MATEC Web of Conferences **378**, 03001 (2023) https://doi.org/10.1051/matecconf/202337803001
SMARTINCS'23

Self-Waterproofing Performance Of Repair Mortars With Inorganic Healing Agents

Padmapriya Arul Kumar[1]* *Sripriya* Rengaraju[1] and *Abir* Al-Tabbaa[1],

[1]Department of Engineering, University of Cambridge, United Kingdom

Abstract. In Europe, about 55% of concrete bridges are about 50 years old and require non-structural rapid repair strategies to reinstate the aesthetic and durability performances. Existing strategies focus primarily on superficial restoration that continues to demonstrate premature deterioration due to inevitable micro-crack formations that further propagate to macro-cracks leading to the ingress of moisture along with harmful ions. In this study, the benefits of self-healing technology to control moisture ingress at the microscale were investigated. For this, tailored microcapsule with inorganic healing agent, specifically, commercially available water-repellent agent (SIKAGARD 705L) was added to mortar with two types of commonly used binders namely CEMI 52.5N and CEMI 52.5R. The compatibility assessment in terms of capsule integration, fresh and hardened properties was done. The baseline healing efficiency of the mortars without any healing additions was obtained to understand the autogenous healing capacity of the reference mortars. Subsequently, the reference mortar mixes were compared with mixes containing varying fractions of microcapsules (3, 5, and 10%) for autonomous healing efficiency with capillary absorption as the main durability function. The healing efficiency was further investigated for two different crack mouth widths (<250 µm and >350 µm); representative of non-structural residual crack widths. In mortars with microcapsules, a maximum reduction of sorptivity coefficients up to 82% and 78% with CEMI 52.5N and CEMI 52.5R mortars, respectively, for specimens cracked after 7 days of curing was observed. Subsequently, a synergetic effect of autogenous healing action and autonomous water-repellent action for durability recovery was identified and proved useful for repair mortar applications. The healing agent investigated, capsule content, and healing environment considered in the current study lay a foundation for further optimisation to improve the performance and to suit different applications.

1 Introduction

Repair mortars are commonly used cementitious systems either to replace deteriorated concrete or to restore a lost portion of the concrete. Their functionalities are primarily aimed at restoring the basic mechanical and durability properties of the deteriorated elements to prevent further damage to the underlying substrate.

The global cost of repair mortars is estimated at USD 2.1 billion in 2019 and is predicted to rise further. Despite the established desired characteristics of existing commercial repair mortars, the redundancy of their application i.e., the need to re-repair systems has been identified as the reason for high costs. Furthermore, corrosion has also been observed as the commonly reported mortar and concrete deterioration in highways and the pavement industry with repair mortars in place [1,2]. Furthermore, a study revealed that about 20% of the in-place repair mortars have failed within their early periods i.e., 5 years indicating pre-mature damage [3–6].

Most of the current solutions, continue to tackle these damages at a superficial level, i.e., focusing on mechanical enhancement, primarily bond strength, rather than addressing the underlying cause. This being the formation of microcracks due to the inherent heterogeneous nature of cementitious systems, which leads to ingress of harmful ions through transport of moisture. Ultimately causing macro-crack development when left neglected by extent to affect their serviceability properties. While microcrack formations are inevitable, the potential to control moisture and harmful ion ingress without external interventions is desirable.

Furthermore, the existing solutions have limited feasibility in terms of hard-to-reach applications such as tunnel linings and bridges where damage ratification is elusive. Additionally, these repair methodologies incur high costs of repair due to repetitive installation. Thus, to address these challenges, an approach to develop a one-time application or repair mortar systems that are capable of self-detecting and self-repairing is identified through self-healing technology.

In this paper, the self-healing mechanism to control moisture ingress was explored using encapsulated water-repellent agents which hold the ability for instant mobilisation of appropriate reactants upon fracture and continue to render default functionalities of repair mortars specifically durability restoration.

