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Applicability of cementitious capsules in concrete production: initial assessment on capsule robustness, mechanical and self-sealing properties of concrete

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Abstract. The use of macrocapsules in self-healing applications offers a potential benefit by carrying a larger amount of healing agent in comparison with microcapsules. However, the application of macrocapsules is still limited to paste and mortar levels on lab-scale. This is due to a concern that most capsules might be broken when mixed with concrete components. In this study, cementitious tubular capsules were used and they were considered as a partial replacement of coarse aggregates (2 vol% gravel). The capsules have a dimension of 54 mm and 9 mm in length and outer diameter, respectively. A water-repellent agent (WRA) was entrapped in the capsules as a proposed agent to seal the crack. Initial results revealed high survivability of capsules during concrete mixing: 100% survival ratio when tested in a drum mixer and 70–95% when tested in a planetary mixer. The mechanical and self-sealing properties of concrete containing embedded capsules were evaluated. With the addition of capsules, around 8% reduction of compressive strength was noticed, but no further effect on splitting tensile strength was detected as compared with concrete without capsules. Ultrasonic pulse velocity (UPV) tests confirmed that the presence of capsules also did not significantly affect the compactness of the hardened concrete. Furthermore, the embedded capsules were able to break when a crack was introduced and it was found that 90% sealing efficiency was achieved by capsule-based concrete as a result of the successful release of sealing agent into the crack.

1 Introduction

In recent self-healing/self-sealing concrete technology, some healing/sealing agents, which will be introduced in the cementitious composite, are stored inside a vessel. The agent is often called as healing agent if it is able to autonomously heal the crack with (precipitated) products ensuring a crack closure phenomenon (e.g. bacteria, sodium silicate, polyurethane). On the other hand, sealing agent refers to a specific agent with the ability to seal the crack in the way that water or liquid cannot penetrate into the cementitious matrix (e.g. water-repellent agent). In this case, the sealing agent seals the crack without having a crack closure phenomenon. A lot of research is done on encapsulation techniques for both healing and sealing agents. There are numerous vessels such as capsules [1,2], aggregates [3], and polylactic acid particles [4]. Especially capsules are often developed in two distinct technologies: micro-encapsulation and macro-encapsulation. In the micro-encapsulation method, the microcapsules are produced through a series of chemical processes (e.g. in-situ polymerization, emulsification, etc). In fact, a tiny amount of agent is stored inside a microcapsule but the microcapsules are normally produced in bulk. In contrast, the macro-encapsulation method comprises storage of agent in bigger-sized capsules. These capsules can be made from commercial materials such as glass [5], ceramic [6], or polymeric [7] tubes, that can be cut into different sizes. In this case, the agent can be stored in a bigger amount as compared with microcapsules. However, there is sometimes an incompatibility issue between the healing/sealing agent, capsule material and cementitious material. Gruyaert et al. [1] reported a premature polymerization of healing agent (i.e., polyurethane) inside the polymeric capsules. Araujo et al. [5] stated that the use of glass capsules might induce alkali-silica reaction which is critical for concrete durability. To mitigate these issues, cementitious capsules have been recently developed and

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proposed using a polymer-modified cement paste [8,9]. The advantages of using the cementitious capsules are reduced brittleness, reduced risk of alkali-silica reaction, and higher compatibility with the surrounding matrix, with respect to the glass capsules [10]. A mechanical regain of the cementitious composite containing cementitious tubular capsules (with polyurethane as healing agent) under static and cyclic loading was also found [11]. Nevertheless, the application of any type of macrocapsule is still limited to the paste and mortar level [12], and only few studies [5,13] applied the capsules into the concrete. The assessment of the fresh and hardened properties of the capsule-based composite is also rarely done because the main objective of using capsules still relies on the healing or sealing performances. This has been identified by the authors as a research gap. Eventually, this paper attempts to investigate the effects of the macrocapsules in the fresh and hardened concrete. The cementitious capsules (54 mm in length and 9 mm outer diameter) are used in this study. The robustness of capsules toward mixing forces is initially evaluated. The self-sealing properties of capsule-based concrete are assessed by means of a capillary water absorption test. An additional test is conducted to quantify the sealing coverage area as a result of the released sealing agent from the capsules.

