Recent advances on self-healing in cement-based materials through the addition of cementitious macro-capsules

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ABSTRACT: Recently, increasing attention has been directed toward developing technological solutions that introduce self-healing properties into cement-based materials. This approach aims to mitigate the intrinsic sensitivity of these materials to crack formation, thereby preserving their load-carrying capacity, enhancing durability, and reducing overall maintenance costs. Among the various strategies explored to achieve self-healing, the incorporation of capsules stands out as one of the most promising due to its versatility. This study presents recent advances in self-healing cement-based materials through the use of tubular macro-capsules with a cementitious shell, developed by the authors. The responsiveness of the capsules to cracking was demonstrated under realistic flexural conditions. Their influence on the intrinsic material strength was evaluated in comparison to reference materials without capsules. The capsules' ability to store and release various healing agents was studied, focusing on organic (polyurethane-based), inorganic (sodium silicate and silane-based), and biological (bacteria-based) agents. Optimization of the capsule shell formulation, coating, and manufacturing procedures was conducted accordingly. The self-healing effect was evaluated in terms of the restoration of mechanical properties, using static reloading tests up to failure, and the improvement in water-tightness, assessed through various permeability and sorptivity tests. The stability of the self-healing performance over time was also studied under the effect of repeated mechanical and thermal actions, specifically for the polyurethane-based healing agents. The results demonstrate the significant potential of cementitious macro-capsules for self-healing concrete applications.

1 INTRODUCTION

Concrete is the most widely used construction material in the world due to its wide availability and cost-effectiveness, combined with its good mechanical properties and versatility. However, despite its extensive use, concrete is inherently prone to cracking, which can significantly reduce its structural integrity and longevity. Cracks in concrete often arise from a variety of factors, including mechanical stress, environmental conditions, and temperature variations. Over time, these cracks can lead to the ingress of water, chlorides, and other aggressive substances, resulting in reinforcement corrosion, further deterioration, and ultimately in the need for costly repairs and maintenance.

To address these challenges, the concept of self-healing concrete has gained increasing attention in both academic and industrial research. Self-healing concrete refers to a material that can autonomously repair cracks or damage that occur during its service life, thus prolonging its functionality and reducing the need for external interventions (de Belie *et al.* 2018). Research on self-healing concrete has evolved over the past few decades, with various strategies being explored. Among them, it is worth mentioning the stimulation of the intrinsic autogenous healing capacity by means of mechanical restriction of the crack width (Maddalena *et al.* 2020), or the use of crystalline admixtures (Cuenca *et al.* 2020), mineral additives (Litina *et al.* 2021), and superabsorbent polymers (Lefever *et al.* 2023). Other promising strategies consist in the introduction of an autonomous healing mechanism through micro-encapsulation (Al-Tabbaa *et al.* 2019), macro-encapsulation (van Mullem *et al.* 2020), or vascular systems (Shields *et al.* 2021). A large variety of healing agents can be used in these cases, either polymeric (van Mullem *et al.* 2020), mineral (Panza Uguzzoni *et al.* 2023), or bacterial (Romero Rodríguez *et al.* 2020).

Among these strategies, the incorporation of capsules into concrete has emerged as one of the most promising solutions due to its versatility and ease of integration (van Mullem *et al.* 2020). In particular, recent developments in the formulation of macro-capsules with a cementitious shell have shown significant potential for self-healing applications, because of their: inherent compatibility with the surrounding concrete matrix; triggering capability, in response to the mechanical stimulus induced by formation of a crack in the concrete matrix; and ability to effectively store and release diverse types of healing agents.

This study contributes to the growing body of knowledge on self-healing concrete by reviewing the use of tubular macro-capsules with a cementitious shell, focusing on their ability to enhance the material's performance in terms of mechanical recovery after pre-cracking, regain of water-tightness, and long-term stability under repeated mechanical stress and thermal cycles.

