

# Evaluation of the self-healing capacity of concrete with low-cost macro-capsules

Niranjan Prabhu Kannikachalam<sup>1,2\*</sup>, Emmanuel Cailleux<sup>3</sup>, Nele De Belie<sup>2</sup>, and Liberato Ferrara<sup>1</sup>.

<sup>1</sup>Department of Civil and Environmental Engineering, Politecnico di Milano, Italy

<sup>2</sup>Department of Structural Engineering and Building Materials; Magnel-Vandepitte Laboratory, Ghent University, Belgium

<sup>3</sup>Buildwise (previously BBRI: Belgian Building Research Institute), Research Facility, Limelette, Belgium

**Abstract.** This study focuses on the evaluation of the efficiency of a low-cost macrocapsule, using commercially available pharmaceutical capsules with specific modifications, for self-healing concrete. The macrocapsules were developed by the Belgian Building Research Institute in a previous study. The healing agent is a resin based on alkyd-urethane, a low-cost commercial product, which was selected for its compatibility with concrete and shell, and also for the following reasons: resin release, adhesion to concrete, and reduction in capillary water absorption. After their manufacturing, the macrocapsules were carefully integrated within the concrete mix at 5 volume-%, and cubes and slabs for compressive and impact tests were cast. Small beams 160 x 40 x 40 mm<sup>3</sup> containing each three capsules (placed 15 mm above the bottom surface) were tested for flexural strength and capillary water absorption. The effect of self-healing was evaluated by sorptivity test for two different crack mouth opening displacements of 0.5 mm and 0.9 mm. In both cases, the cracks were partially or completely healed, and the mechanical properties of the macrocapsule specimens were quite the same as the reference specimens. This demonstrates that the modified low-cost macrocapsules are sufficient to heal large cracks without losing the concrete mechanical properties.

## 1 Introduction

The scientific community has been increasingly focused on developing technologies that can provide cement-based materials with self-healing properties to address their tendency to crack. This has involved exploring various methods, including using vascular networks, capsules, bacteria, and smart aggregates [1]. The use of capsules as a self-healing technology has been extensively studied regarding the materials used for the capsules and the healing agents they contain. These capsules can be spherical or cylindrical and can be made from various materials, including glass, silica, natural fibres, gelatin, polypropylene, polyurethane and ceramic [2]. This self-healing technology relies on the idea that when a crack encounters a capsule, the capsule will break open and release the healing agent it contains into the crack, repairing the damage [3]. Capsules can be divided into two categories based on their diameter: microcapsules, which are smaller than 1 mm, and macrocapsules, which are larger than 1 mm. It is important that the material used for the capsules can withstand the alkaline environment of concrete and does not react with the healing agent inside. The capsules must also form an airtight seal between the healing agent and the concrete environment in order to prevent any reaction of the healing agent inside the capsule. Additionally, the material used for the capsules should be brittle enough to break open when a crack occurs, releasing the healing agent.

The development of self-healing technology using capsules has made significant progress, but creating a capsule with desirable properties remains a challenge. One issue is the survival of cylindrical capsules when added to concrete mixes, as well as the difficulty in breaking spherical capsules, which are less likely to be crossed by cracks. Another concern is choosing a suitable healing agent that hardens at an appropriate rate. This means it should not solidify too quickly after a crack appears, allowing enough time for it to spread into the crack, but also should not take too long to harden to prevent harmful substances from penetrating the concrete. Additionally, placing the capsules in or near the cover zone, where most cracks form, can make the capsule technique economical. On the other hand, for ease of practical application, a technique or equipment is needed to mix the capsules in the concrete without breaking them in the process. To address these issues, the Belgian Building Research Institute developed low-cost macrocapsules using commercially available pharmaceutical standard capsules and modified them for use in concrete [4]. Alkyd-urethane was selected as healing agent based on viscosity, reactivity, reaction time, and ability to scale up. They also created equipment for mixing the capsules with fresh concrete as it is placed in the formwork. This study evaluated the mechanical and durability performance of these low-cost macro-capsules, including their effect on the compressive strength, flexural strength,

\* Niranjan Prabhu Kannikachalam : [niranjanprabhu.kannikachalam@polimi.it](mailto:niranjanprabhu.kannikachalam@polimi.it)

sorptivity, and impact resistance of the self-healing concrete.

