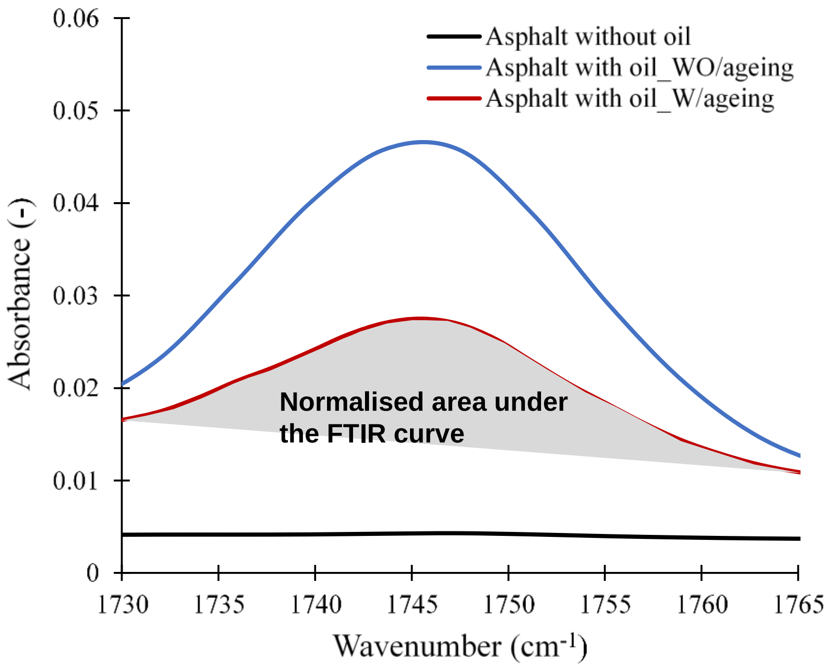

Figure 7. FTIR curves for asphalt mixture samples with and without oil.

Thus, FTIR absorbance spectrum was normalised in the selected wave length range and the absorbance area under the spectrum was measured using the trapezoidal rule of numerical integration [26]. Figure 7 shows an example of the results of FTIR absorbance spectrums for asphalt samples with, and without, oil and an explanation of the absorbance area. Finally, the percentage of broken capsules, *BC*, inside the asphalt beams after healing tests was defined as follows:

$$BC\ (\%) = \left[\frac{A_{1745-Mi}}{A_{1745-Ref}}\right]_j \times 100 \qquad (9)$$

where, $A_{1745-Mi}$ is the absorbance area at the peak 1745 cm^{-1} for the different types of asphalt with capsules (M1-M4), and $A_{1745-Ref}$ is the absorbance area at the peak 1745 cm^{-1} for the asphalt samples with the 100% of capsules broken, and where j indicates the state of the asphalt beams, i.e., with or without the ageing process.

3.8 Measurement of viscosity and ageing index.

To evaluate the variation of the physical properties of the bitumen and sunflower oil used in the asphalt mixtures and capsules manufacturing, viscosity measurements were performed on test samples of bitumen and oil at different ageing times according to Section 2.3. The bitumen viscosity test was performed by means of a Dynamic Shear Rheometer (DSR) supplied by Malvern Instruments Ltd. Before tests, bitumen samples were pre-conditioned in an oven at 85ºC for 9 different ageing times: 2, 24, 48, 72, 96, 144, 180, 210 and 240 h. The aged bitumen samples were tested at 20ºC with an oscillatory sweep frequency of 0.1-10 Hz. A parallel-plate geometry of 8 mm diameter was used with a gap of 2 mm between the spindle and the sample to allow the shearing movement as required by the standards. Moreover, the viscosity of bitumen was also measured on the bitumen samples pre-conditioned in an oven at 140 and 160ºC for 4 h.

Furthermore, the viscosity of sunflower oil was measured under the same ageing conditions of bitumen samples but using a different test. With this purpose, a simple laboratory tool using a Brookfield Rotational Viscometer at low shear rate was used. Samples of 10 g of each aged oil were transferred into separate Brookfield viscometer tubes. After that, a rotational spindle (SC4-27) was lowered into the oil tube and rotated at a constant speed of 200 rpm. Viscosity was recorded after reaching the constant value at 200 rpm. The test temperature was of 20ºC in all the measurements.

Likewise, based on the viscosity results of the bitumen and sunflower oil, an ageing index (AI) was defined as follows:

$$AI\,(\%) = \frac{\eta_{unaged} - \eta_{t-aged}}{\eta_{unaged}} \times 100 \qquad (1)$$

where, η_{unaged} is the viscosity of the bitumen or sunflower oil at 20ºC (Pa·s) without ageing process, and η_{t-aged} is the viscosity of the bitumen or sunflower oil measured at 20ºC (Pa·s) for the different ageing times (h).

3.9 Bulk density and air void content of asphalt specimens.

The maximum density of asphalt mixture samples was determined through the BS EN 12697-5 [18] by the mathematical method. In addition, the bulk densities of the test specimens were determined through the BS EN 12697-6 [19] by measuring the bulk density-saturated surface dry (SSD method). Furthermore, the air void content of each test specimen was calculated based on the previous calculation of the maximum and bulk densities, as follows:

$$AVC(\%) = \frac{\rho_{m-}\rho_b}{\rho_m} \times 100 \tag{2}$$

where ρ_m is the theoretical maximum density of the mixture without voids in g/cm^3, ρ_b is the bulk density in the mixture in g/m^3, and AVC is the air void content in the mixture, in %.

3.10 Indirect tensile stiffness modulus (ITSM) measurements.

The stiffness of the asphalt mixture specimens was measured by means of the indirect tensile stiffness modulus test. Stiffness modulus was measured at 20ºC following the protocol defined in the BS EN 12697-26 [20]. This methodology consists of doing an indirect tensile test on cylindrical specimens, applying sinusoidal loading pulses and resting periods to reach an established horizontal strain. Ten loading pulses were vertically applied on two orthogonal diameters. Stiffness modulus of each test specimen at the test temperature was taken as the average of the values measured over two perpendicular diameters obtained applying the following equation:

$$S_M = \frac{F \cdot (\upsilon + 0.27)}{(z \cdot h)} \tag{3}$$

where, S_M is the measured Stiffness Modulus in MPa, F is the maximum vertical load applied in N, z is the horizontal strain amplitude in mm, h is the average thickness of the specimen in mm, and ν is the Poisson's ratio of 0.35.

3.11 Assessment of the flexural strength of asphalt test samples.

Flexural strength of asphalt mixture samples was measured by means of semi-circular bending tests. Flexural tests were carried out at 20ºC on semi-circular test samples with 10 cm of diameter and 2.5 cm of thickness cut from cylindrical specimens. Before testing, samples were kept in the temperature-controlled chamber at the test temperature for 2 h, to prevent any temperature gradient within them. The test setup for the semi-circular tests consisted of two support rollers at the straight (bottom) edge and one loading roller at the mid-point of the semi-circular arch. The spacing between the two support rollers was 80 mm. The equipment used for testing was a servo-hydraulic Instron with capacity of 10 kN and environmental chamber.

The test load was of 1 kN applied upon sample at a loading rate of 5 mm/min. Finally, flexural measurements were developed on the all asphalt mixtures after applied different ageing times from 2 to 240 h, see Figure 5. In total, more than 100 semi-circular samples with, and without, capsules were tested.

3.12 Indirect Tensile Fatigue Test (ITFT)

Indirect Tensile Fatigue Tests were used to evaluate the effect of capsules on the fatigue life of asphalt specimens with, and without, capsules under a controlled stress mode according to BS DD ABF [21]. Fatigue tests were carried out at 20ºC, and maximum stress levels of 200, 250, 300, 350 and 400 kPa (σ_x^{max}) were used in the study. The test consists of applying an indirect tensile stiffness test to the specimens at each stress level before the fatigue test was started. Once the test started, loading period (1.5 s), loading effect time (0.124 s), stress level, diameter and height of the specimen were registered. The Indirect Tensile Fatigue Test was sustained until the breakage of SMA specimens. Under diametric loading by linear elastic analysis, the maximum horizontal tensile strain (ε_x^{max}) occurring at the centre of the cylindrical specimen can be defined as:

$$\varepsilon_x^{max} = \frac{\sigma_x^{max}\,(1+3\nu)}{S_m} \quad (4)$$

where, S_m is the stiffness modulus in MPa, σ_x^{max} is the maximum tensile stress at the centre of the specimen in MPa; ν is the Poisson's ratio of 0.35. In total, more than 30 cylindrical SMA specimens with, and without, capsules were tested.

3.13 Water sensitivity analysis

Based on previous studies, the rehydration of the capsules may affect the preservation of the encapsulated sunflower oil [27]. Likewise, swelling of capsules may affect the capsule/mastic bonding. The A-method defined in standard EN 12697-12 [23] was adopted to assess the effect of saturation and accelerated water conditioning of SMA mixture specimens with, and without, capsules. With this purpose, two subsets of test specimens were conditioned as follows:

(1) Dry group of SMA specimens: cylindrical SMA specimens with, and without, capsules were conditioned in a temperature controlled room at 20ºC during 72 h.

(2) Wet group of SMA specimens: cylindrical SMA specimens with, and without, capsules were conditioned in a water bath at 40ºC during 72 h. Before placing the specimens in the water bath, they were saturated in a vacuum container for 30 min, using a residual pressure of 6.7 kPa, as defined in the Standard [23].

Water sensitivity of the SMA mixtures was measured using the Indirect Tensile Strength (ITS) method of the cylindrical asphalt specimens, following the methodology defined in the EN 12697-23 [24], at the test temperatures of 20ºC and 25ºC. The Indirect Tensile Strength (ITS) in units of kPa was calculated by applying the following equation:

$$\mathrm{ITS} = \frac{2 \cdot \mathrm{F}}{\pi \cdot \mathrm{L} \cdot \mathrm{D}} \quad (6)$$

where, F is the peak value of the applied vertical load in kN, L is the mean thickness of the test specimen in m, and D is the specimen diameter in m.

Furthermore, to quantify the effect of the accelerated water conditioning on the ITS values with respect to the ITS reached for the SMA specimens tested in dry conditions, the Indirect Tensile Strength Ratio (ITSR) was determined by the following equation:

$$\mathrm{ITSR} = \frac{\mathrm{ITS_{wet}}}{\mathrm{ITS_{dry}}} \quad (7)$$

where ITS_{dry} and ITS_{wet} are the ITS values calculated for the dry and wet conditioned specimens in kPa, respectively. In total, more than 40 cylindrical SMA specimens with, and without, capsules were tested.

3.14 Skid resistance test.

It was proven that the capsules can release small amounts of rejuvenator during the mixing and compaction processes [*14*]. With the aim of evaluating the effect of the oil release on the skid resistance of the SMA slabs, dynamic pendulum test measurements on the SMA slabs with, and without, capsules were carried out following the protocol defined in the standard BS EN 13036-4 [22]. In this test, the measured values represent the friction properties of the surface given by the British Pendulum Number (BPN). The test consists of applying water on the surface of the slabs and re-wetting it every time after swinging the pendulum. Then, the pendulum is raised to a fixed height and it is released to make contact with the surface of the slab. A drag marker shows the BPN value. With increasing friction between the pendulum and the test surface, the BPN value increases. All the tests were measured at 20ºC and the BPN value was calculated as the average of five repetitions. In total, more than 10 SMA slabs with, and without, capsules were tested.

3.15 Measurement of the self-healing of a single crack.

To evaluate the effect of mixing method and ageing time on the self-healing properties of asphalt under real conditions, an experimental crack-healing methodology was used [13]. Self-healing of asphalt mixtures with, and without, capsules was quantified as the healing level reached from the flexural strength recovery of cracked asphalt mixture beam tested under a three-point bending (3PB) test after a healing (rest) time of 120 h. This time was defined based on previous results published by Al-Mansoori et al. [13], where asphalt beams reached the maximum healing levels for healing times close to 120 h. The healing test was implemented according to the following steps, see Figure 8:

- Step 1 - Crack generation: asphalt mixture beams with, and without, capsules were conditioned at -20ºC for 4 h and 3PB tests were carried out at a loading rate of 2 mm/min until the beams were broken in two pieces.

- Step 2 – Capsules' activation: after the 3PB test, a plastic membrane adaptable to the faces of the crack, was placed between the two broken pieces of the beam. Then, the two pieces were put back together and placed in a steel mould. To break the embedded capsules, a strain controlled compressive load was applied at a rate of 2 mm/min on the top surface of the beam, at 20ºC, until the vertical deformation reached 5 mm. Later, the plastic sheet was removed, and finally the two pieces of the beam were put back together into the steel mould.

- Step 3 – Healing process: to start the healing process of the broken beam, the asphalt beam in the steel mould was placed into a temperature-controlled chamber at 20ºC for 120h. Once the healing (rest) time was reached, the healed beam was removed from the chamber and steel mould, and Step 1 was repeated thus completing a damage-healing cycle.

The healing level, HL, reached for each cracked asphalt mixture beam after the healing time was defined as the relationship between the maximum load of the beam initially tested, $F_{initial}$, and the maximum load measured in the same beam after the healing process, F_{healed}:

$$HL = \frac{F_{healed}}{F_{initial}} \quad (8)$$

Finally, crack-healing asphalt measurements were developed on the all asphalt mixtures, see Figure 5, before and after applying an ageing process in an oven at 85ºC for 240 h. In total, more than 30 asphalt beam specimens with, and without, capsules were tested.