2 CEMENTITIOUS TUBULAR MACRO-CAPSULES

2.1 *Polymer-modified cement paste*

Macro-capsules with a cementitious shell were adopted for their envisaged survivability during casting and inherent compatibility with the matrix since they could be sufficiently strong to withstand mixing and brittle to break upon crack formation. Since at first, extrusion was adopted for their production (§ 2.2) (Formia *et al.* 2015, 2016), it was necessary to obtain a paste with good workability and to avoid the collapse of the fresh extruded tubes, therefore adding several compounds to cement (CEM I 52.5 R) and demineralized water, namely:
- Calcium carbonate ($CaCO_3$);
- Hydroxypropyl methylcellulose (HPMC);
- Metakaolin (MK, halloysite calcined at 650 °C for 3 hours);
- Copolymer of ethyl acrylate (EA) and methyl methacrylate (MMA);
- Polyethylene glycol (PEG).

The mixing methodology and compounds were first defined in (Formia *et al.* 2015) and modified by the research group in the following years (Anglani et al. 2020a,b). The current composition of the polymer-modified cement paste is defined in Table 1.

Table 1. Composition of the polymer-modified cement paste.

Cement	Water	$CaCO_3$	HPMC	MK	EA/MMA	PEG
wt%	wt%	wt%	wt%	wt%	wt%	wt%
42.5	19.6	19.6	0.7	0.3	15.7	1.6

2.2 *Production methods*

The first manufacturing process was based on extrusion of the fresh cement paste, using a manually operated screw extruder with replaceable nozzles. This allowed to obtain ridged tubes of variable length and dimensions. In (Formia *et al.* 2015), two typologies of cementitious tubes were produced, using two nozzles with an outer and inner diameter respectively of 5 mm or 10 mm and 2 mm or 7.5 mm. The study proved the higher efficiency of the latter with larger diameters. The tubes curing consists of 7 days in a moist environment (temperature T ≈ 20 °C, relative humidity RH >95%) and subsequently in air (T ≈ 20 °C, RH ≈ 60%). Afterward, they were further cut with a saw into smaller tubes with the desired length. However, deformation of the cross section happened during the first stages of curing due to their own weight, resulting in an ovoid cross section.

To overcome this limitation, a second manufacturing process was developed (Anglani et al. 2020b) in which the fresh paste is rolled around an oiled bar with a circular cross section of the target diameter (usually 5 or 6 mm), until obtaining a smooth cementitious tube with a thickness above 1 mm. The bar is removed after first hardening of the cement paste, i.e., after 24 h storage in a moist environment. This allows them to keep a perfectly cylindrical shape, unlike the extruded ones. The subsequent curing follows the procedure used for the extruded tubes. After curing, the tubes can be further cut with a saw into smaller tubes. However, the rolling procedure allowed to overcome the need of saw cutting, replacing it with pre-notch of the fresh tubes to the target length. Figure 1 shows the cementitious tubes obtained with the two procedures.

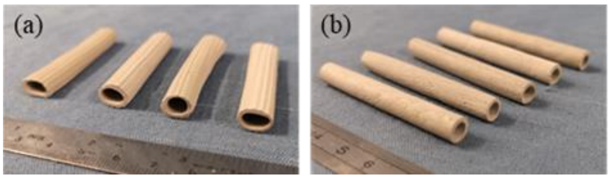


Figure 1. Cementitious tubes produced by (a) extrusion and (b) rolling. Adapted from (Anglani et al. 2020b).

Nonetheless, to ensure proper healing agent storage, the tubes must be further coated, as water can diffuse through the cementitious shell, risking premature reaction (e.g., in case of moisture reactive healing agents) or leakage of the agents itself from the shell. The same applies to the sealing of the capsule ends. To achieve the correct waterproofing, a two-component epoxy primer is applied by complete immersion of the tubes to prepare the surface. Subsequently, a layer of two-component epoxy resin is applied to coat the capsules, either internally or externally, with an overall coating thickness of about 1 mm. After coating, one end of the tubes is sealed using an epoxy-based two-component plaster and further coated with the two-component epoxy resin used for the shell. At this point, the healing agent can be injected with a syringe until complete filling. Finally, the second end of the tubes can be sealed in the same way as the other end. Finally, it is possible to add a finishing layer of sand to improve the bonding with the surrounding matrix, either only on the capsule ends or on the whole capsule (Fig. 2).