## 2 Material and methods

### 2.1 Production of macrocapsules

The Belgian Building Research Institute (BBRI) developed and patented [4] a low-cost macrocapsule technique for self-healing concrete in the CAPDESIGN Project - Encapsulation of polymeric healing agents in self-healing concrete. This technique uses readily available pharmaceutical gelatin capsules and a healing agent that meets certain criteria, such as compatibility with the capsule shell and the concrete and rupture capability of capsule. Zero-size pharmaceutical capsules, which can hold each up to 0.68 ml of the self-healing agent, were selected to seal crack widths from 0.3 to 1 mm. Standard capsule sizes and their holding capacity are shown in Fig. 1. Another advantage of zero-size capsules among the other sizes is the lowest diameter to length ratio, which enhances rupturing of capsules during crack propagation. The highest diameter to length ratio would be 1 for a spherical capsule, where cracks tend to pass around the capsules instead of slicing through them.



Fig. 1. Standard capsule sizes and their holding capacity

An alkyd-urethane healing agent was selected for this study because it is commercially available as a varnish, is compatible with both capsules and concrete, is single-component, and hardens within 24 hours in contact with air. The viscosity of the resin was measured using a viscometer and was found to be 458 mPa.s at shear rate 100 s<sup>-1</sup>. By adding 10% of solvent, the viscosity was reduced to 229 mPa.s, which is approximately 50% of the original viscosity, as shown in Table 1. Using a high quantity of solvent can impair the bonding of the alkyd-urethane with the concrete, so it was important to limit to 10% solvent. The reduction in viscosity was necessary to fill crack widths ranging from 0.3 mm to 1 mm.

Table 1. Change in resin viscosity with the addition of solvent

Amount of solvent added to the resin (%)	Measured viscosity (mPa.s)	Percentage decrease in viscosity (%)
0	458	-
5	318	31
10	229	50
15	175	62

The capsules were filled with the resin using a manual capsule-filling machine, as shown in Fig. 2. The body and cap of the capsules were placed on opposite sides of the manual capsule machine (Fig. 2a), and the bodies were filled with the diluted resin using a pipette. The lid of the machine was then closed (Fig. 2b) and lifted (Fig. 2c),

gently pushing on the top of the lid to remove all the filled capsules (Fig. 2d). The caps of the filled capsules were then pushed to lock them into place using the push-lock mechanism of the capsules. This made it difficult to remove the caps and it was safe for moving and storing the capsules.

The capsules used in this study are made of gelatin and are susceptible to reacting with water, which can cause them to deform when mixed with concrete. Additionally, the smooth surface of the capsules can hinder their bonding with the concrete matrix, allowing cracks to pass around the capsules instead of through them. To prevent these issues, a layer of epoxy was applied to the capsules and fine sand was sprayed on top of the fresh epoxy. This not only protected the capsules from reacting with water, but also improved the roughness of their surface. In more detail, after filling each capsule with the healing agent, the capsules were coated with epoxy and excess epoxy was drained by placing them on a mesh for a few minutes as shown in Fig. 3a. Then, sand with maximum grain size equal to 0.5 mm, was applied to the epoxy-coated capsules and allowed to dry for 24 hours (Fig. 3b-c). As a result of this outer surface modification, the capsules became more airtight and ready to be mixed with concrete. And this modification increased the capsules outer volume by circa 23% from 0.98 ml to 1.28 ml.