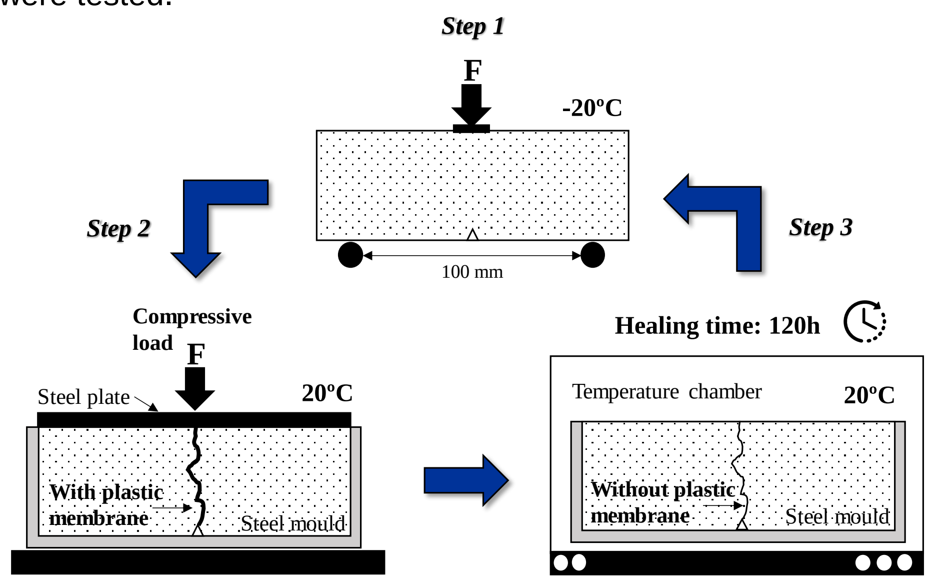

Figure 8. Schematic diagram of the asphalt mixture crack-healing test: Step 1. crack generation in the asphalt beam, Step 2. capsules activation to begin the oil release, and Step 3. healing process.

3.16 Measurement of the self-healing of fatigue damage.

The effect of capsules on the self-healing of fatigue damage was evaluated by means of 3-point bending fatigue tests over an elastic foundation. Such test configuration was selected to produce clean vertical cracks at the midpoint of the specimens, easy to assess and replicate in different samples. In order to remove the characteristic effect of permanent deformations in this type of tests, an elastic membrane was placed under the specimens.

The loading wave oscillated at a frequency of 4 Hz between a maximum value of 2.5 kN and a minimum of 0.15 kN, which ensured consistent contact between actuator and sample. A resting period of 0.15 s was included between consecutive loading pulses. Finally, the tests were carried out at the 20 ± 1ºC.

During the tests, the lateral previously white-painted side of the specimens was continuously observed using a static camera with a f/2.8 aperture and a 12 MP resolution taking a picture every 40 loading cycles. By using the free image processing software ImageJ, the total length of emerging cracks could be accurately measured in each picture until the sample split in half. This study's adopted failure criteria was the number of cycles at which the crack length reached 20% of the total crack length registered at the end of the test.

To determine the fatigue life of each studied material, 15 samples per material were subjected to the test until failure, and the Weibull Distribution was fitted to the data. Such probability distribution was selected considering that fatigue damage is produced by the progressive accumulation of mechanical energy on each applied cycle. The samples' average life time was estimated as the number of cycles whose probability of breaking the specimens is 50%. Such number of cycles will be referred hereafter as $N_{0.5}$ (Figure 1–left).

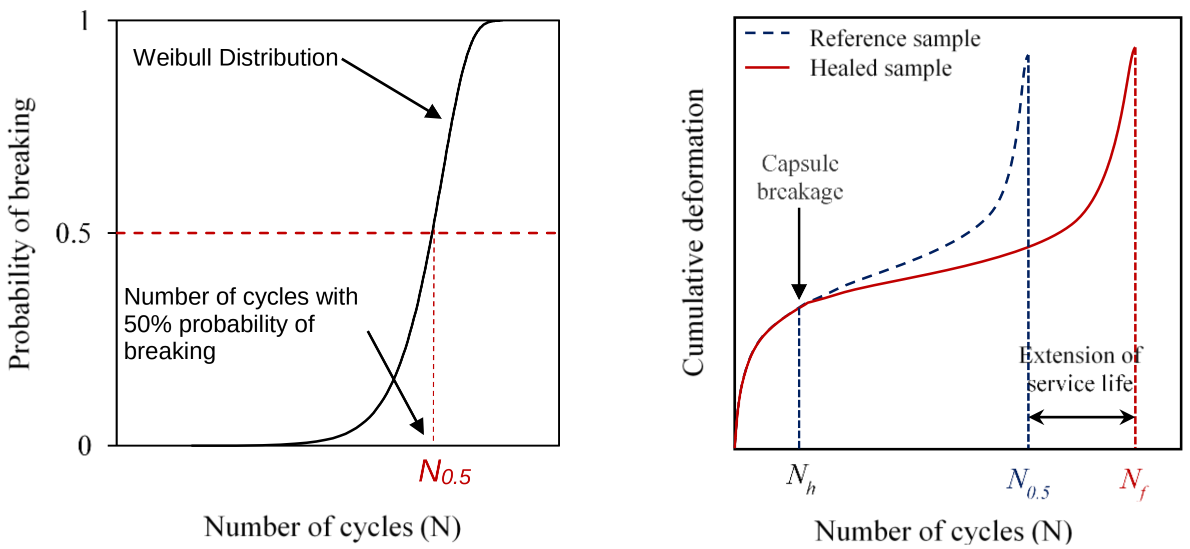

Figure 7. Scheme of how the number of cycles with 50% probability of breaking can be obtained based on Weibull probability distribution (left) and how service life extension can be evaluated (right)

In order to evaluate the capsules capacity to extend their service life of asphalt roads, a series of 40 samples was manufactured and tested, as described above. The 3PB fatigue test was implemented according to the following steps:

- **Step 1.** With the aim of determining the fatigue life of asphalt mixtures at each studied load, 12 beams for each load were manufactured.
- **Step 2.** Asphalt mixture beams with, and without, capsules were conditioned at 20ºC for 4 h before starting 3PB fatigue tests. The test continued until the sample broke in two pieces.
- **Step 3.** The average lifetime of the samples was estimated as the number of cycles whose Weibull probability of breakage is 50% ($N_{0.5}$). The cycle number at which cracks reach an approximate length of 1 cm from the beginning of the tests will be referred hereafter as $N_{0.5}$.

Besides, the healing test was implemented according to the following steps:

- **Step 1.** In order to evaluate the capacity of the asphalt mixture to extend its service life, series of 10 samples of each type of mixture were manufactured. Asphalt mixture beams with, and without, capsules were conditioned at 20ºC for 4 h and 3PB fatigue tests were carried out at different loadings (1.75, 2.75, 3.75 and 4.75 kN) until the beams were broken in two pieces.
- **Step 2.** In this case, each fatigue test was paused at different number of cycles depending on the calculated $N_{0.5}$. Then, the samples were placed into a temperature-controlled chamber at 20 ± 2ºC for 48 h. Once the healing (rest) time was reached, the healed beam was removed from the chamber and returned to the test until the failure was reached.
- **Step 3.** The extension of the service life was assessed for each sample through the concept of Healing Index (*HI*), which compares the total number of cycles resisted (N_f) and the reference ($N_{0.5}$), as follows.

$$HI = \frac{N_f - N_{0.5}}{N_{0.5}} \tag{5}$$

3.17 X-ray computed tomography.

To evaluate the capsule distribution and their integrity inside the asphalt mixtures described in Section 2.2, X-ray computed tomography (CT scan) was employed. To do that, asphalt cylindrical samples of 3.5 cm diameter and 60 mm height were extracted from asphalt mixture beams with capsules, see Figure 4(b). The X-ray tomography scans were developed using a GE Sensing and Inspection Technologies GMBH Phoenix VTomeX M operated at 200 kV and 180 µA. Thus, each tested sample was mounted on a rotational table at distance of 163.74 mm (FOD) from the X-ray source and the distance between the X-ray source and the X-ray detector was 818.698 mm (FDD). A spatial resolution of 40 µm was achieved. Scan settings were

200 ms per radiograph capture with 2520 projections captured on a 360º rotation using an average of four images and skip of one. Detector panel sensitivity was set at 2. Reconstruction of radiographs was performed using Phoenix Datos X2 Reconstruction software. Radiographs were uploaded into the software and corrections were applied for: movement using a scan optimiser algorithm; beam hardening; cone beam ROI; and air observation ROI. Once reconstructed into 3D volume 2D image slices were exported in TIFF format along the XY axis. Image processing of the capsule distribution was prepared by segmenting the materials found in a specific volume, based on simple thresholding to separate the asphalt mixture of the capsules. The software used for 3D volume and 2D image processing was VGStudio MAX version 2.2.

4. RESULTS AND DISCUSSION

4.1 Effect of temperature on the mechanical properties of capsules

Polymeric capsules with encapsulated sunflower oil were added to the asphalt mixtures to improve their self-healing properties. However, the mixing and compaction processes of the asphalt mixtures may possibly affect their physical, thermal and mechanical properties [13]. This section presents and discusses the characterisation results of the capsules.

The effect of the temperature on the density of the capsules was studied. The capsules registered average density values of 1.116 g/cm^3, 1.059 g/cm^3 and 1.021 g/cm^3, at temperatures of 20ºC, 140ºC and 160ºC, respectively. This demonstrated that the density of the polymeric capsules reduced with the increase of temperature, which was due to the loss of mass experienced by the capsules when exposed to increasing temperature, as evidenced in the TGA profiles, see results shown in Figure 9(a). It can be observed in Figure 9(a) that the capsules designed in this study reduced their mass with the increase of temperature, registering mass loss values of 3% and 4%, at 140ºC and 160ºC, respectively. This mass loss registered by the capsules at high temperatures is mainly attributed to the degradation of the calcium-alginate polymer structure and water evaporation.

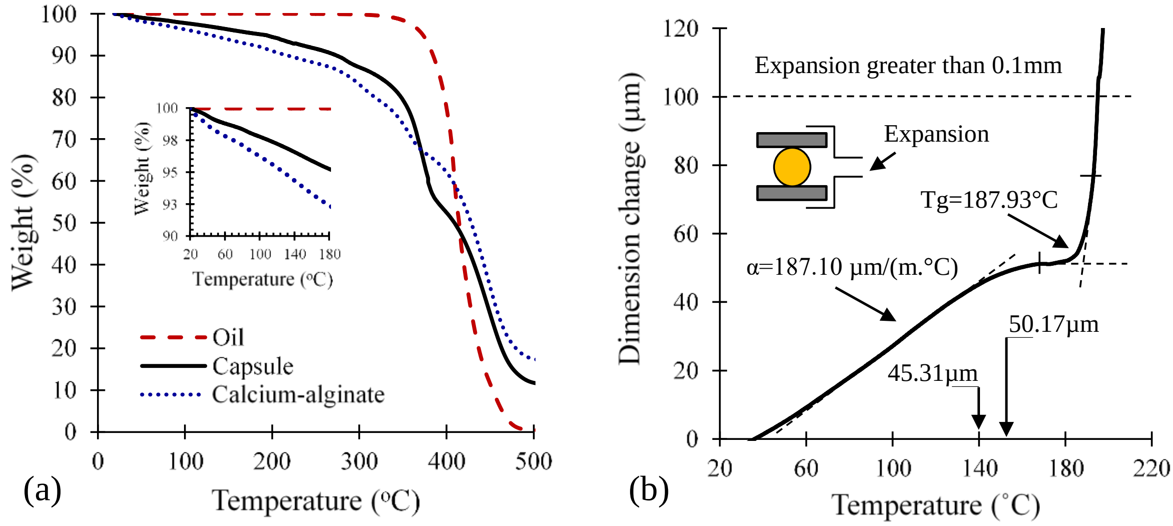

Figure 9. (a) TGA test result of a capsule and its components, and (b) thermal expansion curve of a capsule.

Furthermore, their change in colour to a darker one at high temperatures, see Figure 3(a), was due to the partial peroxidation of the encapsulated oil [28]. This chemical phenomenon occurs when the sunflower oil is exposed to high temperatures (160-200ºC), causing the hydrogen atoms to separate from the oxygen and carbon bonds. As a result, peroxides are formed in the bonds that oxidise the oil thus making it a darker colour with a more viscous texture.

Moreover, the reduction of the capsules' density with the temperature could also be due to the variations in their volume caused by thermal expansion. In this context, Figure 9(b) shows the average curve obtained from the thermal expansion test carried out on the capsules. In this Figure, it can be observed that the size of the capsules linearly increased with the temperature, from 20ºC to 170ºC, registering changes on the diameter of 45.31 µm and 50.17 µm, at mixing temperatures of 140ºC and 160ºC, respectively. Thus, the variation of the capsules' diameter at mixtures manufacturing temperatures (~140ºC) was apparently not significant.