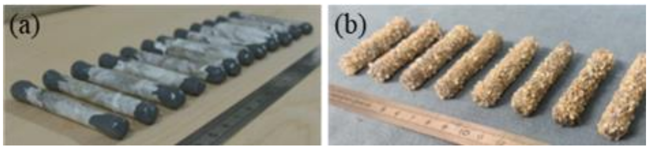


Figure 2. Cementitious capsules with either (a) internal coating (adapted from (Anglani *et al.* 2022)) or (b) external coating and finishing layer of sand.

2.3 *Capsule characterization*

The compatibility with the surrounding matrix can be characterized through mechanical testing of the capsules to confront their characteristics with those of cementitious materials. In (Formia *et al.* 2015) the flexural strength of extruded tubes was tested, either without coating or with a polyester and sand coating, with average values of 1.8 and 1.6 MPa, respectively. The coating did not significantly influence the flexural strength, which was comparable to normal cement. In (Riordan *et al.* 2023) the uniaxial tensile and flexural strength of rolled cementitious capsules with an internal coating were tested, along with the bond strength with surrounding mortar. The measured tensile and flexural strengths were higher than the previous study (>8 MPa), and it is interesting to highlight that the capsules tested for bond strength broke before slippage (Fig. 3), meaning that the bond strength was higher than the tensile strength of the capsules. This is a positive result since a good bond is a prerequisite for the breakage of the carriers upon crack formation.

Figure 3. Capsules after (a) tensile, (b) flexural, and (c) bond tests performed in (Riordan *et al.* 2023).

In the same study, Riordan et al. (2023) assessed the waterproofing of the capsules. First, the watertightness of the shell was measured by filling the capsules with a known volume of water, successively sealing and storing them in laboratory conditions, and measuring the mass change every hour for 24 h with one additional reading at 48 h, positively showing a percentage decrease in mass of only 0.09%. Water uptake of the shell was also measured after full water immersion, storing in laboratory conditions and successive weighing at 24 h and 48 h, showing also in this case a low percentage increase in mass of 0.9%.

3 SELF-HEALING CEMENT-BASED MATERIALS WITH INCORPORATED CEMENTITIOUS CAPSULES

To test the efficiency of self-healing cement-based materials with incorporated cementitious capsules, the majority of tests were performed using cement mortar prisms, either with or without capsules for comparison. Cement mortar was selected as a prototypal cementitious material since the healing agents' reactions usually do not depend on the mortar fraction and the aggregate-cement ratio (van Mullem *et al.* 2020). In addition, mortar is more homogenous than concrete, and it also allows for a fast and cost-effective screening of different self-healing techniques before upscaling for use in concrete (van Mullem *et al.* 2019). However, in recent years, the technique has been upscaled and tested in concrete samples.

3.1 *Incorporation methods*

The incorporation of capsules into cement-based materials can be either obtained by positioning the capsules at fixed positions in the moulds of concrete elements, using for examples the rebars as anchoring points and some devices to anchor the capsules (e.g. threads, grids, supports, etc…), or either by mixing the capsules obtaining a random distribution inside the elements.

The major advantage of fixed positioning is that capsules can be strategically positioned in areas most susceptible to cracking and where crack formation would increase the risk of aggressive substances penetrating the steel reinforcement. The major drawback is the necessity to have additional devices and specialized workers, with a possible cost increase. In several studies involving the use of cementitious capsules, the fixed positioning in mortar prisms was adopted (Anglani et al. 2020a,b, 2022, 2023, Riordan et al. 2023). However, in some cases, a manual placing in a defined location but without fixing devices was also adopted, both in mortar and in concrete samples (Formia *et al.* 2015, 2016, Hermawan *et al.* 2023).

On the other hand, with a random distribution, capsules could be incorporated like aggregates and admixtures, facilitating their use in standard construction practices and industrial concrete production. However, capsules should be capable of enduring the mechanical forces involved during casting (§ 3.2) and could be randomly placed in locations where they would not be necessary, also in this case causing an economical and functional drawback. The possibility of using cementitious capsules with a random distribution in concrete samples was also studied (Formia *et al.* 2016, Hermawan *et al.* 2023, 2024).