---

* Corresponding author: pa456@cam.ac.uk

2 Materials and Experimental Work

2.1 Materials and mix design

Portland Cement (PC) is by far the most commonly used binder material in the formulation of repair mortars, owing to preferred compatibility with underlying PC-based concrete substrate. Further, the existing PC-based repair mortars systems require smart functionality of self-healing, and thus, in this study, two different PC-based cement binders are investigated, namely commercial portland cement CEMI 52.5N with a normal rate of hydration and CEMI 52.5R; a rapid hardening PC cement. Both cement types are supplied following BS EN 197-1[7]

CEMI 52.5N, hereafter indicated as N, was chosen as the reference system and has been studied for its self-healing property in previous literature[8,9][10]. The CEMI system will be compared to CEMI 52.5 R, hereafter indicated as rapid hardening cement (R). R cement was chosen for comparison that is currently used in conventional repair solutions for highways and bridge applications. Also, this type of cement was included to investigate the self-healing properties, to overcome the shortcomings of rapid hardening cement such as shrinkage crack-induced water permeability.

The mortar systems were designed with water to cement ratio of 0.4 along with a binder-to-fine aggregate ratio of 1:3. Consecutively, microcapsule dosages of 3, 5, and 10% by weight of binder were investigated. The microcapsules were dispersed to the mixing water that was subsequently added to the dry mortar mix to ensure homogenous distribution.

Table 1. Water-repellent microcapsules characteristics

Property	Description
Shell	Polyurethane
Cargo	Alkoxy Silane(Water repellent agent)
Average size (D_{50})	88.5 μm

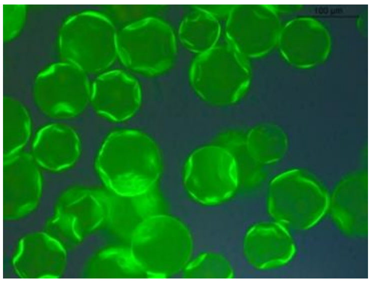

Fig. 1. Water-repellent microcapsules as observed under UV light

The self-healing technology used here is by addition of microcapsules containing water-repellent agents as seen in Fig. 1. The microcapsules are thereby intended to rupture upon cracking of the mortars and subsequently release the healing agent. The characteristics of the microcapsules are described in Table 1. The capsules were produced by the cost-effective method of membrane emulsification interfacial polymerisation.

2.2 Experimental Methodology

2.2.1 Compatibility evaluation

To evaluate the effect of microcapsule additions on the repair of mortar properties, a compatibility evaluation was conducted. According to BS EN 104, the basic criteria to be fulfilled for repair mortars entitle fresh and hardened properties, such as workability and strength respectively [11].

To assess how the microcapsules integrate with the repair mortar mixes during its fresh state, a flow-table study (Fig. 2) complying with EN 1015-3 was carried out [12]. The diameters of the mortar spread were measured at 90˚ and 180˚and mean diameter was measured. Based on the flow value, the consistency of the mortar was categorised according to EN 1015-6 [12].

Fig. 2. a) Flow-table set-up b) Compressive strength test

The effect of microcapsule additions on the mechanical characteristics was tested. A compressive strength evaluation was conducted on all mixes with specimen dimensions of 40 x 40 x 40 mm in line with EN 196-1 standard. A loading rate of 2400 N/s and maximum load capacity of 250 kN was taken as input parameters in CONTROL Advantest9 machine as in Fig. 2(b). To understand the effect of microcapsules on strength development, the age of specimens to be tested was chosen as 1, 3, 7, and 28 days from the time of casting. All specimens were tested in triplicates unless mentioned otherwise.

2.2.2 Healing efficiency evaluation

To evaluate the durability recovery performance with and without healing additions, the sorptivity test was carried out. The sorptivity measure of a cementitious system is defined as its ability to absorb or desorb a fluid by capillary action. The testing protocol was adopted from RILEM TC 116-PCD guidelines to measure permeability of mortars to suit cracked specimen configuration along with dimensional relevance [13]. Thus, for this investigation cylindrical mortars of 50 mm diameter and 40 mm thickness were cast and cracked for assessing the healing performance.