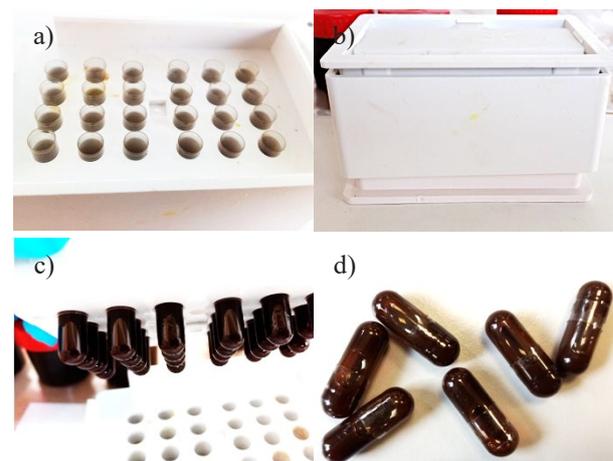


Fig. 2. Manual capsule filling machine

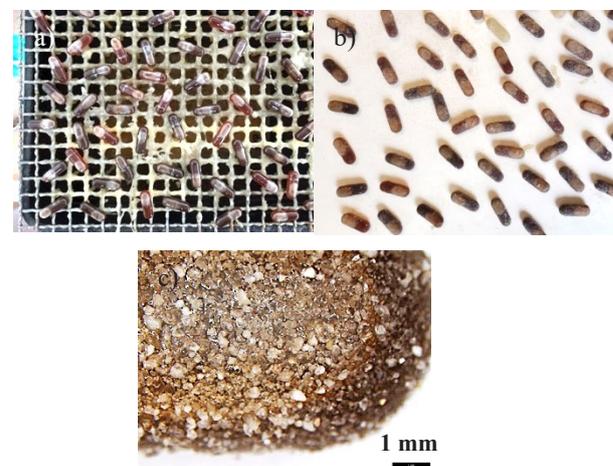


Fig. 3. a) removing excess epoxy on the capsules, b) drying the capsules after the sand coating, c) microscopic image of a capsule outer surface.

## 2.2 Specimen preparation

The reference concrete mix design and type of specimens used are shown in **Table 2** and **Table 3**. Macrocapsules are fragile and would be destroyed by mixing them in a concrete mixer, so they were mixed into the reference mix by hand before being placed in the mould. Each cube and slab specimen produced contained 132 capsules, approximately 5% by volume, arranged randomly. To prepare beam specimens for studying water absorption and flexural strength, three capsules were placed in the middle of the specimen, 15 mm from the bottom surface. This was done by first filling the mould with 15 mm of concrete, then positioning the capsules in the middle, and finally pouring the remaining concrete over the capsules and compacting the specimen lightly. All the specimens were demoulded after 24 hours and cured in a 95% relative humidity chamber until testing. Cube specimens were used to compare compressive strength, and slab specimens were used to compare impact behaviour.

Mixing the capsules by hand, as done in this study, is not viable at the industrial level, so BBRI designed a machine to mix them on-site. However, during a trial process, up to 10% of the capsules were crushed, requiring a few more iterations to fine-tune the machine before adopting it on the site.

**Table 2.** Mix design

Constituents	kg/m <sup>3</sup>
Cement CEM I 42,5 R	470
Superplasticizer	2.3
Water	196.5
Natural Aggregate 0/4	1012.7
Natural Aggregate 4/8	676.4
Steel fibres (3D, l <sub>f</sub> = 60 mm, d <sub>f</sub> = 0.75 mm)	20

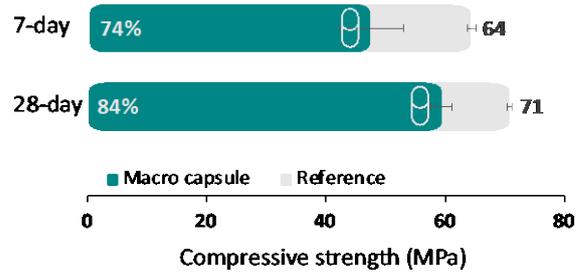
**Table 3.** Specimen details

Specimens	Dimension (mm <sup>3</sup> )	Quantity (each mix)	Capsules (each specimen)
Cube 7 & 28-day (Compressive test)	150 x 150 x 150	3+3	132 (randomly distributed)
Beam	160 x 40 x 40 (Flexural test)	5	3 (Placed in the centre, 1.5 cm from bottom)
Slab	290 x 290 x 40 (Impact test)	2	132 (randomly distributed)