3.2 *Resistance to mixing and casting*

A major challenge of random distribution lies in developing capsules that can be mixed into fresh concrete, withstand the mechanical forces involved, and break upon crack formation. This aspect was addressed by developing capsules that could act as "enhanced aggregates" (Anglani 2020) with an internal diameter of 12 mm, both an internal coating and an external coating provided with a sand layer for adhesion, an external diameter of about 15 mm and length of about 23 mm after finishing (Fig. 4). The resistance to mixing of the cementitious capsules was tested by directly adding 24 capsules to the fresh concrete (maximum aggregate=8 mm) in a tilting drum mixer. After some minutes of mixing, water was added to dilute the mixture and cast it into a large plastic container to manually search and count the intact capsules. The survival rate under mixing was defined as the ratio between the intact capsules and the total number of capsules, obtaining a 100% survival rate and the confirmation of the suitability of using these capsules as addition during the mixing process.

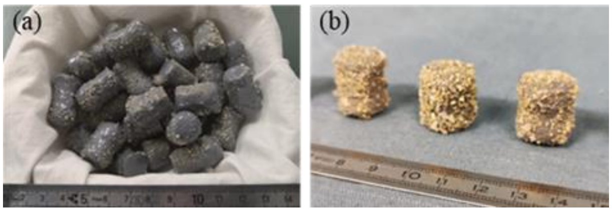


Figure 4. "Enhanced aggregates" cementitious capsules.

The survivability of extruded tubes was preliminary assessed in (Formia *et al.* 2015) and later confirmed for rolled capsules (Fig. 2b, L=54 mm, d_{int}=6 mm, d_{out}=9 mm) in (Hermawan *et al.* 2023), adding them to the fresh concrete (maximum aggregate=16 mm) using both:

– a tilting drum mixer, adding either 40 or 80 capsules and mixing for 3 minutes;
– a planetary/rotary pan mixer, adding either 20 or 40 capsules and mixing for 2 minutes;

successively retrieving the intact capsules after casting. A survival rate of 100% occurred using the drum mixer, while 6 out of 20 and 2 out of 40 capsules were damaged due to the high shear mixing force of the planetary mixer, reaching a survival rate of 70% and 95%. These results confirmed that the cementitious capsules are strong enough to resist mixing.

3.3 *Influence of capsules on fresh and hardened properties*

The addition of capsules may affect the fresh and hardened properties of the cement-based materials. In (Formia *et al.* 2016), the effect of extruded capsules (L=5 cm, d_{out}=1 cm) addition in different orientation in cubic samples (side=10 cm) made of self-compacting concrete was evaluated. The tubes were placed manually in each specimen to dispose them horizontally, vertically, and randomly with respect to the load direction, which resulted in a reduction of 0.3%, 1.6%, and 8.5% compared to the samples without capsules. This variation can be considered negligible. Moreover, tubes resulted in adherence to the concrete matrix even after failure. In (Anglani et al. 2020b) the reduction in flexural strength was evaluated placing in fixed position in the prisms either one extruded internally coated, one extruded externally coated, or two rolled capsules, which resulted in a reduction of 13%, 6%, and 8%, respectively. This showed that the flexural strength is not significantly influenced by the presence of the cementitious capsules. In (Hermawan *et al.* 2023) the effect of adding tubular capsules (Fig. 2b, L=54 mm, d_{int}=6 mm, d_{out}=9 mm) in concrete, with a dosage of 2% by the volume of coarse aggregates, was studied. In terms of fresh properties, the slump passed from 130 mm without capsules to 100 mm, however, both can be categorized in the S3 class. The introduction of capsules increased the air content of the fresh mixture from 3% to 4%, while the fresh density was not altered. Concerning the hardened properties, the concrete without capsules had a compressive strength of 42.9 MPa, and the introduction of capsules caused an 8% strength reduction but had no effect on tensile splitting strength and ultrasonic pulse velocity. A reason for compressive strength reduction was probably due to the packing disturbance that was confirmed in (Hermawan *et al.* 2024), in which it was proved that the introduction of macro-capsules considerably increased the voids ratio of coarse aggregates. Moreover, it was examined how the use of short capsules (Fig. 4) may be beneficial in terms of packing since they can better blend with aggregates than tubular capsules.