To prove this hypothesis, the average diameter of 20 individual polymeric capsules before, and after, the mixing process was measured by taking photographs under an optical microscope. After-mixing capsules were randomly taken by hand from a blend batch of un-compacted hot mixture asphalt, see Figure 3(c). As a result, the average values of the capsules diameter before, and after, the mixing process were, respectively, 2.572 mm and 2.895 mm. Likewise, after-mixing capsules were surrounded by a bitumen membrane with an average thickness of 0.271 mm, see Figure 3(d). Therefore, the increase of the diameter of capsules after mixing was not significant (<53 µm), and can be considered as thermally stable at the mixing temperatures (140-160ºC) used to manufacture asphalt mixtures. Additionally, Figure 9(b) shows that the capsules presented a thermal expansion coefficient of 187.10 µm/m.ºC, with changes in the diameter over 100 µm at temperatures over 200ºC, which can be translated into capsules' mass loss close to 10%, see Figure 9(a).

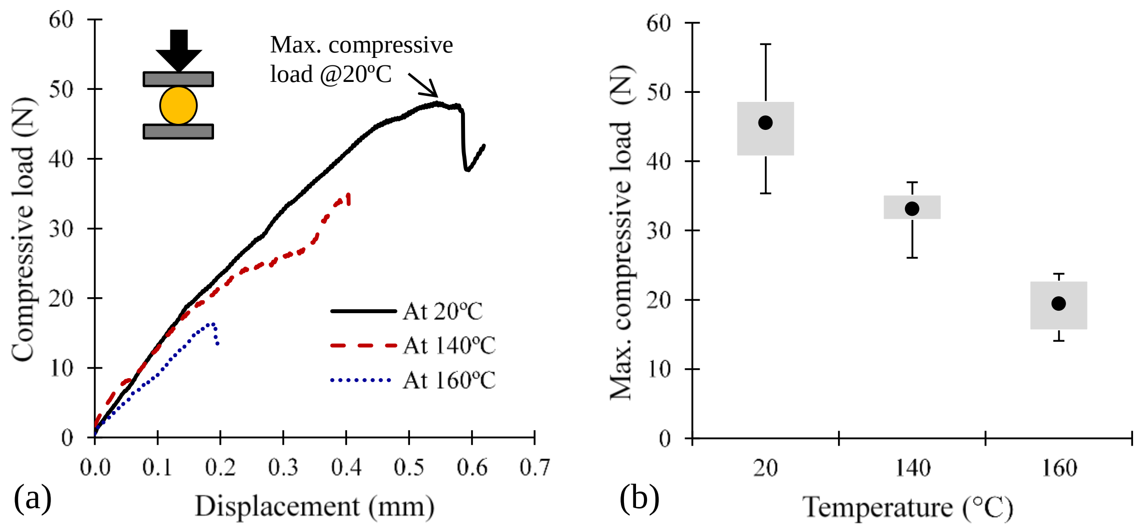

Figure 10. (a) Mechanical strength, and (b) maximum compressive load resisted by the capsules.

On the other hand, Figure 10 shows the results of compressive strength of the polymeric capsules tested at 20ºC, 140ºC and 160ºC. The stress-strain curves of the compressive tests showed a ductile behaviour where the peak load was reached at deformation levels between 0.6 mm and 0.2 mm at, respectively, 20ºC and 160ºC, see Figure 10(a). This result proves that the capsules reduced their strength and strain with the increase of temperature. This was because the exposure to high temperatures degrades the calcium-alginate polymeric structure of the capsules making them more brittle and hence less flexible to mechanical loads. Similar results were also recently published by Micaelo et al. [12].

Otherwise, Garcia et al. [11] in their study about characterisation of capsules containing rejuvenators had shown that the minimum compressive strength of capsules required to resist the mixing and compaction processes was 10 N. Consequently, the measured compressive strength of the polymeric capsules at 20ºC, 140ºC and 160ºC, was 46 N, 33 N, and 19 N, respectively (see Figure 10(b)), which proved that the designed capsules can resist the mixing and compaction processes. Likewise, compressive strength reached by calcium-alginate capsules at low and high temperatures, was two times greater than that of the capsules presented by Al-Mansoori et al. [13]. This proved that the addition of a higher amount of sodium alginate to the capsules' composition helped to increase their mechanical strength, because the calcium-alginate capsules present a denser internal structure, see SEM image in Figure 2(b).

4.2 Influence of capsules on the bulk density and air voids content of asphalt mixtures

Figure 11 shows the average results of the bulk density and air voids content of the asphalt mixtures with, and without, capsules evaluated after different mixing and ageing conditions. In Figure 11(a), it can be observed that the average density values of the mixtures with, and without, capsules evaluated after the usual compaction conditions, Stage 1, were similarly regardless of the pre-conditioning temperature of the capsules and the mixing order. In general, average results of the mixtures density were similar to the bulk density of the design mixture (2.383 g/cm^3), asphalt mixtures without capsules (WO/C) and M2-mixture with capsules being those which registered the highest and lowest densities, with average values of 2.420 g/cm^3 and 2.404 g/cm^3, respectively. Additionally, it was observed that M4-mixture with capsules added at the end of the mixing process at 20ºC, presented the average bulk density most similar to that of the reference mixture, which was 2.416 g/cm^3.

Moreover, Figure 11(a) shows that the average density of asphalt mixtures evaluated according to Stage 2 was lower than that of the mixtures evaluated according to Stage 1. In general, mixtures according to Stage 2 presented higher average density values at 140ºC than at 160ºC. It can be deduced from the results that the mixtures without capsules (WO/C) presented the highest reduction of density with the temperature, registering values of 2.363 g/cm^3 and 2.337 g/cm^3 at 140ºC and 160ºC, respectively, equivalent to an average density reduction of 2.35% and 3.43% respectively, with respect to the density measured in Stage 1.

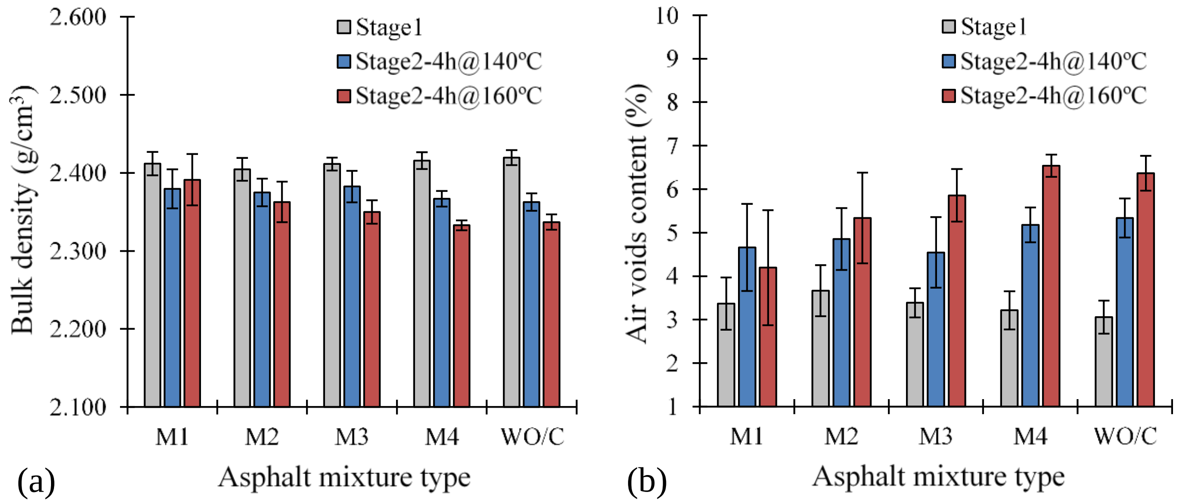

Figure 11. Results of the (a) bulk density and (b) air void content of the asphalt mixtures with and without capsules (WO/C).

In contrast, the average air voids content of mixtures evaluated according to Stage 2 at 140ºC and 160ºC was higher than that of mixtures evaluated according Stage 1, see Figure 11(b). This was due to the reduction of the mixtures density with the temperature, produced by the reduction on the mass of the capsules at the temperatures evaluated, see Figure 9(a). Another reason of the increase in the air voids content of the Stage 2-mixtures was the ageing of the bitumen caused by the ageing time, which hardened the bitumen thus making the mixture less workable. It was proven that ageing times of 4 h in the oven at 140ºC and 160ºC, resulted in ageing indices of 7.57% and 28.46%, respectively. Finally, results shown in Figure 11 demonstrated that the 4h-waiting time (ageing time) before compaction at high temperatures can affect the physical properties of mixtures with, and without, capsules and, that this phenomenon can be due to the mixing order and to the ageing damage of bitumen.

4.3 Influence of mixing and ageing time of capsules on the mechanical properties of asphalt mixtures

The results of the stiffness modulus depending on the asphalt mixtures evaluated and air voids content can be observed in Figure 12(a) and Figure 12(b), respectively. Figure 12(a) shows that depending on the mixing order (i.e., mixture type) and the ageing temperature (4 h in oven at 140ºC or 160ºC), the stiffness modulus was different. In the case of mixtures evaluated according to Stage 1 conditions, the average values of stiffness modulus of mixtures with capsules were lower than that of mixtures without capsules (WO/C). Thus, the reference mixture without capsules presented the highest stiffness modulus, with an average value of 7880 MPa, followed by the mixtures with capsules of types M4, M1, M2 and M3, with average values of 6423 MPa, 5946 MPa, 5861 MPa, and 5474 MPa, respectively. This result demonstrated that the mixing order and the pre-conditioning temperature of the capsules had an influence on the stiffness modulus values. An increase in the porosity was another factor that could have had an influence on the reduction of the modulus in the asphalt mixtures with capsules evaluated according to Stage1. In Figure 12(b), it can be observed that, regardless of the evaluation state, Stage 1 or

Stage 2, the stiffness modulus of mixtures with capsules decreased with the increase of the air voids content.

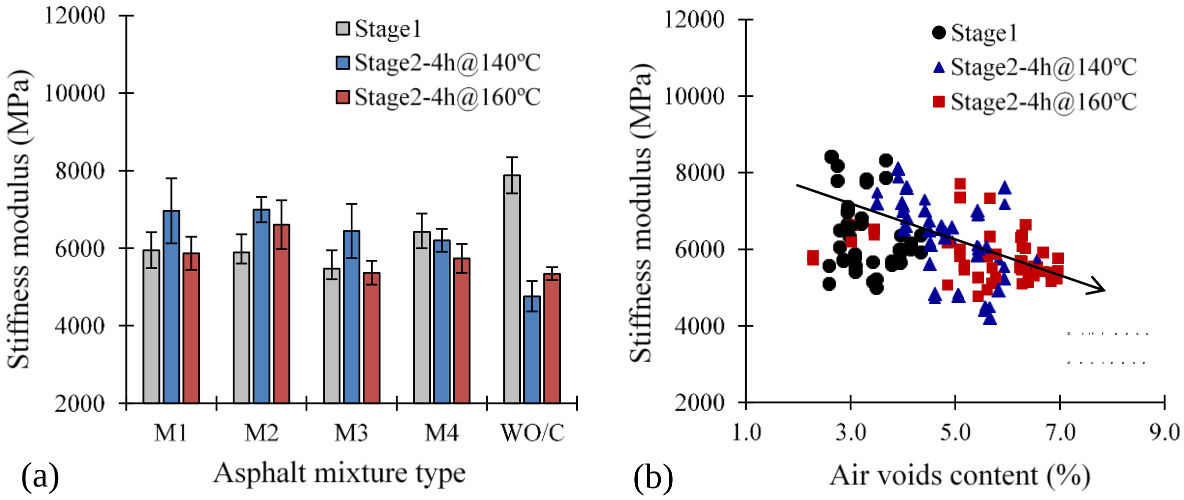

Figure 12. (a) Results of stiffness modulus for asphalt mixture samples with and without capsules (WO/C), and (b) stiffness modulus results of all test specimens versus air voids content.

Furthermore, it can be seen in Figure 12(a) that the mixtures evaluated according to Stage 2 at 140ºC and 160ºC, presented stiffness modulus values different to that of mixtures evaluated according to Stage 1 conditions. These modulus values were different depending on the mixture type evaluated. Therefore, mixture types M1, M2 and M3, presented stiffness modulus higher than in Stage 1, with values 6964 MPa, 6993 MPa and 6445 MPa, respectively. However, M4-mixture presented an average stiffness modulus lower than that obtained according to Stage 1. The same behaviour was observed in the mixture without capsules, registering average values lower than those of Stage 1, such as: 4759 MPa and 5342 MPa at 140ºC and 160ºC, respectively.

Additionally, in Figure 12(a) it can be observed that mixtures with capsules, evaluated according to Stage 2 conditions at 160ºC, presented lower values of stiffness modulus than at 140ºC, this difference being larger or smaller depending on the mixing order. For example, in the case of M3-mixture, where the capsules were mixed with the aggregates at 20ºC at the beginning of the mixing process, the stiffness modulus presented a higher reduction compared with M4-mixture, where the capsules were mixed with the asphalt mixture, at 20ºC, but at the end of the mixing process, see Figure 12(a). As in the case of mixtures evaluated according to Stage 1 condition, the reduction of stiffness in mixtures evaluated according to Stage 2 at 140ºC and 160ºC, can be attributed to the increase of the air void content, Figure 12(b), and the effect of the capsule distribution inside the tested specimens.