2 Materials and methods

2.1 Materials

Table 1 shows the mix designs of reference concrete (REF) and concrete containing capsules (CAPS). CEM III/A 42.5N, having 52% clinker and 48% blast furnace slag, was used as a binder component. Sea sand 0/2.5 was used as fine aggregate, while two fractions of gravels (4/8 and 8/16) were used as coarse aggregates. The specific gravities of sand 0/2.5, gravel 4/8 and gravel 8/16 were 2.67, 2.59 and 2.60, respectively. The water-cement ratio (w/c) was fixed at 0.50 for all mixtures. Polycarboxylate-ether (PCE) superplasticizer (Fluvicon 801, supplied by CUGLA B.V.) with a 20% solid content was used to improve the workability of the mixes.

Table 1. Mix designs

Material	Unit	REF	CAPS
CEM III/A 42.5N	kg/m^3	325	325
Sand 0/2.5	kg/m^3	740	740
Gravel 4/8	kg/m^3	701	678
Gravel 8/16	kg/m^3	378	365
Effective water	kg/m^3	163	163
Superplasticizer	kg/m^3	0.89	0.89
Effective w/c	-	0.50	0.50
Capsules	vol% vs. gravel	-	2

The REF mixture was made without the addition of capsules. In case of CAPS mixture, the cementitious capsules (see Figure 1) were added into the concrete mix with a dosage of 2% by the volume of coarse aggregates. The capsules were treated as a partial replacement of gravels (see Table 1). The length, outer diameter and inner diameter of cementitious capsules were 54, 9 and 6 mm, respectively. On the outer surface of the capsule, a sand layer was applied in order to provide a good bonding between the capsule and concrete matrix. Water-repellent agent (Sikagard 705L) was selected as a sealing agent that was stored inside each capsule with an approximate amount of 1 mL. The detailed composition and manufacturing process of cementitious capsules can be found in [11,14].

Fig. 1. Cementitious capsules

2.2 Preliminary test: capsule survivability test

Before incorporating the capsules into the concrete, the capsule survivability test was initially performed to determine the robustness of the capsules during concrete mixing. To realise this test, two types of mixers were employed: a tilting drum mixer (Lescha SM 145 S) and a planetary/rotary pan mixer (Zyklos ZZ 75 HE). In this regard, the REF mixture was used as a preliminary test with a target volume of 20 L. The capsules were added in two different dosages per type of mixing, with a total of 20, 40, or 80 pcs in each case (see Table 2). A normal concrete mixing procedure was implemented. First, the raw materials (i.e., cement and aggregates) were mixed for 30 sec in the drum/planetary mixer. Second, the mixing water and superplasticizer were added and the mixing was continued. After 6 minutes of mixing, capsules were added into the mixer. For the survivability test with the drum mixer, the fresh mixture with the capsules was continuously mixed for another 3 minutes, while with the planetary mixer, it was only mixed for another 2 minutes. The survivability test was performed by taking the fresh concrete little-by-little from the mixer onto a sieve and then shaking the sieve under water in order to remove the fresh mortar from the aggregates and capsules. In this way, the capsules can be easily found and the number of intact and broken capsules was counted manually. A quick overview of this developed test is depicted in Figure 2. The survival ratio was calculated as the total number of intact capsules over the total number of originally added capsules in percentage.