3.4 *Capsule triggering*

To evaluate the self-healing efficiency, it is first necessary to trigger the release of healing agents by pre-cracking samples. Three-point bending of prisms was adopted in the majority of cases to create a crack, controlling the crack mouth opening displacement (CMOD) up to a target value (Formia et al. 2015, 2016, Anglani et al. 2020a,b, 2022, 2023, Riordan et al. 2023). The capsule breakage could be noticed by a sudden load drop in the load vs CMOD curve (Fig. 5) and a breaking sound, followed in many cases by a visible release of healing agents from the crack (Fig. 6). In Figure 5, it is possible to highlight a consistent load drop before 150 μm. In addition, Formia et al. (2016) positively evaluated the capsule triggering due to restrained shrinkage. Finally, the sum of capsules triggered in a crack plane in concrete samples with randomly distributed capsules was evaluated in (Hermawan *et al.* 2023).

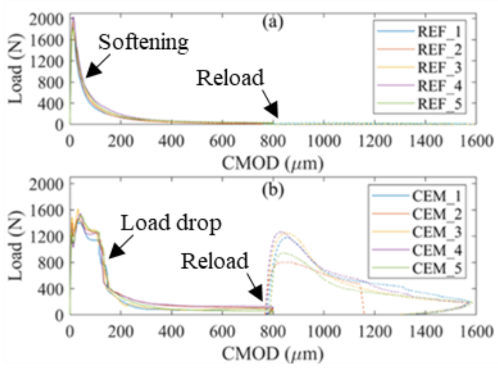
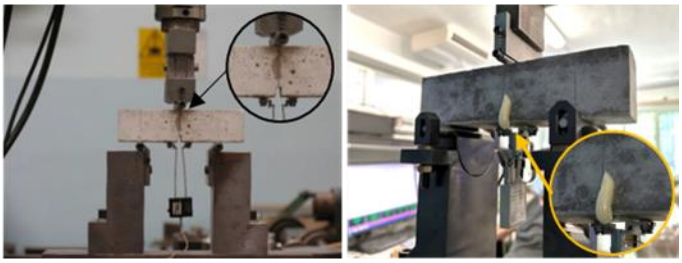


Figure 5. Load vs. CMOD curves of pre-cracking and successive reloading of mortar prisms (a) without capsules; (b) with two rolled cementitious capsules filled with polyurethane.



(a) (b)

Figure 6. Visible leakage of (a) a low viscosity healing agent; (b) a moisture reactive and expansive polyurethane precursor.

3.5 *Self-healing efficiency evaluation*

3.5.1 *Regain of mechanical properties*

The self-healing effect can be evaluated after reloading through a load recovery index (LRI) defined as:

$$LRI\ (\%) = \frac{P_r - P_u}{P_p - P_u} \times 100 \quad (1)$$

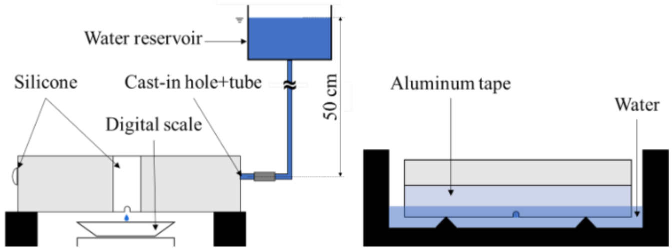
where P_r is the peak load obtained during reloading, P_p is the peak load reached during pre-cracking (i.e., the pristine material maximum load bearing capacity), and P_u is the residual load obtained at unloading after pre-cracking. In Figure 5, it is possible to see the evident absence of mechanical regain after reloading of samples without capsules, and evaluate the good mechanical recovery after the incorporation in fixed position of two cementitious capsules (L=5 cm, d_{int}=5 mm, d_{out}=8.5 mm, internal coating 1 mm, volume of healing agent per capsule V_{ha}=0.7 mL) filled with a highly moisture reactive and expansive polyurethane (PU) precursor, with an average LRI of 70%.