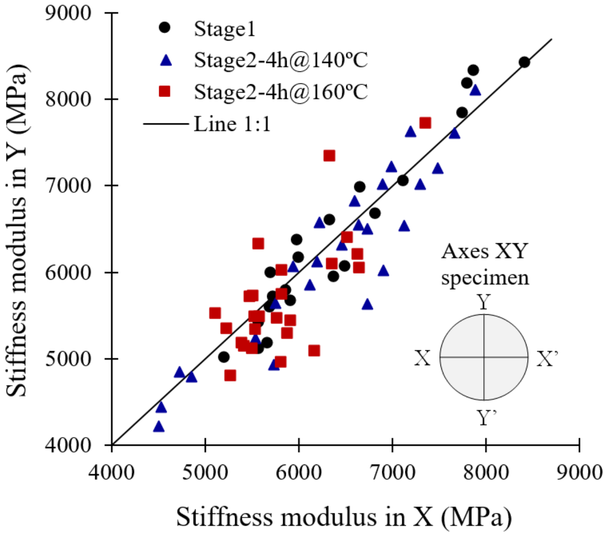

Figure 13. Relationship between stiffness modulus of the test specimens measured in longitudinal (Y) and cross (X) directions.

With the aim of evaluating the influence of the capsule distribution on the stiffness of mixtures, Figure 13 shows the relationship between all the stiffness modulus values on the test specimens in the longitudinal (Y-Y') and cross (X-X') directions. In this Figure, it can be observed that regardless of the mixing order, Stage, and temperature evaluated, the stiffness modulus values were very similar in both directions (X and Y), for most of the asphalt specimens tested. This proved that asphalt mixtures with capsules did not present an anisotropic mechanical behaviour for the plane orthogonal to the coaxial axis of the specimens, which can directly affect the average stiffness modulus.

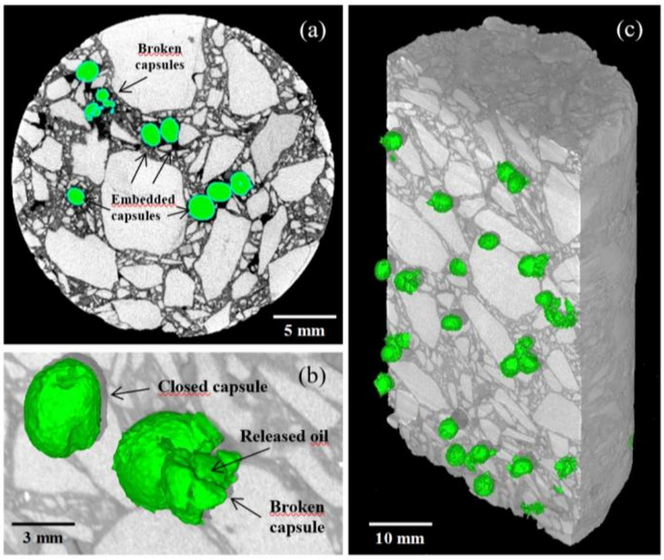

Figure 14. CT-Scans reconstructions of the asphalt mixture M4 with capsules highlighted in green colour: (a) 2D cross-section image of sample with capsules, (b) 3D reconstruction of the embedded capsules in asphalt, and (c) 3D reconstruction of the capsule spatial distribution inside of the asphalt sample.

Additionally, previous results can also indirectly prove that the capsule distribution inside the asphalt mixtures can be considered as uniform, not presenting a negative influence on the stiffness modulus value. This result can be verified by observing the CT-Scans reconstructions shown in Figure 14. In this Figure, it is shown that the capsules presented a good distribution inside the mixture, with some damaged capsules due to the mixing and compaction processes. Overall, it can be concluded that capsules added in a percentage of 0.5 did not improve the stiffness modulus of asphalt mixtures respect to the mixtures without capsules.

Furthermore, Figure 15 shows the results of the maximum force resisted by the semi-circular bending samples evaluated at different ageing times. It can be observed that 1) flexural strength slightly increased with the ageing time, and 2) mixtures without capsules (WO/C) resisted, in general, higher flexural forces than mixtures with capsules (M1-M4). This result can be attributed to the hardening of the bitumen because of ageing, which increased the stiffness of asphalt mixtures. In fact, Figure 16(a) shows how the maximum flexural force resisted by asphalt samples without capsules increases with the viscosity of bitumen because of the ageing time. Additionally, comparing the results for the mixtures evaluated according to Stage 2 at 140ºC and 160ºC, see Figure 15(a) and (b), it can be observed that these results were similar, regardless of the experimental Stage evaluated. In this way, it can be concluded that the addition of capsules did not improve the flexural strength of asphalt mixtures for the different ageing times.

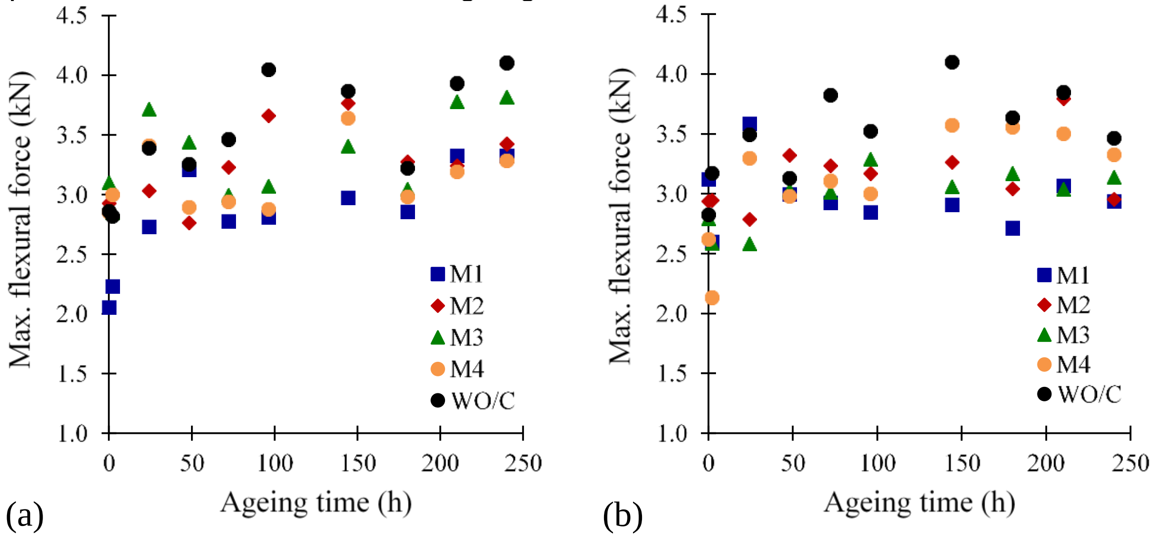

Figure 15. Variation of the maximum flexural forces versus ageing times for all the asphalt test samples measured under (a) stage 2@140ºC, and (b) stage 2@160ºC.

In order to prove that the capsules did not have a significant influence on the flexural strength of asphalt mixtures, a ratio between the maximum flexural force of all the test specimens with capsules and that of the mixture without capsules (WO/C) tested at the same ageing time was calculated, regardless of the mixing order and ageing time. For reasons of simplicity, these data have not been presented in the paper. Nevertheless, to prove that the difference between them is due to the scatter (see dispersion of results in Figure 15), the Normal Probability of these values has been calculated and presented against the data percentiles in Figure 16(b). Results showed that all data can be aligned in a straight line with a 1:1 slope, regardless of the experimental Stage evaluated. This means that the differences in the flexural strength between tests specimens with, and without, capsules were due to statistical variations, and that the mixing order and ageing time did not have a significant influence on the flexural strength reached by the mixtures.

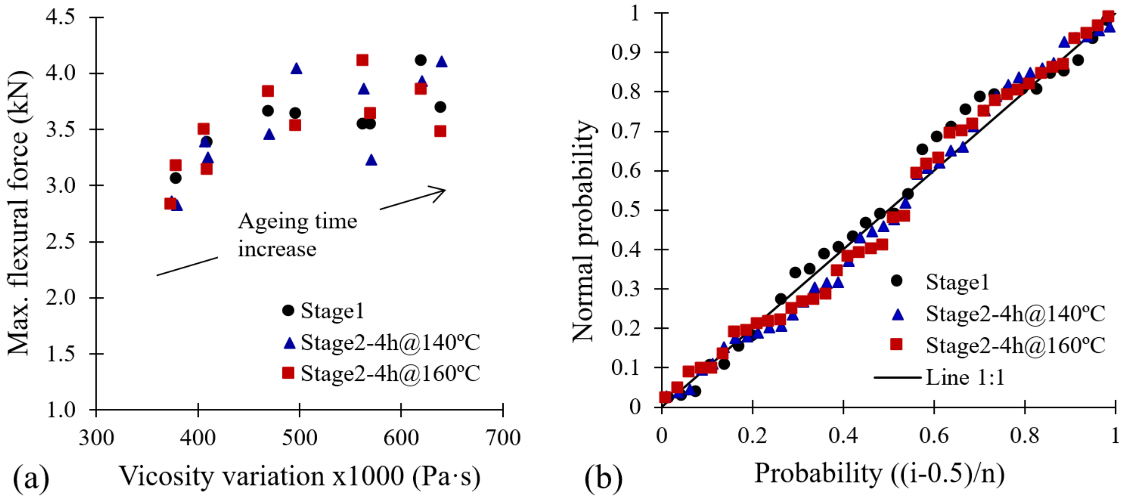

Figure 16. (a) Relation between the maximum flexural force resisted by samples without capsules versus viscosity variation of bitumen with the ageing time. (b) Normal probability-probability plot of the ratio between the flexural force of the test samples with (W/C) and without (WO/C) capsules at different ageing times.

4.4 Tensile strength and water damage of SMA mixtures

Figure 17(a) shows the average results of the Indirect Tensile Strength (ITS) of the SMA specimens with, and without, capsules after dry and wet conditioning at the test temperatures, 20ºC and 25ºC. Likewise, Figure 17(b) presents the results of the relationship between the ITS values (strength ratio, ITSR) measured on the specimens under dry and wet conditions, also at the test temperatures, 20ºC and 25ºC.

In Figure 17(a) it can be observed that the Indirect Tensile Strength of the SMA specimens reduced with the increase of the temperature and that the SMA specimens evaluated in wet conditions presented lower values of tensile strength than specimens tested in dry conditions, for both test temperatures. Overall, SMA mixtures with capsules tested in dry conditions showed slightly higher average ITS values compared with the specimens without capsules, see Figure 17(a).
This increase was of 4.86% and 4.42% for test temperatures of 20ºC and 25ºC, respectively.

In addition, in contrast with specimens in dry conditions, specimens with, and without, capsules evaluated under wet conditions presented similar ITS values for both test temperatures, which shows that, under conditions of moisture damage, SMA mixtures can show a similar mechanical performance, see Figure 17(a). This result proved that the capsules manufactured in this study showed a positive influence on the tensile strength of SMA mixtures. Based on the study recently published by Micaelo et al. [12], this result can be mainly due to strong adhesion of the calcium-alginate capsules in the bituminous mastic matrix and to their good interlocking with the aggregate skeleton.

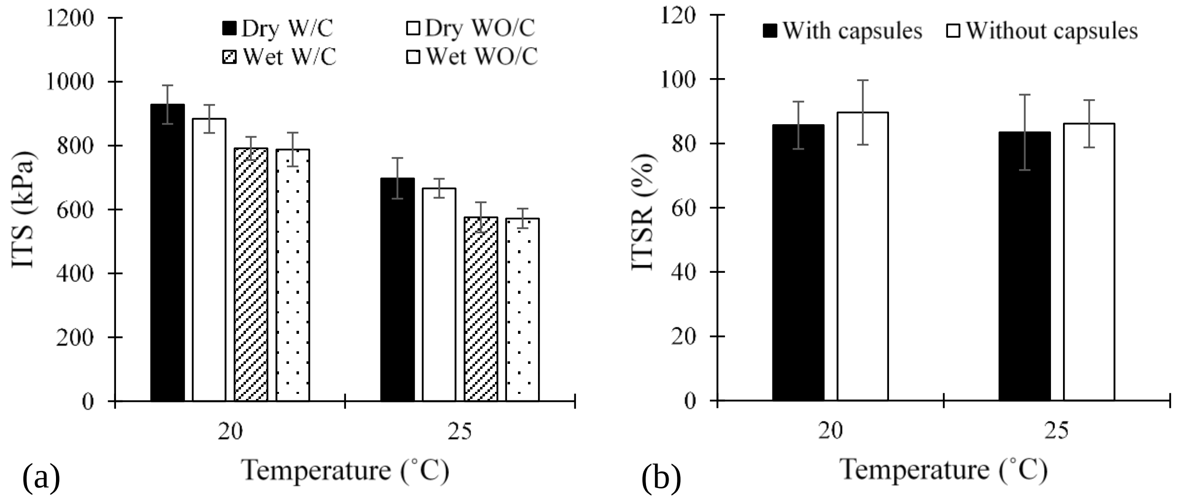

Figure 17. Average values and standard deviation bars of the: (a) Indirect Tensile Strength (ITS) of the SMA specimens with (W/C), and without (WO/C), capsules after dry and wet conditioning evaluated at two different test temperatures, and (b) Indirect Tensile Strength Ratio (ITSR).