## 3 Experiments

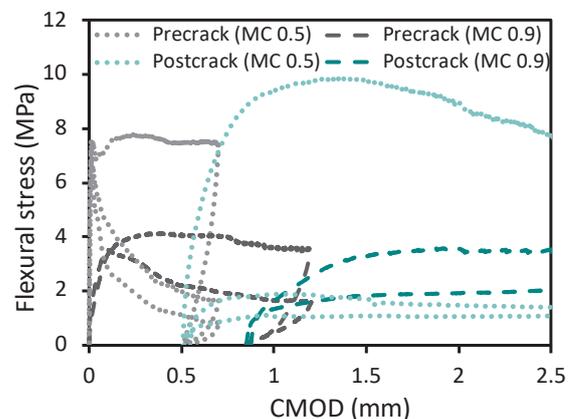
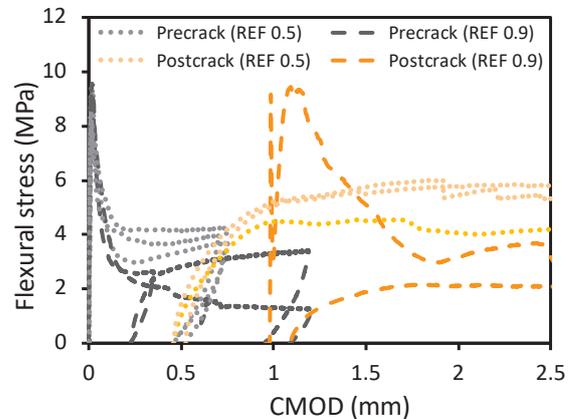
### 3.1 Compressive and flexural test

On the 7th and 28th day, three samples from each mixture were examined for compressive strength. The average compressive strengths of the reference (REF) specimen were 64 MPa (7 d) and 71 MPa (28 d), whereas those of the macrocapsule (MC) specimens were 47 MPa (7 d) and 59 MPa (28 d). Hence, the compressive strength of the MC specimens was 84% of the reference for a 28th-day test. This is a considerable decrease, but could be deemed acceptable because of the specimens' potential to heal. The standard deviation was slightly higher for the specimens with capsules; the values are shown in **Fig. 4**.



**Fig. 4.** Compressive strength of reference and macrocapsule specimens on the 7th and 28th day

Small beams were tested in 3-point bending after 28 days of curing in a 95% humidity room. A 5 mm deep notch was cut at midspan to ease the test control and measure the crack opening displacement at the notch mouth. Loading was applied at a rate of 0.5 μm/s in the pre-peak regime, and raised to 2.5 μm/s in the post-peak part. The specimens were unloaded at a particular displacement to reach two different residual crack mouth opening displacements (CMODs), 0.5 and 0.9 mm. For each mixture, three samples were stopped at a residual CMOD of 0.5 mm (CR 0.5), and two samples were stopped at a residual CMOD of 0.9 mm (CR 0.9), as shown in **Fig. 5**. The average peak flexural stress for reference and macrocapsule specimens was 9 MPa and 6 MPa, respectively, as shown in **Fig. 6** and there was considerable scattering for the macrocapsule specimens, likely due to the disturbance effect by the same macrocapsules.



**Fig. 5.** Flexural stress vs CMOD of reference and macrocapsule specimens

After pre-cracking, the specimens were stored for 50 days in a 50% relative humidity room and sorptivity tests were performed, as detailed below, followed, 60 days after pre-cracking, by three point bending tests to failure to check for mechanical recovery, applying the same loading rate of 2.5 μm/s. The reloading curves are also shown in Fig. 5. There is no noticeable effect of the healing agent on the recovery of flexural strength. This leads to the conclusion that any regain in flexural strength may be due to the hydration of the concrete matrix and the influence of fibres in the concrete matrix rather than the curing of the healing agent, as a similar pattern is seen in the reference and MC specimens.