The cylindrical specimen configuration was chosen where the specimens were subjected to direct compressive load rate of 150 N/s until failure. Once the cylinders were split apart, they were reinforced with zip ties to hold them together and tightened to maintain the crack width. Further, two different crack mouth opening sizes were investigated. To obtain a wider and uniform crack mouth, a silicone tape of uniform thickness was used at the extremities as seen in Fig. 3(b). This was observed under the optical microscope and measured to be 200 ± 50 µm for smaller crack width specimens and 300 ± 50 µm for larger crack width specimen category.

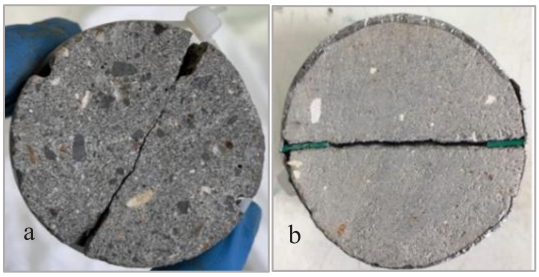

Fig. 3. Discs with a) smaller cracking width (<250 µm)

b) larger crack width (>350 µm)

The test procedure involved obtaining the capillary absorption coefficient before cracking, immediately after cracking at 7 days, and the end of the healing period. Specimens after crack induction were exposed to healing environment of 98% relative humidity for 3 days and were tested subsequently for healing performance.

The time taken for the capsule cargo. i.e., water-repellent agent to undergo polymerisation and form the healing product was under 5 hours under completely dry conditions as per product specifications [14]. Thus, to evaluate the benefits of this instantaneous product formation with a limiting environment of high humidity, a healing period of 3 days was chosen.

Prior to testing period, the specimens were allowed to air dry for 24 hours at ambient temperature at 50% relative humidity. Following this, the lateral surfaces of the specimens were sealed with adhesive aluminium foil with only the cracked surface exposed as seen in Fig. 4. This was done to ensure direct capillary absorption through the cracked region.

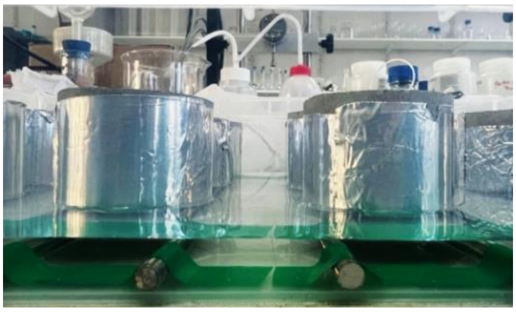

Fig. 4. Specimens sealed laterally with aluminium foil and partially submerged in water for capillary absorption.

The specimens were then placed into a container with water up to 10 mm in height, with a crack mouth facing downwards as observed in Fig. 4. Subsequently, weight change at intervals of 10 min for up to 4 h and 16 min were monitored sorptivity coefficients were then calculated by a slope of the curve between weight change per unit area versus squared root of the interval time [15].

3 Results and Discussion

3.1 Compatibility Evaluation

3.1.1 Fresh properties

Fig. 5 presents the effect of varying capsule addition on flowability in mortar mixes. In N-mortars, minor alterations for workability with the increase in capsule content were observed. The variations appeared to have a linear increase in flowability, with considerable difference between the reference mix i.e., without capsule addition, and with 10% capsule content. In all addition levels the workability (<200 mm) was classified as plastic consistency, desirable for repair overlay applications. This trivial effect of capsule additions indicates that the effect of increased volume fraction was insufficient to obstruct flow in N-mortars. Subsequently, the morphology of capsule shells i.e., smooth, and large particle size indicating lower surface area translating to lesser adsorbed water is expected to yield in flow of N-mortars with capsule additions [16].