Moreover, in Figure 17(b) it can be observed that SMA specimens with capsules presented lower average values of ITSR than the specimens without capsules. Nevertheless, based on the dispersion of the ITS and ITSR values measured in the SMA specimens with capsules (see standard deviation bars in Figure 17(a) and (b)) it can be concluded that the capsules did not significantly affect the tensile strength of the SMA mixture. Consequently, the reduction of the tensile strength of the SMA specimens was mainly associated with the increase of the test temperature and not with the environmental test conditions or with the content of capsules. This result can be better observed in the values of stiffness modulus measured by indirect tensile tests at different temperatures.

4.5 Stiffness modulus and resistance to fatigue of SMA mixtures

The stiffness modulus results for the SMA mixtures with, and without, capsules measured at four different temperatures are presented in Figure 18 (a). It can be observed that the stiffness modulus reached by the SMA specimens decreased with the increase of the test temperature, from 5ºC to 30ºC, and that overall, the average values of stiffness modulus were similar for specimens with, and without, capsules regardless of the test temperature, see Figure 18 (a).

At low test temperatures (5°C), it can be observed that the addition of capsules positively affects the stiffness modulus of SMA mixtures with respect to the reference SMA mixtures without capsules, registering values of 9801 MPa and 9772 MPa for specimens with, and without, capsules, respectively. However, when the test temperature increased, their stiffness modulus significantly reduced, see Figure 18(a). For example, test specimens tested at 30ºC showed modulus values of 1102 MPa and 1119 MPa for mixtures with, and without, capsules, respectively, which represents a reduction of approximately 90% in the stiffness modulus. This result can be explained due to the viscoelastic behaviour of bitumen, which undergoes a loss of stiffness when the temperature increases [29], thus resulting in a decrease in the viscosity of SMA mixture and the consequent reduction of the stiffness modulus.

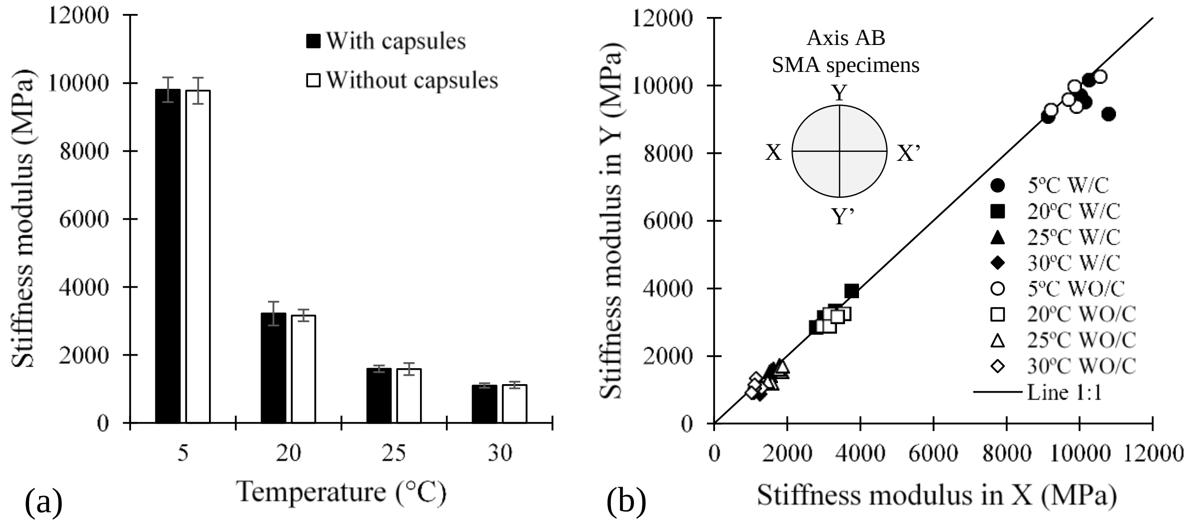

Figure 18. Average values and standard deviation bars of the: (a) stiffness modulus of SMA specimens with (W/C), and without (WO/C), capsules versus test temperature, and (b) relationship between stiffness modulus of the SMA specimens measured in longitudinal (Y) and cross (X) directions.

Moreover, Figure 18(b) shows the relationship between all the stiffness modulus values measured by the indirect tensile test on the SMA specimens in the longitudinal (A-A') and cross (B-B') directions. It can be observed that, independent of the type of mixture tested, with or without capsules, and of the test temperature, the stiffness modulus measured in both directions was very similar, see line 1:1 in Figure 18(b). This result proved that SMA mixtures with 0.5% of capsules did not present an anisotropic mechanical behaviour in the orthogonal plane to the coaxial axis of the specimens, which can directly affect the average stiffness modulus. This result also proved that capsules did not negatively affect the stiffness of SMA mixtures, with respect to the mixtures without capsules.

Furthermore, the number of cycles until fatigue failure of SMA mixtures with, and without, capsules is presented in Figure 19 for the different tensile stresses applied on the specimens, from 200 to 400 kPa. In addition, the number of cycles to failure shown in Figure 19 have been plotted versus the initial maximum horizontal tensile strain obtained by using the Equation (4) from section 2.12. In this Figure, it can be observed that the number of cycles to failure decreased with the increase of the stress level applied, and that the maximum horizontal tensile strain of the specimens increased with the increase of the stress level applied, see Figure 19. These results were similar regardless of the type of mixture tested (with, or without, capsules), which proved that the addition of capsules in the SMA mixtures tested did not have an influence on the fatigue life of the specimens. Besides, the linear trend of the results, shown in Figure 19, suggest that the capsules did not produce a decrease of the fatigue resistance of SMA mixtures, and that the similarity of results of number of cycles to failure obtained by the specimens with, and without, capsules was mainly due to the equivalence of values between the stiffness modulus reached by the SMA specimens with, and without capsules, see Figure 18.

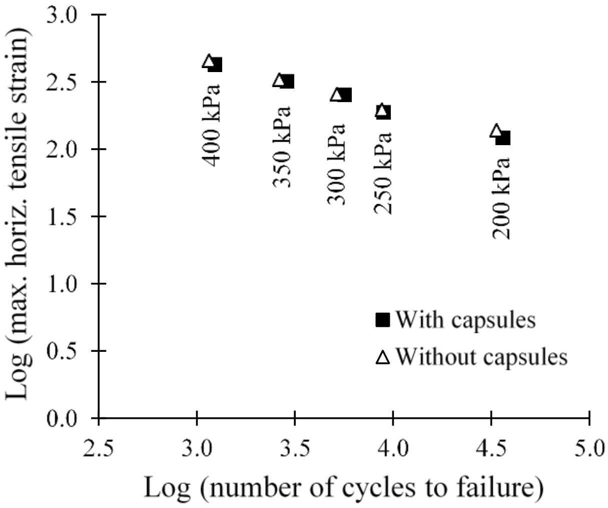

Figure 19. Maximum horizontal tensile strain average results versus the number of cycles to failure reached by the SMA specimens with, and without, capsules measured by indirect tensile fatigue tests.

Moreover, the results shown in Figure 18(b) and Figure 19 can also indirectly prove that the embedded capsules inside the SMA mixtures can be considered as uniformly distributed not presenting a negative influence on the stiffness modulus values and fatigue resistance of the SMA mixtures. This result can be verified observing the CT-Scan reconstruction results in Figure 14. In this Figure, it is shown that the capsules presented a good spatial distribution inside the SMA specimens, with some damaged capsules due to the mixing and compaction processes. Additionally, it was found that the physical properties of SMA mixtures with, or without, capsules were very similar registering non-significant differences for the bulk density and air void content values, see Table 2. Hence, this result proved that the capsules did not affect the volumetric properties of SMA mixtures.

Table 2. Physical properties of the SMA mixtures with, and without, capsules: Average (Std. deviation) values.

Property	Unit	With capsules	Without capsules
Bulk density	g/cm^3	2.229 (0.011)	2.230 (0.011)
Air void content	%	5.969 (0.453)	5.899 (0.480)
Skid resistance	BPN	48.1 (1.893)	51.5 (2.334)

4.6 Influence of capsules content on the healing properties of single cracks

Figure 20 presents self-healing results of asphalt beams containing different percentages of capsules (0.1%, 0.25% and 0.50% by total weight of the mixture) and tested at a healing temperature of 20ºC. Figure 20(a) shows the healing levels results reached for all the asphalt mixture beams, with, and without, capsules, at different healing times ranging from 6 to 192 h. In this figure, it can be observed that the healing levels in the asphalt beams with capsules were higher than in beams without capsules and that the healing level of asphalt mixtures with and without capsules increased with the healing time until a maximum value, and then remained constant. Based on the results, maximum healing level value was reached, which is, the value that healing level stops increasing until the end of the test. In general, the healing level at 96h is the highest value that can be reached and remained mostly constant until 192 h, see the box in Figure 20(a).

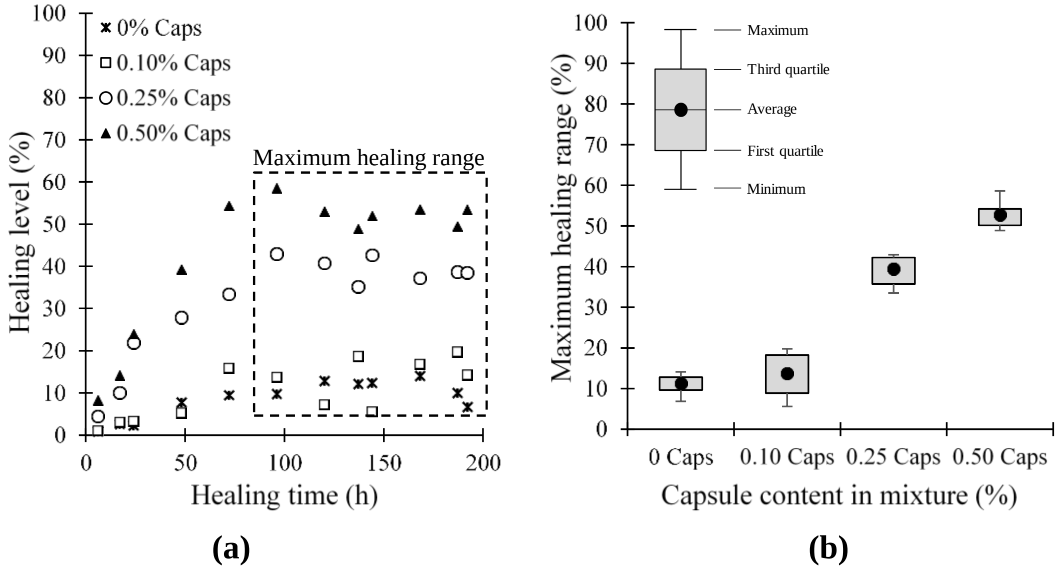

Figure 20. Self-healing results for asphalt mixture samples with and without capsules: (a) healing level depending on healing time, and (b) box plots of the maximum healing range (96-196h) with capsule content in the mixture.

The healing level curve start to be constant after a certain healing time, namely after 96h until the end of the test. The average healing level obtained from these healing times is called the maximum healing range. Results reached in the maximum healing level are shown in box plots in Figure 20(b): 13.75%, 39.44% and 52.72% for asphalt mixtures with capsule content of 0.10%, 0.25% and 0.50%, respectively. Likewise, asphalt mixtures without capsules (WO/C) presented an average maximum healing level of 11.22%. These results prove that a higher capsule content in the mixture resulted in higher healing levels for all the healing times studied. This conclusion is reasonable due to the fact that a higher capsule content increases the probability of breaking more capsules when the mixture is subjected to external damage, which increases the potential released oil content in the mixture. To prove this, Figure 21 presents the results for the quantity of broken capsules as a percentage of the total capsules used for asphalt mixtures with different capsule content and tested at healing temperature of 20ºC. The results of broken capsules were represented before, and after, the external compression load applied to the samples in the laboratory as described in Figure 7 (Step B). From Figure 21 it can be observed that asphalt mixtures with the highest capsule content, 0.50%, presents the highest percentage of broken capsules after the compression loading of 56.31% of the total amount of the capsules in the mixture, compared to 49.20% and 31.29% of mixtures with capsules contents of 0.25% and 0.10%, respectively.

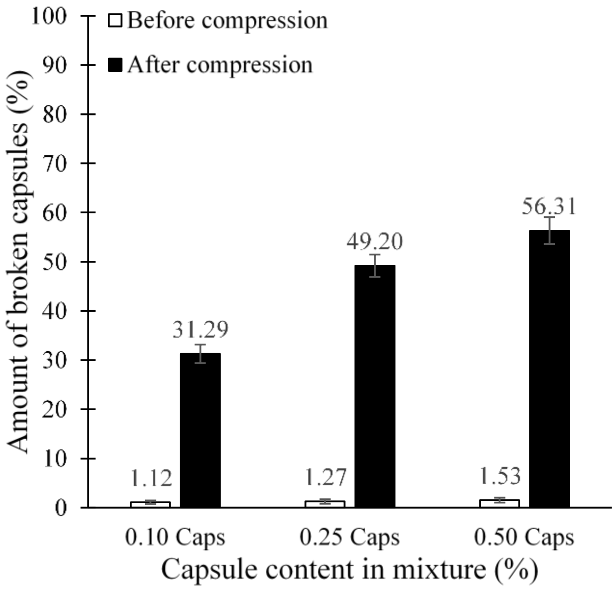

Figure 21. Amount of broken capsules for asphalt mixtures with different capsule content in mixture.