Fig. 2. Capsule survivability test

2.3 Modified mixing and casting procedure for capsule-based mixture

A concrete volume of 35 L was aimed both for REF and CAPS mixtures. Especially for the CAPS mixture, a new mixing process was developed. Initially, all dry materials (i.e. cement, sand, gravel) were put into the drum mixer and were mixed for 30 s. Mixing water was then added into the mixer and after 2 minutes of mixing, the mixer was stopped and the superplasticizer was added. Next, the mixing process was continued. After 6 minutes of mixing, a big portion of the fresh mix (~20 L) was poured into a bucket and around 15 L of fresh mix remained in the mixer. Fifty-two capsules (corresponding to 2 vol% of gravel in a 15 L fresh mixture) were added into the mixer and the leftover mix (~15 L) was mixed again for 90 s. Hence, the fresh mixture containing capsules was tested by means of slump and air content tests. Fresh concrete from the bucket was then prepared to be combined with capsules and cast into moulds. Three cube moulds (150 mm in side), three cylindrical moulds (Ø100×200 mm) and three prismatic moulds (100×100×400 mm^3) were prepared to cast the capsule-based concrete. It was initially calculated that 2% of capsules by the volume of gravel corresponds with 14 capsules per cube specimen and 6 capsules per cylindrical specimen. A small amount of fresh mix was taken from the bucket into a bowl and depending on the volume of the designated specimens, the desired correct number of capsules was added into the bowl. Next, the fresh mix and capsules in the bowl were manually mixed by a scoop until a homogeneous mixture was obtained which was then poured into the mould. The fresh mix with capsules was compacted by a vibrating table. In this way, a random distribution of capsules was attained. This method is proposed to have the target amount of capsules per specimen. A preliminary test also showed that the capsules were completely intact after concrete mixing in the drum mixer, thus this method can be used to simulate the application of capsules in the concrete and to understand the effects of capsules toward mechanical performance of hardened concrete.

For the prismatic specimens, the capsules were manually placed into the moulds by the following procedure:

- the first layer of fresh mix was poured into the mould with a height of approximately 40 mm,
- on top of the first layer, three capsules were placed in the middle span of the mould,
- the second layer of fresh mix was poured into the mould until reaching a height of 60 mm from the bottom of the mould,
- three capsules were placed again in the middle span of the mould and finally, the fresh mix was poured until it fully covered the entire mould.

The cross section of the prism can be found in Figure 3. Unlike a random distribution of the capsules in cube and cylindrical specimens, the prismatic specimens were not aimed to assess the mechanical performance of the concrete. It was targeted to assess the ability of capsules to break during cracking and at the same time to evaluate the sealing properties. As the point of interest here is a crack that will be generated in the middle span of the prism, it was decided to manually place the capsules instead of adopting a randomly placement. In order to keep the same condition, the REF mixture was cast in the same way as the CAPS mixture but no capsules were added. One day after casting, the specimens were demoulded. The cube and cylindrical specimens were stored in a water tank at a temperature of 20±2 °C, while the prismatic specimens were stored in a curing chamber at 20±2 °C and 60% RH.

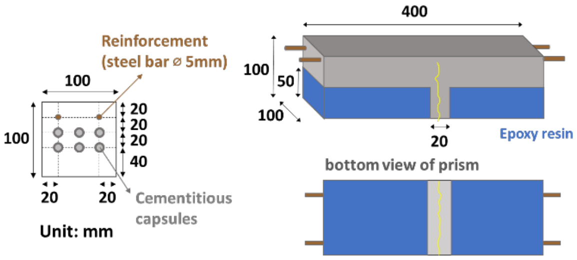

Fig. 3. Schematic design of the capsule-based prism

2.4 Test methods

The fresh properties of concrete were assessed by means of slump, air content and fresh density tests (EN 12350-2,6,7), while the hardened properties of concrete were evaluated by means of compression and tensile splitting tests at 28 days (EN 12390-3,6). An ultrasonic pulse velocity (UPV) test based on EN 12504-4 was also conducted on the hardened cube specimens to evaluate the compactness of the specimens with and without the presence of capsules. During the UPV test, the transmitter and receiver were always arranged in the same way per each specimen, either REF or CAPS, i.e. the UPV probes were placed at the center of two opposite faces of cube, and the wave propagation direction was perpendicular to the casting direction. The prisms were subjected to the three-point bending test to induce a crack at the age of 14 days. During the three-point bending test, the loading was manually controlled by the user with a relatively slow loading rate and it was directly unloaded as soon as a crack occurred. Since the rebars were embedded 20 mm from the top of the prisms, the specimens did not experience a sudden failure and after unloading, the crack width slightly reduced due to the rebar relaxation. Then, the cracked prisms were stored in an oven at 40°C for 10 days until constant weight was achieved. Constant weight was considered to be achieved when the change in mass over a period of 24 hours was less than 0.2% [15]. Next, the sides and the bottom of the specimens were partially covered with epoxy resin (Episol Designtop SF) as shown in Figure 3. The prisms were later returned to the oven for 3 days and the capillary water absorption test was finally conducted. A small area around the crack (20 mm in width) was left uncovered which was assigned as the contact area for this test (see Figure 3). The specimens were immersed in water with the water level 3 mm above the bottom surface of the prism. The weight of each specimen was recorded after 10, 20, 30, 60, 90, 120, 180, 240, 360, 480, and 1440 minutes. During the capillary water absorption test, it was ensured that the water level remained constant. As a note, the capillary water absorption test was performed on 3 uncracked reference prisms (REF_UNCR), 3 cracked reference prisms (REF_CR), and 3 cracked capsule-based prisms (CAPS_CR). In addition, the water droplet test was later executed on the split cracked specimens. After performing the capillary water absorption test, all prisms were deliberately cracked until failure, resulting into two parts of prisms per specimen. One part of the specimen was taken and the cross section was tested by the water droplet test. The droplet test was performed by releasing water droplets from the pipette to the crack surface (entire area of the cross section), and the water droplets were remained for approximately one minute. Next, the crack surface was captured by the camera and the area where the water droplets were not absorbed by the matrix indicates a sealed area and the wet area where the water droplets were absorbed by the matrix indicates an unsealed area.