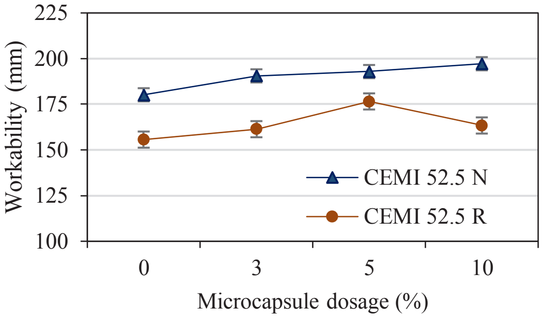

Fig. 5. Workability flow values for repair mortars

A similar increasing flow trend was seen for R-mortars until 5% capsule addition although, a drop at 10% capsule content for rapid hardening mortars was also observed. This phenomenon is indicative of a crowding effect, whereby an increase in solid contents results in increased obstruction for movement in the fluid media[17,18]. This effect is furthermore clear in CEMI 52.5 R mortars, which could be a result of accelerated hydration product formation witnessed by rapid hardening cement, complementing the crowding effect of capsule addition.

3.1.2 Hardened properties

Strength development with increasing dosage of microcapsules was obtained at 1, 3, 7 and 28 days of standard curing and presented in Fig. 6 and Fig. 7. The compressive strength observed for the N-mortar systems showed trivial change with the increase in capsule content for ages up to 7 days. The variations appeared more pronounced after 28 days of curing, whereby a linear increase in strength was observed with increasing capsule content. Specifically, about 24% increase was observed with a 10% capsule fraction.

The increase in strength after 28 days is expected to rise from improved bonds between the polyurethane shell and cement matrix. Also, the texture of shell material has potential to enable stronger contact with cement matrix, allowing strengthening of interfacial zone. Subsequently, the difference in material stiffness between microcapsule and matrix is expected to be low and hence have limited effect on strength.

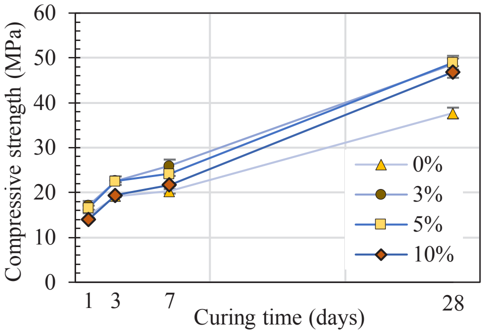

Fig. 6. Compressive strength development of N mortars

For the R-mortar system's compressive strength values, until 7-day were obtained to be inconsequentially lower than reference mortars with capsule dosages of 3 & 5% i.e., the percentage decrease was below 9%. However, this decrease was more evident for over 10% capsule content with a 17% decrease after 7 days and 20% reduction after 28 days.

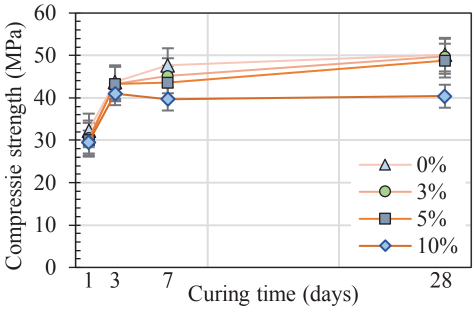

Fig. 7. Compressive strength development of R mortars

The reduction of strength is expected to emerge from a significantly lower stiffness of the microcapsule compared to the R-mortar matrix, which would demonstrate rapid strength property and subsequently result in a weaker interface between the bonded capsule and matrix system. Therefore, higher addition levels could reflect in larger strength reduction in comparison to the reference system [19]. Additionally, the gradual bond development favouring N-mortars does not appear to enhance the strength of R-mortars. The reason for the reduction despite the expected bondage could be two-fold. The first could be a result of accelerated hydration of cement matrix, causing irregular adhesion with the shell material. The second reason is expected to be due to higher shrinkage stresses, normally experienced in rapid hardening cements, that induce micro-crack formations leading to weaker interface bonds [20–24].