Moreover, Figure 20(b) presents the maximum healing range that can be obtained from asphalt mixtures with three different capsule contents compared to a mixture without capsules. Apparently, the healing range is dependent on the capsules content in the mixture, where a higher capsule content presents the higher healing range obtained. However, the healing range of the asphalt mixture without capsules was similar to that of mixtures with 0.10% capsule content, see Figure 20(b). In fact, these asphalt mixtures without capsules were treated with some oil during the mixing process. The amount of oil added to these mixtures was similar to the amount of oil released from damaged capsules during mixing and compaction process because of mixing temperature or compaction loads.

Figure 21 presents the quantity of broken capsules before compression, which means during mixing and compaction processes. These amounts were evaluated as 0.1 g, 0.2 g and 0.5 g from mixtures with capsule content of 0.1%, 0.25% and 0.50%, respectively. So, the reason for adding these amounts of oil to mixtures without capsules is to exclude the effect of this released oil from self-healing evaluations and keep it restricted to the oil released from capsules after applying compressive loads to damage the capsules in the mixture. The amount of oil added to the mixture without capsules in Figure 20(b) was 0.5 g, which was similar to the oil released from asphalt mixtures with 0.50% of capsule content.

4.7 Influence of temperature on the healing properties of single cracks

Figure 22 shows the results of healing level for all asphalt mixture beams without capsules (Figure 22(a)) and with capsules (Figure 22(b)), tested at eight different healing temperatures (-5, 5, 10, 15, 20, 30, 40 and 50ºC) in the healing times range from 6 to 192 h. The capsules content used in this test was 0.50% of the total weight of the asphalt mixture. Similar to Figure 20(b), the results from Figure 22 show that the healing level of asphalt mixtures increased with the time until a maximum value where it remained constant. In general, this maximum healing level was reached after approximately 96 h and remained approximately constant until the end of the test for both mixtures with, and without, capsules. Evidently, this behaviour depends on the healing temperature and the quantity of broken capsules in the asphalt beams after compressive load. Figure 23 shows the quantity of capsules broken by external compressive loads at different temperatures. In this figure, it can be observed that the stiffer the asphalt samples the fewer the broken capsules, and that the percentage of broken capsules increased with temperature as mixtures become less viscous. Therefore, in less stiff asphalt samples tested at -5ºC the broken capsules were 10.39% of the total capsules in the mixture, while in asphalt samples tested at 50ºC the percentage of broken capsules reached 60.47%.

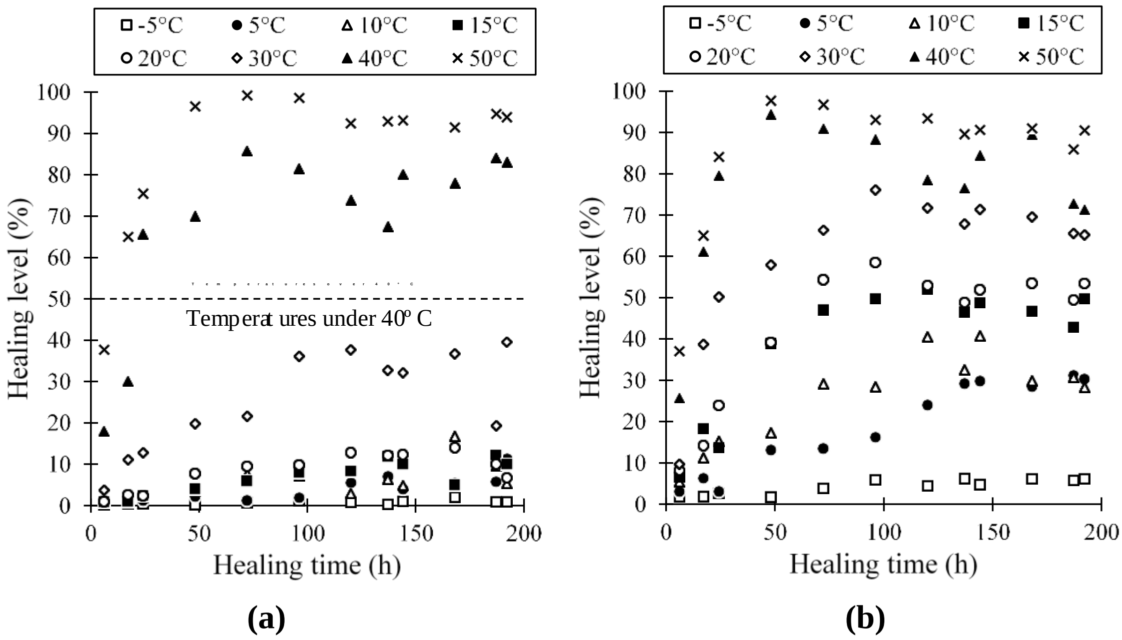

Figure 22. Crack-healing results for asphalt samples tested at different temperatures, (a) without and (b) with capsules.

Furthermore, from Figure 22 it can be noticed that tests performed at 30ºC or below show higher healing levels for asphalt samples with capsules than those without capsules, except at -5ºC where the healing levels results were similar. This could result from the reduction of the bitumen viscosity with temperature in the presence of the rejuvenator (sunflower oil) as shown in Figure 21(a). In this figure, it can be observed that the viscosity of bitumen (the asphalt binder) considerably decreased with temperature in the presence of oil, except at -5ºC, which remains almost the same. Likewise, comparing Figure 22(a) and (b) it can be seen that for tests at healing temperatures of 40ºC or higher, the healing level of samples without capsules begin to rise reaching similar or higher values than those of the samples with capsules.

To prove this, Figure 24 shows the average maximum values of healing levels for the asphalt samples with, and without, capsules versus the different healing temperature evaluated. In Figure 24, it can be seen that the maximum average healing level for samples with capsules is almost linear with temperature, while there is a jump in the curve of the healing levels for samples without capsules. The curve of samples without capsules is linear until 20ºC, then jumps considerably intersecting the curve of asphalt samples with capsules when it reaches 40ºC, see Figure 24. So, the effect of the capsule addition on the healing levels of asphalt is more easily appreciable at temperatures lower than 40ºC.

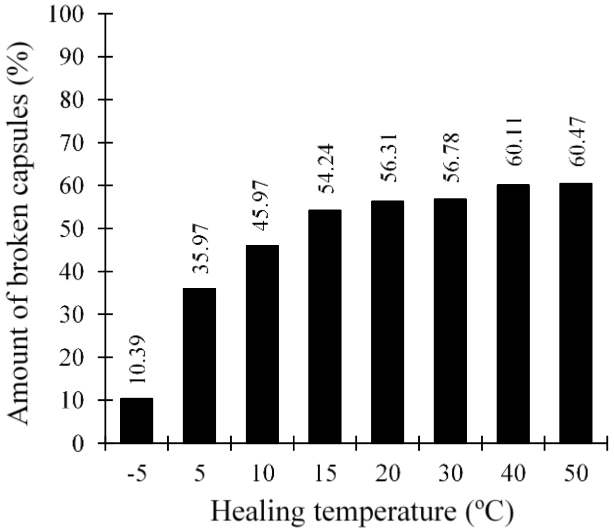

Figure 23. Amount of broken capsules in asphalt mixtures with 0.5% capsule content at different temperatures.

This result may be due to the combined effect of decreasing viscosity and thermal expansion with temperature, as presented in Figure 25. In this way, as can be seen in Figure 25(b), for the temperature range evaluated between 5ºC and 50ºC, the thermal expansion of asphalt mixture samples without oil, 15.62 µm/(m.ºC), was higher than that for asphalt mixture samples with oil 9.202 µm/(m.ºC), and it continues until reaching the melting point, which is 48.05ºC and 36.37ºC for asphalt mixture samples without and with oil, respectively. Therefore, from Figure 25 and Figure 24 results it can be concluded that: 1) asphalt mixture samples with, and without, capsules present a proportional increase of the healing level when the temperature increases, and 2) there is a relationship between the healing level results of asphalt mixture samples with, and without, capsules, which in turn is a function of the variables of viscosity and thermal expansion with the temperature.

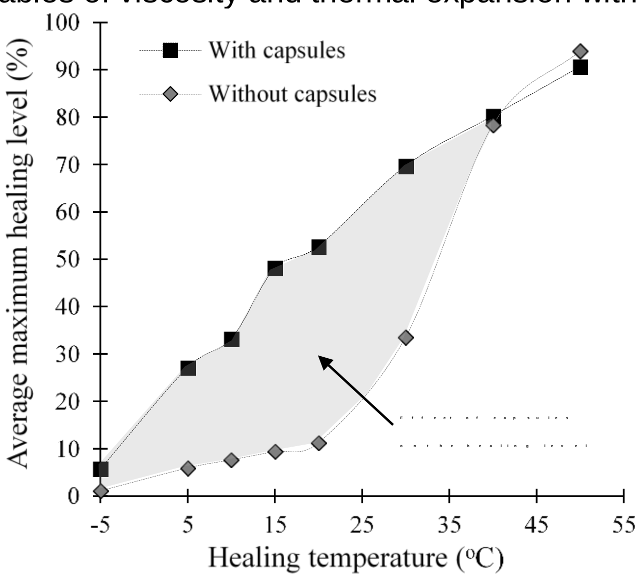

Figure 24. Average healing levels achieved for asphalt mixture samples with and without capsules at different temperatures.

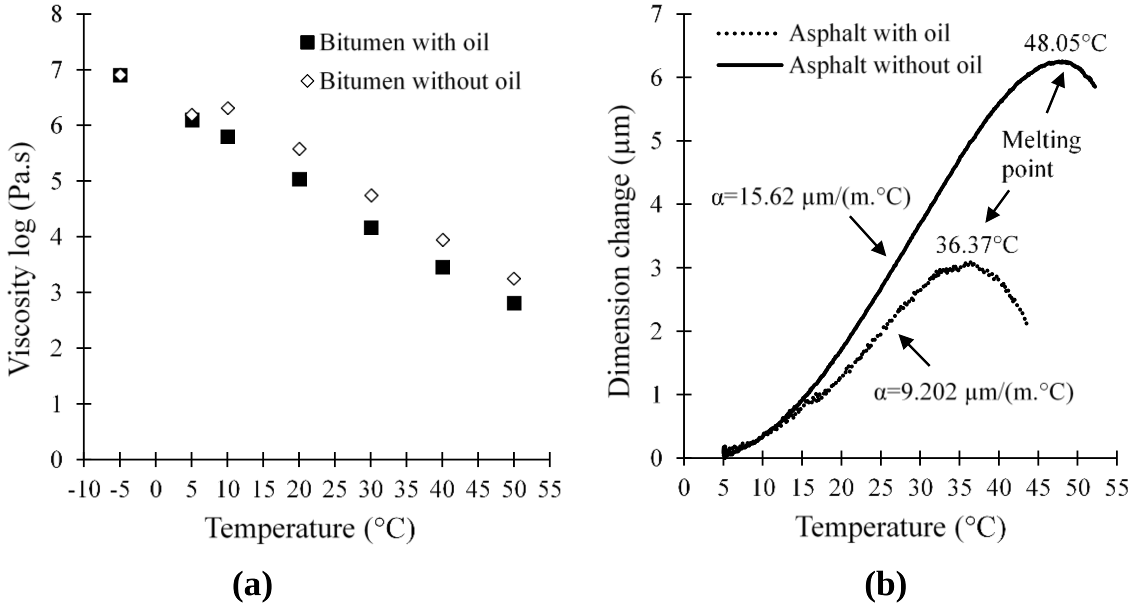

Figure 25. Oil effect on bitumen and asphalt: (a) viscosity test for bitumen, and (b) thermal expansion for asphalt samples.

4.8 Influence of mixing and ageing time of capsules on the self-healing of single cracks in dense asphalt mixtures

The results of the healing levels of single cracks are presented in Figure 26(a). In this Figure, it can be observed that: 1) the healing level reached by the mixtures was different depending on the mixing type, and 2) healing levels of mixtures that were not aged were higher than those of mixtures submitted to ageing process. In this study, it was proven that ageing the mixtures in an oven at 85ºC for 240 h caused an increase in the viscosity of bitumen and oil of 71.09% and 65.84%, respectively (see Table 4), which causes a reduction of the healing properties of mixtures.

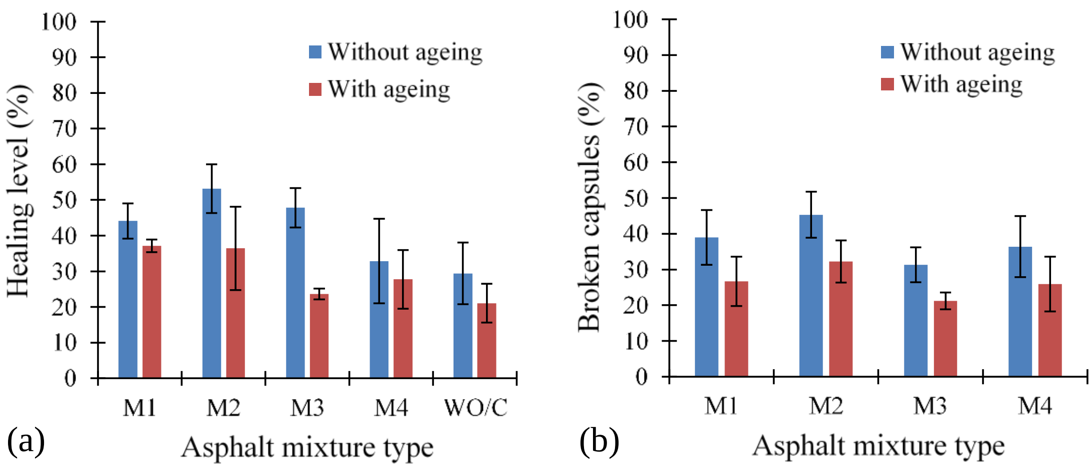

Figure 26. Results of the (a) healing levels for the different asphalt mixture types, and (b) percentage of broken capsules inside the different asphalt mixtures evaluated at healing time of 120h.