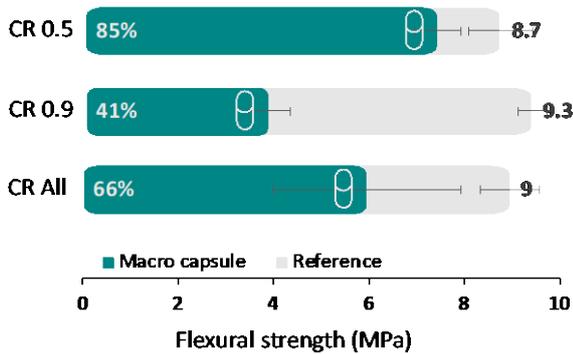


Fig. 6. Average flexural strength of the specimens cracked to CMODs 0.5 mm and 0.9 mm, and average flexural strength of all the cracked specimen (CR All)

### 3.2 Sorptivity test

Small beams were subjected to sorptivity testing before and after the formation of controlled cracks. The specimens were cracked (CR) for two different crack widths of CMOD 0.5 mm and 0.9 mm. The bottom surface and the lateral surfaces over a height of 20 mm of the specimens were coated with silicon, with the exception of a 20 mm wide zone in the centre around the crack for capillary water absorption. This silicon layer was freshly applied to each specimen and dried in an oven for 24 hours, followed by another 24 hours in a chamber with 50% relative humidity. The specimens were placed on two supports to maintain a water level 3 mm above the depth of the notch. After 15 minutes, 30 minutes, 1, 2, 3, 5, and 7, and 24 hours, the water uptake was recorded. After the pre-cracking, the specimens were allowed to heal for a month before the next sorptivity test, mainly because the healing agent in the inner part of the crack may take longer to harden (although probably a week would have been enough). The healing agent generally heals/hardens within 24 hours after exposure to air as per the label mentioned on the product.

Water uptake versus square root of time curves are shown in Fig. 7. The legends on the graphs start with the reference (REF) or macrocapsule (MC), followed by a number, representing the crack width in mm, or uncracked (UNCR). The uncracked reference and macrocapsule specimen's water uptake was the same, and the capsule influence was minuscule. For the cracked specimens, MC water uptake was lower than the reference. To calculate the sorption coefficient (SC) for 24 hours of water uptake,

the amount of water uptake was divided by the area exposed to water and by the square root of time. From Fig. 8, for uncracked specimens, the sorption coefficient was 0.4 kg/m<sup>2</sup>/√hour for both REF, the sorption coefficient was 2.5 times higher for reference specimens, and 1.9 times higher for macrocapsule specimens compared to the UNCR specimens. For CR 0.9, the reference specimen's sorption coefficient wasn't much different from CR 0.5. However, for the macrocapsule specimens, it was 0.67 times that of the UNCR reference and it was almost 200% lower than that of reference specimens with the same crack width. In any case, the water absorption of healed specimens was lower than for reference specimens, and the variation among healed specimens of different crack widths may be due to the viscosity of the healing agent, which could result in a better crack filling for wider cracks.