Previous studies conform to the same effect, where microcapsule additions with similar shell-type material and dimensions have displayed strength reductions with increasing volume fractions [25,26].

3.2 Self-healing assessment

3.2.1 Base-line healing capacity

By observing the base-line healing capacity of the mortar systems i.e., without any healing additions, repair mortars with ordinary portland cement (CEMI 52.5N) indicates a notable drop in sorptivity after 3 days of healing at high humidity with both smaller and larger crack-widths as seen in Fig. 8 and Fig. 9 . This observation is in line with previous studies which correspond to the healing mechanism by continuous hydration of unhydrated clinkers present [27–29]. On the contrary, this reduction in sorptivity was less prevalent in the case of repair mortars with rapid hardening cement (CEMI 52.5 R) irrespective of the crack widths as seen in 0% - R data in Fig. 9. The apparent higher sorptivity for 0%-R system was observed as a result of lack of unhydrated clinkers to participate in autogenous healing mechanisms arising from the speedy hydration kinetics of rapid repair mortars[30–32].

Nevertheless, a higher sorptivity coefficient for both systems immediately after cracking, and non-restorative reduction of sorptivity back to uncracked state after healing period was observed.

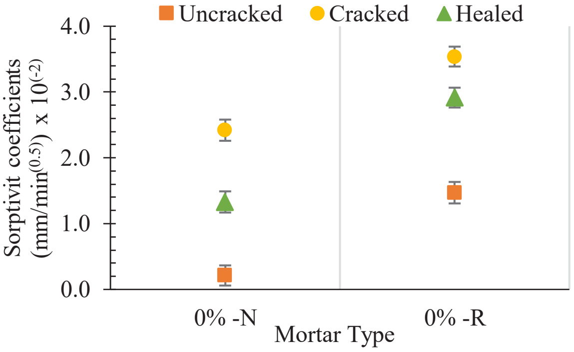

Fig. 8. Sorptivity coefficient for mortars with smaller crack width (<250 µm)

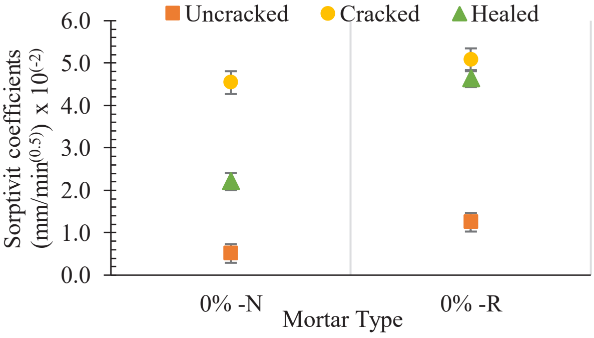

Fig. 9. Sorptivity coefficient for mortars with smaller crack width (>350 µm)

Furthermore, uptake of moisture was significant when the initial crack width was larger and subsequent healing was also observed to be impeded. The larger crack mouth thus has a more undesirable effect on rapid hardening mortars with reduction of sorptivity less than 10% after cracking. This has been deemed undesirable as it allows for transport of harmful ions hence needing an intervention for repair.

3.2.2 Water-proofing healing capacity

Further to baseline healing, capillary absorption of systems with different dosages of microcapsules after cracking and 3 days of healing is seen in Fig. 10, Fig. 11, Fig. 12 and Fig. 13. The sorptivity measurements of mortars with capsules were compared with neat mortars i.e., with 0% capsules. The mortars are hereafter referred to as percentage capsules-type of binder. i.e., 0%-N etc.