3 Results and discussions

3.1 Capsules robustness

The results from the capsule survivability test were summarized in Table 2. It was shown that a 100% survival ratio of capsules was achieved during mixing the capsules in a drum mixer, while in case of using a planetary mixer, the survival ratio was attained in the range of 70–95%. A lower survival ratio in a planetary mixer occurred due to the high shear mixing force which eventually damaged a few capsules. Nevertheless, the capsules were considered robust enough to resist the mixing forces. As a proof-of-concept, in order to guarantee no single capsule was broken during the mixing process, the drum mixer was further opted for the final REF and CAPS mixtures.

Table 2. Capsule survivability (results based on a single test)

Concrete mixer		**Drum mixer**		**Planetary mixer**	
Concrete volume (L)		20	20	20	20
Number of added capsules (pcs)		40	80	20	40
Number of capsules after mixing – manual counting (pcs)	Intact capsules	40	80	14	38
	Broken capsules	0	0	6	2
Survival ratio (%)		100	100	70	95

3.2 Fresh and hardened properties

The fresh properties of REF and CAPS concretes were summarized in Table 3. The slump of REF concrete was 130 mm, while CAPS concrete had a lower slump of 100 mm. Nevertheless, both slump results were categorized in the S3 slump class. The introduction of capsules increased the air content of the fresh mixture from 3% (REF) to 4% (CAPS). This may be due to the fact that the presence of capsules that disturb the packing of the concrete materials (especially on the packing of aggregates). The fresh densities of both mixtures were identical regardless the addition of capsules.

Table 3. Fresh properties (note: single measurement for slump and air content tests)

Fresh properties	**Unit**	**REF**	**CAPS**
Slump	mm	130	100
Air content	%	3.0	4.0
Fresh density	kg/m^3	2328 ± 14	2329 ± 6

Figure 4 illustrates the mechanical performance of REF and CAPS concretes and the evaluation of UPV results. The REF concrete had a compressive strength of 42.9 MPa and the introduction of 2% capsules caused an 8% strength reduction. Statistical analysis by a t-test revealed that the difference of compressive strengths between REF and CAPS concretes was statistically significant (p-value = 0.002 < 0.05 as significance level). Interestingly, the tensile splitting strength of both mixtures had the same value of 4.1 MPa. Additionally, the UPV of hardened concretes with and without capsules had identical values at roughly 4810 m/s. It can be concluded that the addition of capsules mainly affects the compressive strength and no notable effects are found on the tensile splitting strength and the matrix compactness. It may be attributed to the effect of the random capsule distribution inside the concrete. One possible reason for strength reduction was due to the packing disturbance and the capsules were regarded as 'weak' spots in the matrix.