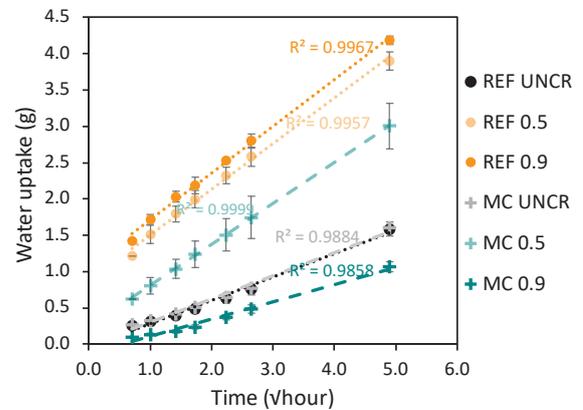


Fig. 7. Water uptake vs time is square root

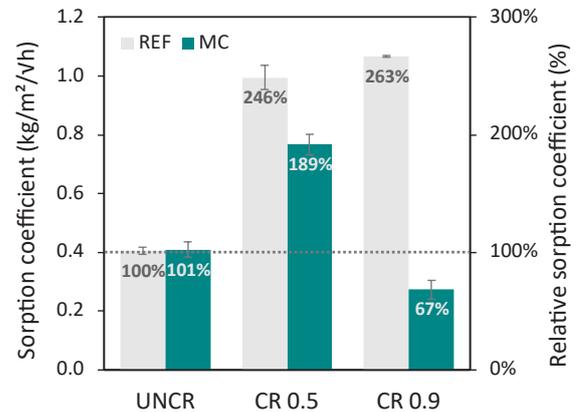


Fig. 8. Sorption coefficient and relative sorption coefficient of the specimens

Self-healing efficiency (SE) of the macrocapsule system was evaluated by the formula (1):

$$SE = \frac{SC_{REF\ CR} - SC_{MC\ Healed}}{SC_{REF\ CR} - SC_{REF\ UNCR}} \times 100 \quad (1)$$

Where  $SC_{REF\ CR}$  is the sorption coefficient of the cracked samples,  $SC_{MC\ Healed}$  is the sorption coefficient of the healed samples by the healing agent, and  $SC_{REF\ UNCR}$  is the sorption coefficient of the un-cracked samples.

Lower healing of specimens with narrow cracks, 39% for MC 0.5 as compared to 120% for MC 0.9, as shown in Fig. 9 is due to the healing agent viscosity, and the actual crack opening at the capsule level, which was about one third of CMOD, which might have hindered the flow-out of the resin from the capsules. Its efficiency could probably be improved by choosing a healing agent with lower viscosity.

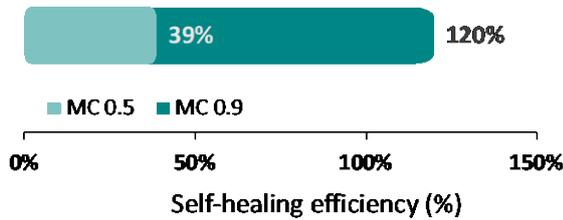


Fig. 9. Self-healing efficiency of the macrocapsule specimens

### 3.3 Impact test

The impact resistance of slab specimens was evaluated using the test setup shown in Fig. 10. It consists of a specimen holder, a 6 kg impactor of diameter 63 mm with a cylindrical body and rounded end, an electro-magnet to control the height at which the impactor was released, a transparent plexiglass tube to guide the impactor's trajectory, a high-speed camera (120 fps) to record the rebound height of the impactor, a laser device to measure mid-deflection of the specimens kept underneath the specimen aligned with the trajectory of the impactor, and load cells to record the impact reaction force. The laser device had a resolution of 2 μm to 120 μm, and the capacity of the impact load cells was 89 kN. The specimen was clamped to the support on the four corners. The test was conducted by releasing the 6 kg impactor from a height of 0.5 m onto the centre of a 10-day-old specimen, with the load cells and laser recording the impact reaction force for the next 5 ms. The rebound of the impactor was captured on video using the high-speed camera. A trigger controlled the release of the impactor and the data storage. The test was repeated on two specimens of each mix, and each specimen was impacted four times.