Additionally, in Figure 26(a) it can be observed that the mixtures with capsules with, and without, ageing presented, on average, higher healing levels than mixtures without capsules (WO/C). Overall, mixtures with capsules presented a similar trend in the average healing levels, regardless of the ageing state, see error bars in Figure 26(a). For example, in the case of mixtures without ageing, M2-mixture with capsules presented the highest healing level with an average value of 53.16%, followed by mixtures M3, M1 and M4, with average healing levels of 47.82%, 44.11% and 32.86%, respectively, while the healing level in the reference asphalt mixture without ageing was 29.40%.

Table 4. Average viscosity and ageing indices for the bitumen and sunflower oil at different ageing times.

Ageing time (h) in oven @85ºC	Viscosity value (Pa·s)		Ageing index (%)	
	Bitumen	Sunflower oil	Bitumen	Sunflower oil
0	373900	0.073	--	--
2	379260	0.074	1.43	2.04
24	407400	0.075	8.96	3.29
48	409700	0.086	9.57	17.28
72	469100	0.090	25.46	23.87
96	496000	0.091	32.66	24.69
144	562500	0.100	50.44	36.63
180	570300	0.101	52.53	37.86
210	620500	0.109	65.95	49.79
240	639700	0.121	71.09	65.84

These healing level results directly depended on the percentage of broken capsules in the mixtures. Figure 26(b) shows the percentage of broken capsules inside the different asphalt mixtures after applying external compressive loads. Thus, mixtures with higher percentages of broken capsules presented, in general, higher average healing levels in Figure 26(a). The fact that mixtures with ageing present lower values of oil-release was due to the increase of the oil viscosity with the ageing time, see Table 4. In this study, the oil inside the designed capsules is stored inside small cavities of the calcium-alginate structure, see SEM image in Figure 2(b). Therefore, when the capsule is activated by an external compression load, the oil is released from the cavities into the bitumen in small volumes, thus avoiding the saturation of the bitumen with oil. Hence, if the viscosity of the encapsulated oil increases, the oil release from the capsules is more difficult, thus reducing the healing levels in the mixtures with ageing, see Figure 26(a).

Furthermore, with the aim of verifying the efficiency of the capsules in the healing process of cracked asphalt beams, the viscosity of different bitumen samples (with, and without, ageing) mixed with oil without ageing, was measured. The amount of oil added to the bitumen samples was calculated using the percentage of broken capsules in the mixtures, based on Figure 26(b). Table 5 presents the average results of viscosity for the different mixture types evaluated, from M1 to M4. In this Table, it can be observed that all bitumen samples with oil presented lower viscosity values than the reference values at 0 and 240 h, shown in Table 4, for asphalt mixtures without, and with, ageing, respectively.

Table 5. Average viscosity values and viscosity reduction indices of the oil-release asphalt samples with and without ageing.

Mixture type	Broken capsules in mix (%)		Viscosity value (Pa.s)		Viscosity reduction index (%)	
	Without ageing	With ageing	Without ageing	With ageing	Without ageing	With ageing
M1	38.96	26.68	349847	486985	6.43	23.87
M2	45.30	32.24	332340	471278	11.12	26.33
M3	31.34	21.20	366700	514650	1.93	19.55
M4	36.40	25.94	365510	494949	2.24	22.63

Additionally, it can be observed in Table 5 that the viscosity reduction in samples with ageing was higher than that in samples without ageing, see values of the viscosity reduction indexes in Table 5, which can be interpreted as a measurement to quantify the efficiency of the capsules in asphalt mixtures healing. In this way, the results obtained for the viscosity reduction indexes demonstrated that the addition of capsules with oil inside aged mixtures can help the bitumen to rejuvenate by between 19.55% and 26.33%, depending on the mixing order of the capsules.

4.9 Single crack self-healing properties of SMA mixtures with, and without, addition of capsules

Figure 27(a) presents the results of the crack healing levels of SMA mixtures with, and without, capsules evaluated after different healing (rest) time ranging from 5 to 216 h. In this Figure, it can be observed that: i) the crack healing levels in the SMA mixture beams with capsules were higher than in beams without capsules, and ii) the crack healing level of mixtures with, and without, capsules increased with the healing time until a maximum value, and then the healing level remained constant. Micaelo et al. [12] indicated that after the maximum healing value, a new state of physical equilibrium usually is attained at the crack surface of the asphalt beams, which causes the healing level to remain constant. Based on the results, the maximum healing level value was reached at 96 h and remained mostly constant until 216 h.

In this way, crack healing level values registered for the SMA beams evaluated between the healing times of 5 and 72 h increased linearly, see Figure 27(a), reaching healing growth rates of 0.32 %/h and 0.41 %/h for mixtures with, and without, capsules, respectively. Moreover, the average healing results reached in the maximum healing level for SMA mixtures with, and without, capsules were 55.04% and 43.56%, respectively. These results proved that the capsules in the SMA mixtures resulted in higher healing levels for all the healing times studied, because the encapsulated rejuvenator released from the capsules to the bitumen improved the natural self-healing properties of the SMA mixtures. Therefore, the crack healing level results were directly dependant on the percentage of broken capsules inside the SMA mixtures.

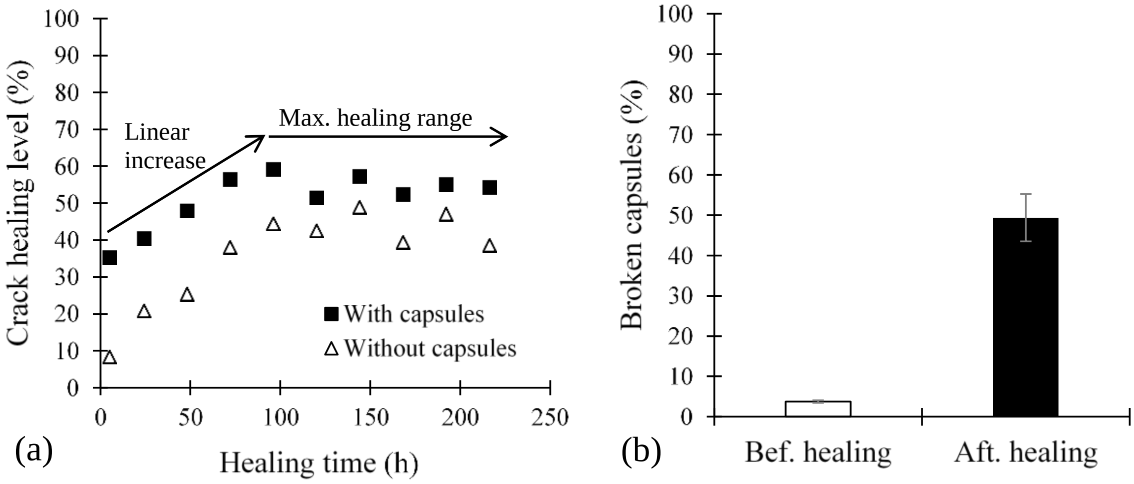

Figure 27. Results of the: (a) healing levels for the SMA specimens with (W/C), and without (WO/C), capsules versus healing period, and (b) percentage of broken capsules inside the different SMA mixtures before, and after, healing process.

In this context, Figure 27(b) shows the percentage of broken capsules inside the different SMA specimens after applying compressive loads. From this Figure, it can be proven that some capsules can be broken due to the mixing and compaction processes, see CT-Scan image in Figure 28(b), registering a content of broken capsules of 3.73%. However, the amount of broken capsules after applying an external compression force of approximately 75 kN on the SMA beams and healing test was 49.39%, which is equivalent to 13 times the percentage of broken capsules before healing.

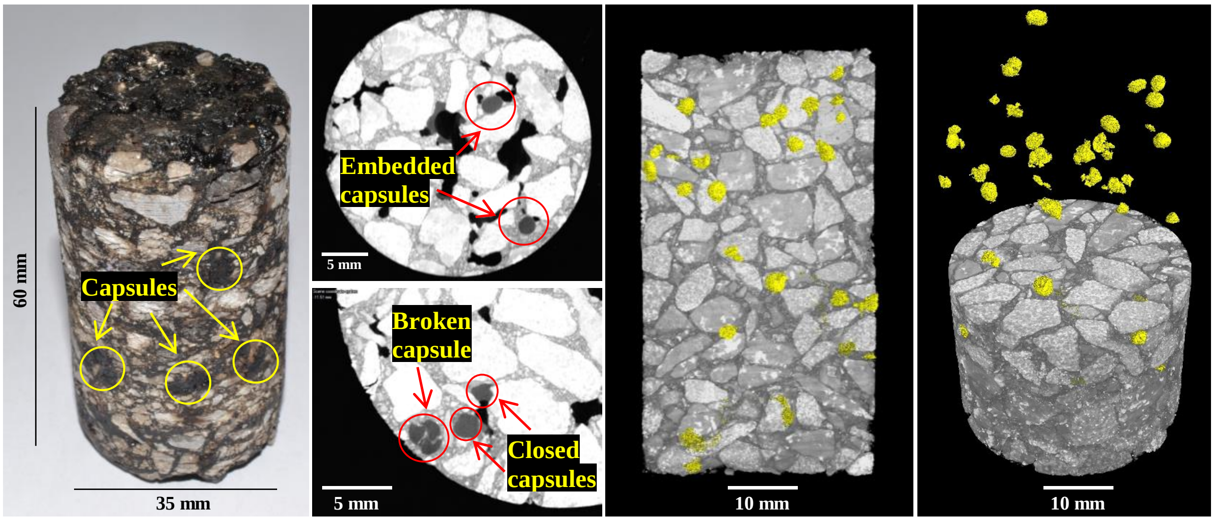

Figure 28. CT-Scans reconstructions of the SMA mixture with 0.5% of capsules: (a) SMA mixture core extracted from a cylindrical Marshall specimen, (b) 2D cross-section images of the sample with embedded capsules inside, (c-d) 3D reconstruction of the capsule spatial distribution inside the SMA sample, capsules have been highlighted in yellow colour.

Likewise, the oil released by the capsules could produce the risk of skid on the surface of the SMA mixtures. To provide evidence, Table 5 presents the average results of the skid resistance measured on the SMA beams with, and without, capsules. In this sense, the results of BPN suggested that the capsules slightly reduced the skid resistance of the SMA pavements, however, considering the variation of the results for the tested mixtures with, and without, capsules this reduction was not significant. Therefore, the oil released by the capsules did not produce risk of skid on the surface of the SMA asphalt pavement compared with SMA mixtures without capsules, which is a guarantee for its installation in real conditions.

Table 6. Probability of breakage by means of $N_{0.5}$ in asphalt mixtures with/without capsules.

Load (kN)	Number of cycles
1.75 W/C	56440
2.75 W/C	5560
2.75 WO/C	22080
3.75 W/C	2360
4.75 W/C	820

W/C: With capsules WO/C: Without capsules

4.10 Self-healing of fatigue damage in asphalt mixture.

Table 6 presents the $N_{0.5}$ probability of breakage for each applied load. As the Table shows, the number of cycles decreased as the load increased. 12 samples were used to obtain the Weibull probability distribution and $N_{0.5}$ breakage for each load, as can be seen in Figure 29. It can be observed that there is a continuous decrease in the number of cycles for failure, when the applied load increases.

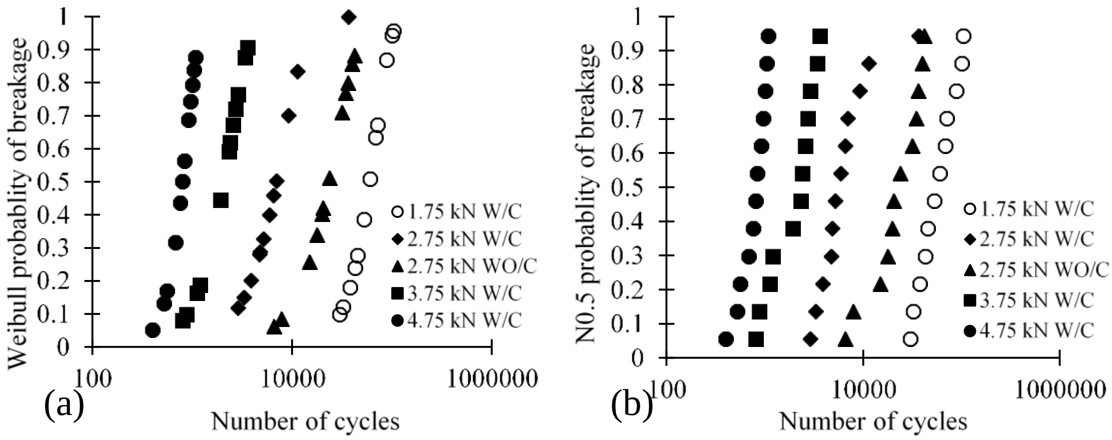

Figure 29. Number of cycles vs. probability of breakage: a) by means Weibull, b) by means $N_{0.5}$.