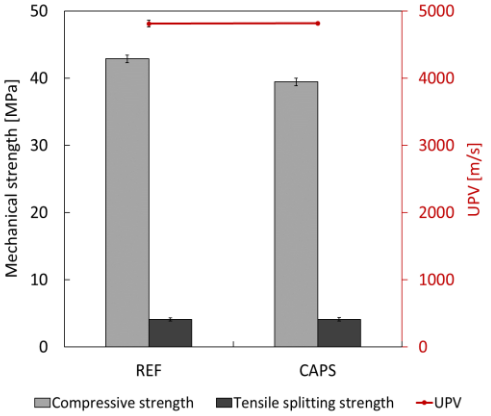

Fig. 4. Hardened properties of the REF and CAPS concretes (notes: n = 3 for compression tests, n = 3 for tensile splitting tests and n = 3 for UPV tests)

3.3 Self-sealing properties

The 14-days-old prisms were cracked by means of the three-point bending test. In case of the REF concrete, the average crack width of three cracked prisms was 135 ± 10 µm. The CAPS concretes were also cracked, but the average crack width (184 ± 17 µm) was slightly higher than for the REF. When the CAPS prisms were cracked, a release of sealing agent was observed as shown in Figure 5. This confirms that the capsules were successfully ruptured during the cracking stage. In this condition, the released water-repellent agent was seen to be rapidly absorbed by the concrete matrix in the crack zone. In order to evaluate the sealing ability, the capillary water absorption was performed on uncracked and cracked specimens. Figure 6 shows the relationship between the water uptake and the water immersion time during the test. Based on the REF concretes, it was clear that the presence of a crack caused a significant increase of the water uptake with increasing immersion time with respect to the uncracked specimen. With the cracked CAPS prisms, the progress of water uptake over time was considerably low and it was less dramatic than cracked REF prisms. Nevertheless, the water uptake of cracked CAPS prisms was slightly higher than the uncracked REF prisms. It proves that the released sealing agent seals the concrete matrix by preventing the penetration of water via the crack. According to Figure 6, the sorption coefficients (*S*) were recorded at 1.18, 6.98 and 1.78 kg/m^{2}h$^{0.5}$ for uncracked REF, cracked REF and cracked CAPS concretes, respectively. The sealing efficiency (*SE*) of the CAPS concretes can be calculated by Equation (1) and it was found that 90% *SE* was achieved by the CAPS concrete. A previous study by Anglani et al. [14] also showcased a 92% sealing efficiency with the use of the cementitious capsules filled with WRA in the mortar matrix and although the definition for the SE was slightly different, the results of Anglani support the finding in this study.

$$SE = \frac{S_{REF\_CR} - S_{CAPS\_CR}}{S_{REF\_CR} - S_{REF\_UNCR}} \times 100\% \qquad (1)$$

Fig. 5. Successful rupture of embedded capsules by releasing the sealing agent through the crack

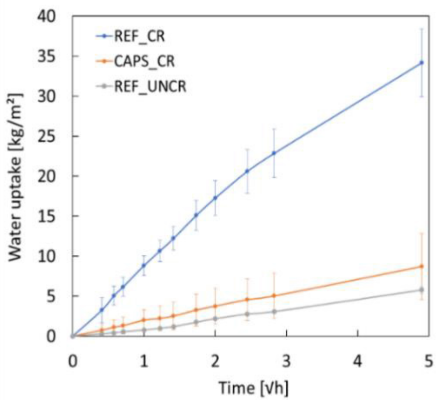

Fig. 6. Capillary water absorption results (note: n = 3 for capillary water absorption tests on each specimen series)

An additional test was performed to assess the sealing coverage area in the crack zone by water droplet test. Figure 7 clearly shows a 100% unsealed area for a REF specimen and a distinction between sealed and unsealed areas for a CAPS specimen. The sealed area represents a hydrophobic coating confirming the presence of the water repellent agent in that area. The AutoCAD software was used to measure the total area of the concrete surface and the total unsealed area. In this way, the sealed area can be quantified. It was found that based on three repetitions, the total sealing coverage area for the CAPS concretes was in the range of 80–88%.