Previous studies showed the recovery using extruded capsules filled with either a sodium or potassium silicate (Formia *et al.* 2015, 2016), with an average LRI of 31% and 8%, respectively. In (Anglani et al. 2020b) the mechanical regain was evaluated placing in fixed position either one extruded internally coated capsule (L=45 mm, d_{int}=7.5 mm, d_{out}=10 mm, V_{ha}=0.6 mL), one extruded externally coated capsule (L=45 mm, d_{int}=7.5 mm, d_{out}=10 mm, V_{ha}=0.9 mL), or two internally coated rolled capsules (L=60 mm, d_{int}=5 mm, d_{out}=8 mm, V_{ha}=0.3 mL), all filled with a highly moisture reactive and expansive PU precursor. These resulted in a LRI of 31%, 47%, and 36%, respectively. Moreover, the study also proved the capacity of the proposed self-healing system to sustain repeated mechanical stress.

3.5.2 *Regain of water tightness*

The regain of water tightness of the pre-cracked and successively healed specimens can be assessed by either measuring:

- the reduction of water permeability using a water flow test (van Mullem *et al.* 2020) (Fig. 7a), measuring the water passing through the crack mouth in samples with a cast-in hole through their length and sealed lateral surfaces, in comparison with reference cracked samples without capsules;
- the reduction of water absorption using a capillary water absorption test (Anglani et al. 2020a, Riordan et al. 2023) (Fig. 7b), measuring the water absorbed through the crack in samples immersed in water, with sealed surfaces apart from the crack mouth, in comparison with reference samples without capsules (cracked and uncracked to account for matrix absorption).



(a) (b)

Figure 7. Test setups for (a) water flow and (b) water absorption.

In (Anglani et al. 2020a) both tests were performed on prisms with extruded capsules in fixed position, either with an internal or external coating, storing a bacterial strain able to induce calcite precipitation (*Bacillus sphaericus* plus deposition medium, BAC), a low viscosity PU precursor, and a silane-based water repellent agent (WRA). The best results in terms of water flow reduction were obtained using PU stored in internally coated capsules (79%) and BAC stored in externally coated ones (78%). Negative results were obtained using WRA. On the contrary, the best results on the water absorption reduction were obtained using WRA (92%). In (Anglani *et al.* 2022) similar results were obtained using a low-viscosity PU precursor stored in extruded and rolled capsules, with a reduction of water flow of 74% and 54%, respectively. The good reduction of water absorption offered by WRA was confirmed in (Riordan *et al.* 2023), with a sealing efficiency of 105%, namely a lower absorption than uncracked samples. Finally, in recent studies, a highly moisture reactive PU precursor (§ 3.5.1) showed positive results both in terms of water flow reduction (100%, namely a complete sealing) and water absorption reduction (92%).

3.5.3 *Long-term stability under repeated degrading actions*

Finally, Anglani et al. (2023) evaluated the ability of expansive PU precursor to endure cyclic flexural loads and temperature variations after being released from cementitious capsules to assess the long-term stability of the sealing. Sealing efficiency was first evaluated through a water flow test immediately after pre-cracking and self-repairing. The test was then repeated at set intervals under force-controlled cyclic flexural loading (10%–70% of the estimated self-repaired strength) and cyclic thermal stress (-20°C to +50°C). The thermal range was selected to achieve an acceleration factor equal to 102 using the Coffin-Manson model (i.e., 1 test day ≈ 102 days under normal conditions).

The mechanical tests showed that when the initial sealing efficiency was 100%, it remained constant throughout the test. Additionally, higher initial sealing efficiency correlated with a greater number of cycles to failure, likely due to the effective spreading of PU over the crack faces, enhancing both mechanical and sealing performance.

A similar result was obtained for thermal aging since the sample with an almost perfect initial sealing (100%, 100%, 99%, 97%) showed just a slight decrease after 50 (simulated) years of aging (83%, 84%, 97%, 94%), showing good perspectives for the long-term stability.

4 CONCLUSIONS

The study highlighted the advances in self-healing cement-based materials using cementitious macro-capsules. The capsules' responsiveness to cracking was validated in controlled conditions, and their impact on fresh and hardened properties was assessed, showing a minimal reduction. Various healing agents were successfully stored and released, with optimization of the shell formulation, coating, and manufacturing processes. Self-healing efficiency was demonstrated through mechanical property restoration and improved watertightness, while the long-term stability of the sealing was evaluated under repeated mechanical and thermal stresses. The findings confirm the significant potential of cementitious macro-capsules for self-healing concrete applications.

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