The relationship between the Healing Index and the number of cycles was obtained from the asphalt mixture samples by subjecting them to tests at four different loads. Healing Index and probability of breakage is presented in Figure 4. It was determined that mixtures submitted to the 1.75 kN load resisted the highest number of load cycles (109,000) in terms of inducing cracking while the mixtures submitted to 4.75 kN load resisted the lowest number of load cycles (620) (see Figure 30). The maximum Healing Indexes were: 0.93, 0.89, 0.47 and 0.36 at 1.75, 2.75, 3.75 and 4.75 kN loading, respectively. The Healing Index value when applying a load of 1.75 kN decreased 61.29% compared with the 4.75 kN load application (see Figure 30).

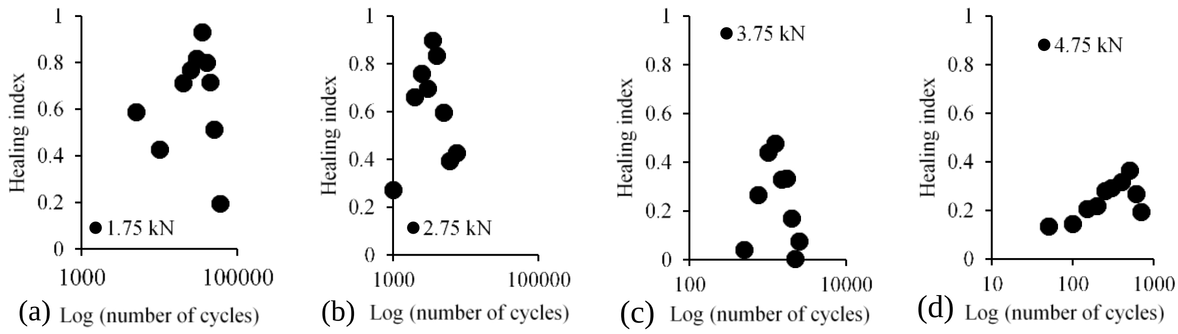

Figure 30. Results of the healing index/number of cycles in the asphalt mixture with capsules for a) 1.75 kN b) 2.75 kN c) 3.75 kN d) 4.75 kN.

The maximum Healing Indexes and Weibull probability of breakage were determined for the different loads: 1.75, 2.75, 3.75 and 4.75 kN, resulting in: 0.14, 0.16, 0.20 and 0.12, respectively (see Figure 31). In Figure 31, it can be observed that the Weibull probability of breakage value corresponding to the maximum Healing Index varied in the range 0.10-0.20.

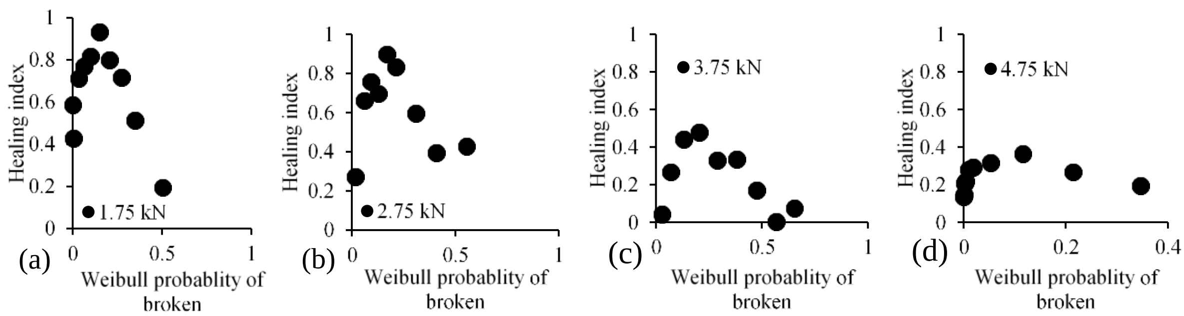

Figure 31. Results of the healing index/Weibull probability of breakage in the asphalt mixture with capsules for: a) 1.75 kN b) 2.75 kN c) 3.75 kN d) 4.75 kN.

In the case of asphalt mixtures with capsules, the Healing Index value of the samples when applying a load of 2.75 kN increased a 5.62% compared to the samples without capsules (Figure 32). This result pointed out the fact that using capsules is effective to increase the Healing Index value. The number of loading cycles resisted by the samples depends on the magnitude of the applied load. The number of resisted cycles decreases with the increase of loads. Moreover, the Healing Index rate in the 3PB fatigue test decreases with the increase of loads. This is because the probability of cracking of the capsules used in the asphalt mixture as a healing agent decreases with increasing loads. Depending on the additionally applied load, the probability of breakage in the maximum Healing Index demonstrates small differences.

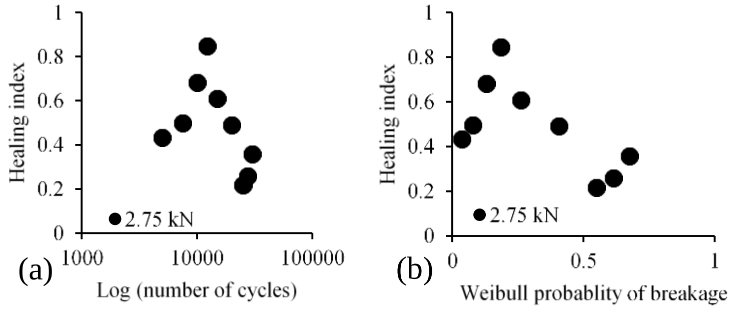

Figure 32. a) Results of the healing index/number of cycles b) results of the index/Weibull probability of asphalt mixtures without capsules.

Figure 33(a) shows the healing level results for all the dense asphalt samples with, and without, capsules tested at a healing temperature of 20ºC and in the loading range from 1.75 to 4.75 kN. From this Figure, it can be observed that the number of cycles and the percentage of broken capsule decreased as the applied load increased. This was because the lifetime of the sample decreases and breaks more quickly as the load increases, so the sample breaks before the capsules. It can also be observed that the optimal Healing Index increased with the increase of average broken capsules in the asphalt mixture (see Figure 33(b)).

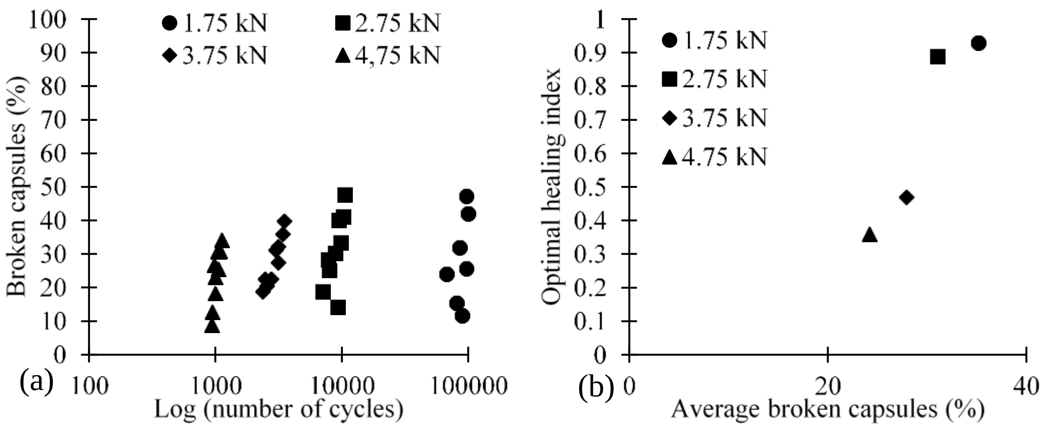

Figure 32. Results of: a) percentage of broken capsules inside the asphalt samples with different loads evaluated, and b) healing indexes for the different loads.

5. CONCLUSIONS

This report has explained the effect of the mixing method and the ageing time on the physical, mechanical and self-healing properties of asphalt mixtures containing 0.5% of capsules with sunflower oil as the encapsulated rejuvenator. In addition, the use of calcium-alginate capsules containing rejuvenator (sunflower oil) on the mechanical performance and self-healing ability of SMA composite mixtures has been evaluated. Based on the results, the following conclusions have been obtained:

- The encapsulation procedure presented in this study allowed the preparation of capsules with an average diameter of 2.5 mm and composed of 75% vol. of sunflower oil and 25% vol. of calcium-alginate polymer. The oil was encapsulated inside a complex microporous structure made of calcium-alginate.

- Mixing temperature did not affect capsules mechanical strength as they can survive mixing and compaction processes during asphalt preparation. Some oil release was noticed but it was about 0.7% of the total encapsulated oil in the mixture. Nevertheless, when the capsules are exposed to high temperatures for long pre-heating periods, they can suffer peroxidation of the encapsulated sunflower oil.

- Density and compressive strength of the polymeric capsules decreased with the temperature, not affecting their mechanical resistance to mixing and compaction processes during the asphalt mixture manufacturing.

- It was demonstrated that the 4h-waiting time before compaction at high temperatures can affect the physical properties of the asphalt mixtures with, and without, capsules. This variation was attributed to the order of addition of capsules and to the ageing damage of bitumen.

- In addition, it was proved that 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 reached by the asphalt mixtures.

- Moreover, it was observed by X-ray computed tomography tests that the capsules' distribution inside the asphalt mixtures can be considered as uniform, not presenting a negative influence on the mechanical properties evaluated.

- The stiffness modulus, tensile strength and fatigue resistance, and water sensitivity of SMA specimens containing calcium-alginate capsules presented similar results to SMA without capsules. This result evidenced that a capsule content of 0.5% did not significantly affect the mechanical properties of the SMA mixtures.

- The oil released by the capsules did not reduce the skid resistance of SMA mixtures, compared with SMA mixtures without capsules.

- Capsules show significant healing levels when added to asphalt samples in comparison to asphalt samples without capsules at healing temperatures equal or less than 30ºC. In addition, the healing level of asphalt samples with, and without capsules increased with the healing time until a maximum value where the healing level remained constant until the end of the test. It is recommended for future research to study the possibility of providing enough rest period on asphalt roads, with respect to rest period that healing start to be constant. Then measuring the self-healing, which might be more economic.

- The healing levels obtained by the asphalt mixtures varied depending on the order of addition of the capsules, and mixtures with capsules showed higher healing levels than mixtures without capsules. Besides, the healing levels for the mixtures without ageing were greater than those of mixtures after ageing process.

- Moreover, the viscosity reduction indices demonstrated that the addition of capsules to aged mixtures can help the bitumen to rejuvenate by between 19.55% and 26.33%, depending on the mixing order.

- Healing levels of asphalt samples without capsules start to increase considerably after 30ºC. At 50ºC, the healing levels of asphalt samples without capsules is higher than that with capsules. This explains the effect of viscosity and thermal expansion on self-healing, in the presence of temperature. In this way, it is recommended to study different types of asphalt binder and how their thermal expansion affect self-healing at different temperatures.

- The healing levels for asphalt samples with and without capsules were almost similar at healing temperatures over 40ºC. This indicates that these capsules are not useful for asphalt self-healing at hot climate. For future work using calcium-alginate capsules with oil, the authors recommend that asphalt self-healing tests be performed at healing temperatures below 40ºC, in order to obtain a significant influence of the capsules on self-healing.

- Finally, based on the results of the physical, mechanical and self-healing properties of asphalt mixtures containing capsules, the authors recommend adding and mixing the capsules with the hot asphalt mixture at the end of the mixing process, without the need to heat the capsules over 20ºC prior to the mixing process.

6. RECOMMENDATIONS

The capsules produced in the project resisted mixing and compaction, in the laboratory at an industrial level. The properties of asphalt mixture that contained capsules were equivalent to those of asphalt without capsules, except for reduced fatigue life that was caused by an excessive size of the capsules. Although it was observed that capsules could repair crack damage in asphalt mixture, it is not yet clear if the capsules will break at the right moment to repair cracks caused by traffic loading, such as bottom-up cracks and ravelling.

It is recommended that the efficiency of the capsules for asphalt self-healing is tested under accelerated conditions that reproduce the real traffic conditions observed in roads, and that the size of capsules is reduced and optimised to improve asphalt self-healing and minimise possible reductions in quality due to their excessive size.

Furthermore, based on previous experiences of the Healroad project, from Infravation, at the moment it is unclear how to use accelerated pavement technology to cause cracks in asphalt mixture. In order to be able to use any accelerated pavement testing machine to cause cracks, it will be required to (i) develop a testing technique that is able to generate cracks in asphalt slabs and (ii) learn how to quantify crack damage in asphalt slabs at the same time as cracks are generating in the slabs.

With regard to understanding the crack development in asphalt slabs under accelerated pavement testing, it will be required to install sensors, such as deformation, damage, temperature and moisture that quantify changes in the materials during the testing period. In order to process all the information generated by the sensors, it is suggested that machine learning techniques are used that will allow detecting hidden patterns in the data and flag the moment when significant cracks start appearing, to compare them against the amount of oil released by the capsules.

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