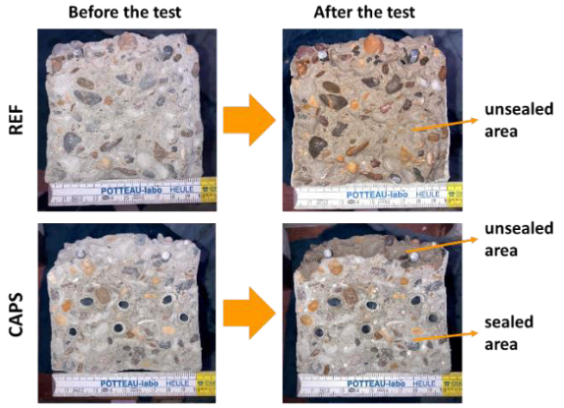

Fig. 7. Sealing coverage area based on the water droplet test

As a proof-of-concept, a qualitative assessment was made to split the cube and cylindrical specimens, which had been previously tested by compression and tensile splitting tests, respectively, in order to count the amount of broken capsules in a certain crack plane. The specimens were placed in a tensile splitting setup, and they were tested until failure, as shown in Figures 8 and 9. The number of broken capsules was counted after performing the splitting tests on three cylindrical specimens and three cube specimens containing 2% capsules. Few capsules in a certain plane of specimens broke during the splitting test. As shown in Figure 8, based on three repetitions, 2-5 out of 14 embedded capsules were present and broken in the crack plane of the cube specimens. On the other hand, as shown in Figure 9, based on three repetitions, 2-3 out of 6 embedded capsules were present and broken in the crack

Fig. 8. Splitting the cube specimens to count the amount of broken capsules in a certain crack plane (note: the darkest concrete surface is the area covered by WRA)

Fig. 9. Splitting the cylindrical specimens to count the amount of broken capsules in a certain crack plane (note: the darkest concrete surface is the area covered by WRA)

plane of cylindrical specimens. Despite the fact that not all of the embedded capsules were broken, the fracture surface covered by the sealing agent appears to be quite large with respect to the total fracture surface. This is due to the fact that a single macrocapsule can carry a considerable amount of sealing agent.

These tendencies occur due to a random distribution of the capsules inside concrete, thus the location of the capsules is not concentrated in one specific location. For the CAPS prisms, the capsules were deliberately placed in the middle span of the specimens, thus when they were split (see Figure 7), all embedded capsules were broken. This observation implies that the distribution/placement of the capsules plays a key role on the successful capsule breakage when a crack penetrates into the concrete. Pros and cons related to the capsule placement are discussed below:

- In the case of a random distribution of capsules in the concrete, it is not guaranteed that all capsules can be broken when a single crack is introduced. It strongly depends whether the capsules are present in the crack plane. However, it can also be beneficial if the cracks occur in different locations. It will open a high possibility of 'in situ' repair where the capsules in random locations are broken and the release of sealing agent may occur in many places.
- In the case of a specific placement of capsules in the concrete (like CAPS prisms), all capsules are most-likely at the 'right' place when a single crack is introduced. However, if cracks occur in any other location in which the capsules are not present, the 'in situ' repair will not be achieved. This concept of a specific capsules placement might be useful in case the cracks are predicted to occur in a specific location, thus the specific capsules placement can be strived to allow a local repair.

4 Conclusions

This study aims to investigate the introduction of macrocapsules in the concrete production by evaluating the capsules robustness, mechanical and self-sealing properties of hardened concrete. Cementitious capsules with a length of 54 mm and an outer diameter of 9 mm were used and the water-repellent agent was stored inside the capsules. The dosage of capsules was fixed at 2% by the volume of coarse aggregates. Based on this study, the key findings are summarized as follows:

1. The cementitious capsules were found to be robust to resist the mixing forces with 100% survival ratio when tested in a drum mixer and 70–95% when tested in a planetary mixer.
2. The addition of capsules slightly reduced the slump value but resulted in a higher air content.
3. A reduction of compressive strength by 8% was found when the capsules were introduced at 2% by the volume of coarse aggregates which potentially occurs due to the disturbance of the packing and the presence of capsules could be 'weak' spots in the concrete matrix. Furthermore, the presence of capsules did not alter the compactness of concrete matrix and the tensile splitting strength of concrete.
4. Based on the capillary water absorption test, the water uptake of the cracked CAPS specimens was almost as low as the water uptake of the uncracked REF specimens. This occurred due to the released water-repellent agent from the capsules that sealed the crack. A 90% sealing efficiency was achieved for the capsule-based concrete with the sealing coverage area of 80–88%.

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