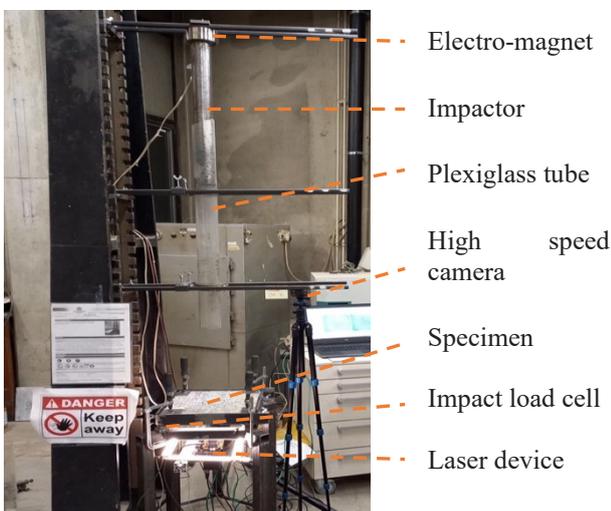


Fig. 10. Impact setup with 6 kg impactor released at the height of 0.5 m above the centre of a specimen

The restitution coefficient (CR) and instant elastic revenue (IRE) were calculated from the obtained data. The restitution coefficient is the square root of the ratio of the rebound height ( $h_r$ ) over the drop height ( $h_d$ ) [eq. (2)], and defines energy dissipated by the plates. The instant elastic revenue is the ratio of plastic deflection to total deflection during loading, as shown in eq. (3), and it assesses the ductility of the plate. Where  $\Delta_{max}$  is the maximum vertical displacement under impact loading at the centre of the specimen and  $\Delta_{steady}$  is the vertical plastic displacement after the impact at the centre of the specimen. Similar techniques were adapted to evaluate impact energy absorption for self-healing concrete by Snoeck et al [5].

$$CR = \sqrt{\frac{h_r}{h_d}} \tag{2}$$

$$IRE = \frac{\Delta_{max} - \Delta_{steady}}{\Delta_{max}} \times 100 \tag{3}$$

In the first impact, the energy dissipated by the slabs made from both mixes was the same. However, after the second impact, the energy dissipated by the slab made from the macrocapsule mix became lower than the value for the reference mix, resulting in a higher CR value, as shown in Fig. 11. Lower energy dissipation was observed in the slabs with the macrocapsule mix due to fewer radial cracks (Fig. 12)

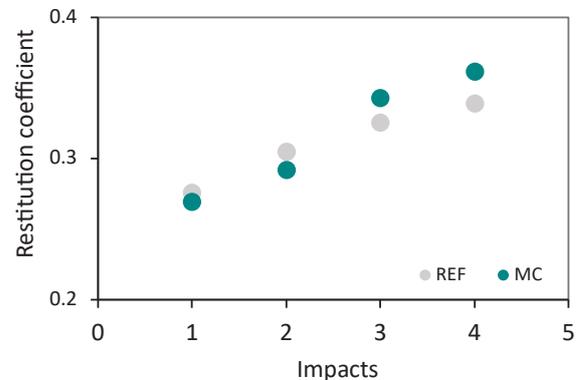


Fig. 11. Restitution coefficient

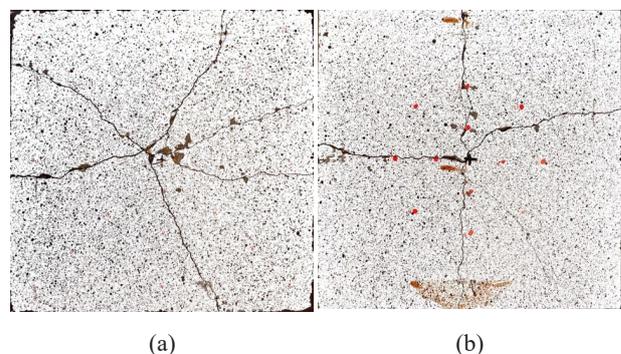
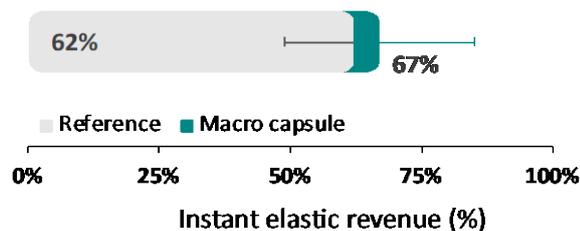