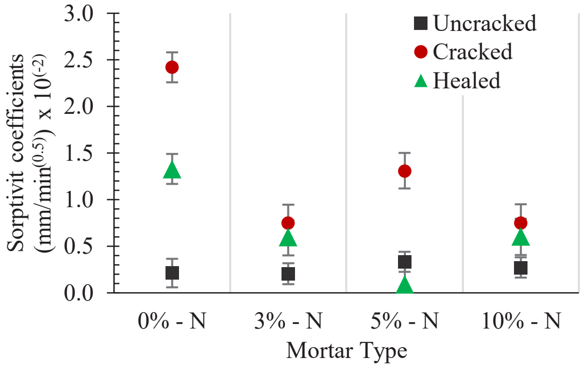

Fig. 10. Sorptivity coefficient for N-mortars with smaller crack width (<250 μm) and varying capsule dosages

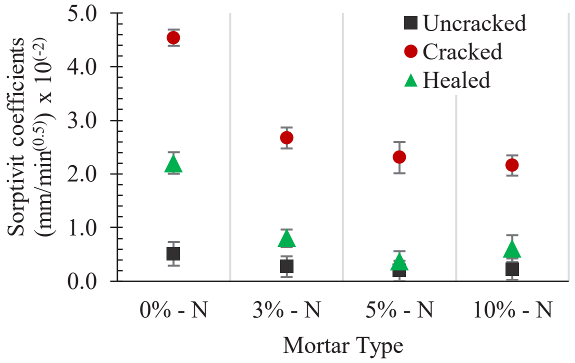

Fig. 11. Sorptivity coefficient for N-mortars with larger crack width (>350 μm) and varying capsule dosages

The healing capacity hereby indicating water-proofing ability is observed as reduced sorptivity coefficients in both cement types irrespective of crack widths. Comparing the cracked and healed coefficients, this reduction was more apparent in systems with larger crack width openings whereby the initial water uptake was higher immediately after cracking (Fig. 11 and Fig. 13).

Evidently, the action of water-repellency was observed to be immediate with a lower sorptivity coefficient at the cracked state of systems with microcapsules compared to the sorptivity of cracked systems with 0% capsules. This is expected as a response to instantaneous curing of the healing agent, the SIKA GARD water-repellent agent (WRA), within few hours. This curing mechanism of the capsules' healing agent imparts impermeability by the formation of hydrophobic alkyl groups that reduce the surface tension [33,34]. Subsequently, the silicone groups have been reported to be chemically reactive with the substrate that further controls the penetration of incoming water [35].

Furthermore, the reduction in sorptivity coefficients continued after the healing period of 3 days, for both CEMI 52.5 N and CEMI 52.5 R mortars.

This can be indicative of two synergetic healing mechanisms. The first mechanism being the action of cured WRA by beading effect whereby the water would be repelled from the hydrophobic surface. Subsequently, this run-off water tends to form a film atop the surface in turn treating the pores [33–35].

Hereby providing a way for the second mechanism of healing, whereby the water present in the capillary pores continue to take part in autogenous hydration mechanisms involving formation of hydration products. Subsequently, indicative of overlapped sorptivity coefficient in healed mortars compared to uncracked mortar coefficients.

For CEMI 52.5 N mortars, the expected healing is by ongoing hydration mechanism of unhydrated cement clinkers resulting in filling of pores.

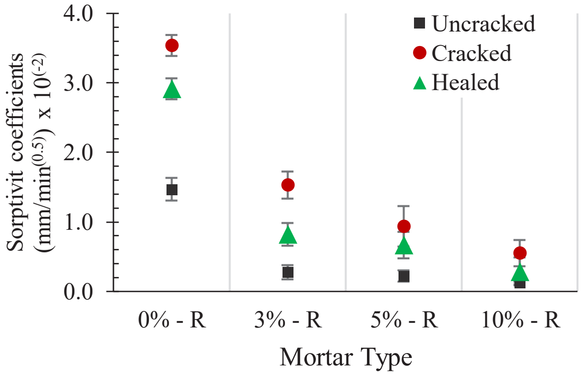

Fig. 12. Sorptivity coefficient for R-mortars with smaller crack width (<250μm) and varying capsule dosages