Fig. 12. Images of bottom surface of the reference (a) and macrocapsule slab specimen (b) after four impacts

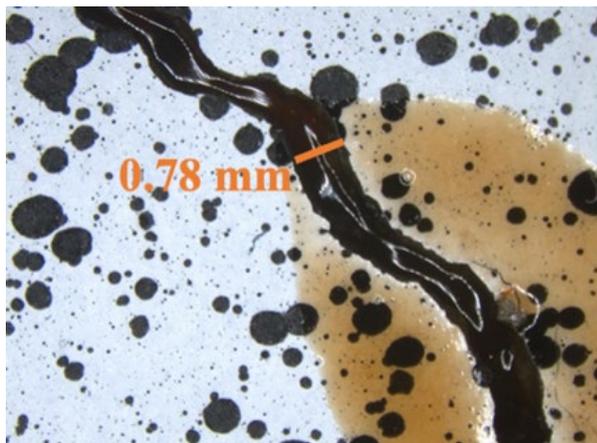
Fig. 13 shows the average IRE of all four impacts, highlighting that the slabs made from the macrocapsule mix were more deformable in comparison to the slabs

made from the reference mix. This is supported by the higher CR value observed in the macrocapsule slabs. It is worth noting that the difference in CR and IRE between the slabs made from the macrocapsule and the reference mix was only marginal, indicating that even under extreme loading, the properties of the concrete did not vary significantly.

After the four impacts, the maximum crack width observed at a certain location was around 3 mm. However, most of the crack widths were around 1 mm, sufficient to contain the healing agent without it flowing out of the crack. **Fig. 14** shows the healed crack. It demonstrates that even under the impact load the cracks were able to contain the healing agents.



**Fig. 13.** Instant elastic revenue



**Fig. 14.** Microscopic image of a healed crack

## 4 Conclusion

This study has examined the performance of cementitious composites containing inexpensive self-healing macrocapsules. The following conclusions can be drawn:

- The introduction of macrocapsules in concrete induce a reduction of its mechanical resistance. The compressive and flexural strengths of the macrocapsule specimens were about 85% of the reference. Nonetheless, this drop can be considered as acceptable from the perspective of the specimens' healing capacity.
- Based on the sorptivity tests, the self-healing efficiency for MC 0.5 and MC 0.9 was 39% and 120%, respectively. It should be possible to improve the self-healing efficiency for both specimens to 100% or more by selecting a more effective healing agent with the appropriate viscosity. Nevertheless, the water

uptake of MC 0.9 was almost 200% lower than that of reference specimens with the same crack width.

- During impact loading, the MC specimens demonstrated lower energy dissipation and more ductile behaviour compared to the reference specimens, although this difference was only marginal. The MC specimens were also able to effectively contain the healing agent within a 1 mm crack width.

So in conclusion, these capsules are simple to produce and can incorporate a large volume of self-healing agent. They are able to fill large cracks but will probably not induce any effect on the mechanical strength of cracked concrete. However, they will significantly reduce the water ingress, especially in the case of large cracks. These effects could probably be modified by changing the self-healing agent: use of a more fluid resin to fill the cracks of smaller dimensions or use of a resin allowing to take up mechanical efforts.



Acknowledgements: This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 860006.

## References

1. N. De Belie, E. Gruyaert, A. Al-Tabbaa, P. Antonaci, C. Baera, D. Bajare, ...H.M. Jonkers, *Advanced Materials Interfaces*, 5(17), 1-28 (2018)
2. D. Snoeck, T. De Schryver, N. De Belie, *Construction and Building Materials*, 191, 13-22 (2018)
3. B. Van Belleghem, K. Van Tittelboom, N. De Belie, *Materiales de Construcción*, 68(330), e159-e159 (2018)
4. O. Vandooren, E. Cailleux, Patent No. BE2019/0020 (2020)
5. C. Xue, W. Li, J. Li, V. W.Y. Tam, G. Ye, *Structural Concrete*, 20(1), 198-212 (2019)