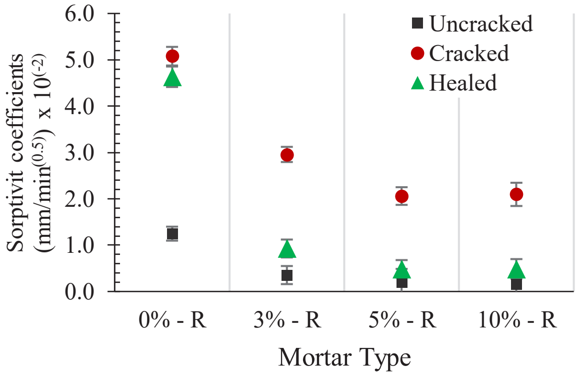

Fig. 13. Sorptivity coefficient for R-mortars with larger crack width (>350μm) and varying capsule dosages

Alternatively, for CEMI 52.5R mortars, due to lack of unhydrated clinkers by rapid hydration, the autogenous healing mechanism is expected by action of carbonation [36]. Previous studies with WRA have reported that when the agents come in contact with moisture, the water film formation caused by hydrophobic network of alkoxy, enables favourable moist conditions below the hydrophobic layer for carbonation reactions [37]. This thereby results in product formation to act as crack filler material for self-healing [36].

4 Conclusions

This paper presented the evaluation of conventional repair mortars incorporated with microcapsules. The performance parameters considered were compatibility assessment in terms of flow properties and strength. The benefit of self-healing was also evaluated in mortars with capsules containing water-repellent agent in terms of capillary absorption recovery when cracked. The following conclusions are made from the work carried out.

1. The flow properties of mortars with capsule additions were demonstrated to deter the flow of R-mortars at higher addition levels i.e., 10%, more than N-mortars owing to accelerated hydration and particle packing effect.
2. The addition of microcapsules demonstrated insignificant changes to the early age strength of both mortar systems. For N-mortars there was a small increase after 7 days for 10% dosage and decrease for R-mortars after 7 days due to stiffness difference and weak bonding of matrix and capsule as an effect of shrinkage-induced microcrack formations in shell-matrix interface.
3. The healing efficiency of mortars cracked at 7 days and healing for 3 days was witnessed by sorptivity coefficient changes. The efficiency was clearly observed after healing period with reduced sorptivity and more pronounced despite larger crack width (>350μm). The effect of microcapsule dosages had minimal effect on healing efficiency. Although no clear difference was seen between N-mortars and R-mortars at early age cracking, the mechanism identified was observed to enable water-proofing efficiency in both systems.
4. Mechanism of healing was identified as water-repellent action of healing agents when released and allowed to undergo polymerisation even in high humidity environment. Subsequently, a synergetic effect of autogenous healing was recognized to enhance the durability after crack formation.

The addition of microcapsules appeared to have notably decreased effect on both workability and strength characteristics of CEMI 52.5 R cement-based mortars 10% addition level. This response however is still considerable for patch repair mortars on class R1, R2, and R3, whereby durability performance is more important.

Thus, the obtained results demonstrate desired effect of microcapsule additions regarding durability recovery. The instant ratification of mortar systems to control moisture ingress by action of water-repellency by healing agents was observed. This attribute of microcapsules to rupture and release the healing agent, subsequently restoring functionality instantaneously upon cementitious system's crack formation, is hereby deemed desirable for rapid repair systems.

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.

The authors would like to acknowledge Claire Riordan, Ph.D. Candidate of the University of Cambridge and Micropore Technologies and Dave Palmer from Micropore for the development and supply of microcapsules to conduct this research.

References

1. M. Farzad, S. F. Fancy, A. Azizinamini, and K. Lau, NACE - International Corrosion Conference Series (2019)
2. Markets and Markets, *Concrete Repair Mortars Market - Global Forecast to 2021* (2016)
3. Conrepnet, (2007)
4. A. M. Vaysburd and P. H. Emmons, Constr Build Mater **14**, 189 (2000)
5. M. S. Ali, E. Leyne, M. Saifuzzaman, and M. S. Mirza, Constr Build Mater **174**, 159 (2018)
6. M. Lukovic, G. Ye, and K. van Breugel, 14th International Conference Structural Faults and Repair, Edinburgh, Scotland, UK, 3-5 July 2012 (2012)
7. BS EN 197-1-2011, (n.d.)
8. C. Litina and A. Al-Tabbaa, ICSHM 2013: Proceedings of the 4th International Conference on Self-Healing Materials 201 (2013)
9. T. S. Qureshi and A. Al-Tabbaa, Smart Mater Struct **25**, (2016)
10. T. Qureshi, A. Kanellopoulos, and A. Al-Tabbaa, Constr Build Mater **192**, 768 (2018)
11. BS EN 1504-4:2004, British Standard **3**, 1 (2016)
12. BS EN 1015-6, (n.d.)
13. J. Kropp, Mater Struct **32**, 174 (1999)
14. P. D. Sheet, 2 (2020)
15. J. Kropp, Mater Struct **32**, 174 (1999)
16. S. G. Sanfelix, I. Santacruz, A. M. Szczotok, L. M. O. Belloc, A. G. de la Torre, and A. L. Kjøniksen, Constr Build Mater **202**, 353 (2019)
17. M. Mooney, J Colloid Sci 162 (1951)
18. A. D. B. J. Chong, E.B. Christiansen, J Appl Polym Sci **15**, 2007 (1971)
19. X. Wang, P. Sun, N. Han, and F. Xing, Materials **10**, (2017)
20. F. Pacheco-Torgal, J. Barroso de Aguiar, Y. Ding, W. Tahri, and S. Baklouti, Handbook of Alkali-Activated Cements, Mortars and Concretes 627 (2015)
21. H. S. Folker H. Wittmann, **13**, 243 (2007)
22. A. Garcia, E. Schlangen, and M. van de Ven, Key Eng Mater **417–418**, 573 (2010)
23. A.-Ai. L. (Hasson KE, Brooks JJ, Cem Concr Compos (2001)
24. A. Barde, S. Parameswaran, T. Chariton, W. J. Weiss, M. D. Cohen, and S. Newbolds, 154p (2006)
25. A. Kanellopoulos, P. Giannaros, and A. Al-Tabbaa, Constr Build Mater **122**, 577 (2016)
26. P. Giannaros, A. Kanellopoulos, and A. Al-Tabbaa, Smart Mater Struct **25**, (2016)
27. E. S. M. De Rooij, K. Van Tittelboom, N. De Belie, Springer (2013)
28. K. van Tittelboom and N. de Belie, *Self-Healing in Cementitious Materials-a Review* (2013)

29. M. Wu, B. Johannesson, and M. Geiker, Constr Build Mater **28**, 571 (2012)
30. E. Sakai, Y. Nikaido, T. Itoh, and M. Daimon, Cem Concr Res **34**, 1669 (2004)
31. F. Edition, H. Member, and C. Society, *Neville* (1954)
32. H. F. W. Taylor, Cement Chemistry (1997)
33. P. Zhang, H. Shang, D. Hou, S. Guo, and T. Zhao, Advances in Materials Science and Engineering **2017**, (2017)
34. D. S. Oehmichen, A. Gerdes, and A. Wefer-Roehl, Hydrophobe V **218**, 205 (2008)
35. F. Li, Y. Yang, M. Tao, and X. Li, RSC Adv **9**, 7165 (2019)
36. M. Wu, B. Johannesson, and M. Geiker, Constr Build Mater **28**, 571 (2012)
37. AndA. G. J.Heinrichs, S. Schmeiser, in *4th Int. Conf. Water Repel. Treat. Build. Mater.* (2005), pp. 27–44