

# Self-healing of cementitious composites using silica precursors as microencapsulated healing agents

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## ABSTRACT

Several different healing agents and mechanisms have been put forward for use in autonomous healing of small cracks in cementitious composites. While the existing research on this topic focuses mainly on the use of polymeric materials as encapsulated healing agents, this study aims at assessing the efficiency of inorganic silica precursors as potential healing materials. Herein, poly-urethane microcapsules were developed to envelope an inorganic aqueous core. Colloidal silica Ludox 40™, an aqueous suspension of fine silica particles, was selected as the appropriate healing agent. The suspended silica particles can react with calcium hydroxide in the matrix to yield further hydration products (C-S-H gel) and fill any existing voids.

Microcapsules were synthesised in situ through a one-step interfacial polymerisation process and were subsequently dispersed in fresh cement paste. Two different mass fractions, i.e. 1 and 5% microcapsules/cement ratios, were investigated to assess the effects on both mechanical properties and healing efficiency. The self-healing capacity was evaluated by crack healing ratio and permeability reduction. Thus, three-point loading tests were performed on cracked, healed cement paste specimens, coupled with acquisition of microscopic images and gas permeability tests. The healing products were isolated and characterised through SEM, TGA and XRD.

The results indicated that the encapsulation of healing agent in polyurethane could be regarded as a promising method for realising self-healing of cementitious composites.

## 1. INTRODUCTION

Civil engineering applications have a built-in redundancy of design to structural safety under a variety of adverse events. But, over the long-term, repair and eventual replacement is inevitable. In this project, a proposed solution evolving the development of an autonomic self-healing system through the incorporation of encapsulated inorganic silica precursors. Therefore, poly-urethane microcapsules were developed to envelope an inorganic aqueous core.

## 2. MATERIALS

Commercially available monomers: diphenyl methylene diisocyanate (MDI) and 1,4-butanediol (BD), gum arabic (GA), polyethylene glycol (PEG400), colloidal silica (CS) Ludox® AS-40 and toluene were obtained from Sigma Aldrich, UK. All the solvents

were of analytical grade and were used without further purification as received. Deionised water was used for all experiments. The studied cement paste mixtures were composed of ordinary Portland cement (CEM I 52.5 N), water and a varying amount of microcapsules expressed as mass% (m%) of cement weight (Table 1). The water to binder (total solids) ratio adopted in this design was kept constant ( $w/c=0.6$  by weight).

Table 1 Studied paste samples with their code, m% of MCs, method of cracking, curing conditions and number of specimens tested.

Specimen	Reference	m % of MCs	Cracking	Curing	Number
Prism	CI	0	3-point bending	Water curing	6
	CIMC1	1			6
	CIMC5	5			6
Cylinders	CI	0	Compressive splitting	Permeability	5
	CIMC1	1			6
	CIMC5	5			6

MCs: microcapsules

### 3. METHODS

The overall preparation procedure of the microcapsules is illustrated in Figure 1. Microcapsules with aqueous core and polyurethane shell were formed as a free flowing powder (particle size  $43.5 \mu\text{m} \pm 15 \mu\text{m}$ ).

Two types of composite specimen including the control (with no particles admixed) and the microcapsule-reinforced specimens were investigated. Series used for three-point-bending tests consisted of six  $160 \times 40 \times 40 \text{ mm}^3$  prism samples. The cylindrical discs used in the permeability tests were 50 mm in diameter and 15 mm high. All specimens were de-moulded after 24h and then cured under water for additional 27 days prior testing.

By means of a displacement-controlled three-point-bending test, cracking was induced on the composite prisms at an age of 28 days. The displacement was increased until the maximum flexural capacity was reached. A large final crack of approximately  $150\text{-}200 \mu\text{m}$  was formed. Cracks of the specimens containing encapsulated healing agent were autonomously healed when allowed to cure. The healing efficiency ( $H_{\text{eff}}$ ) was estimated as the ratio of the recovered ultimate strength over the initial capacity. In between both loading periods, microscopic observations were performed.

For the determination and quantification of the sealing recovery, the gas permeability method and apparatus proposed by Alshamsi and Imran were adopted [1]. Permeability tests were carried out on cylindrical discs after cracking the specimens by a means of laterally confined splitting-controlled compressive test.

A thermogravimetric analysis (TGA) was performed with a Perkin Elmer STA 6000 to determine the composition of the white crystallisation residue found on the crack planes and to identify the healing materials. To further analyse and identify the mineralogy of the extracted healing products X-ray diffraction (XRD) was used.

### 4. RESULTS

The addition of microcapsules in the matrix appears to yield increasing flexural capacity and enhance the consistency of the mix design (Figure 3). Increased healing efficiency was observed (18%) compared to findings [2], [3] for microcapsule induced self-healing in concrete with increased production of calcite in the healing

products. Nevertheless, for less than 5% addition the strength recovery appears minimal.

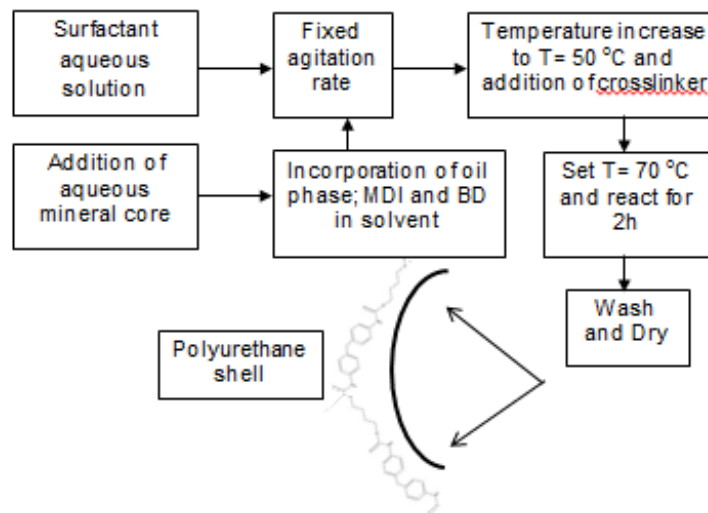


Figure 1: Microencapsulation procedure.

Gas permeability test results report decrease in permeability coefficient by 29.4% and 21.3% for samples prepared with 1 m% and 5 m% of microcapsules after 28 days of curing. The inclusion of both types of micro particles was demonstrated to reduce the gas permeability of hydrated cement composites. The reduced permeability can be attributed to the self-healing effect of the microcapsules. The observations under XRD and TGA (Figure 7) confirm that the microcracks in specimens incorporating microcapsules were healed mainly with calcium carbonate and portlandite [4]–[6].

## 5. CONCLUSIONS

The results presented in this paper show that even with limited control of the crack width, the inclusion of MCs provided cement with the ability to seal microcracks and to partly regain its mechanical properties.

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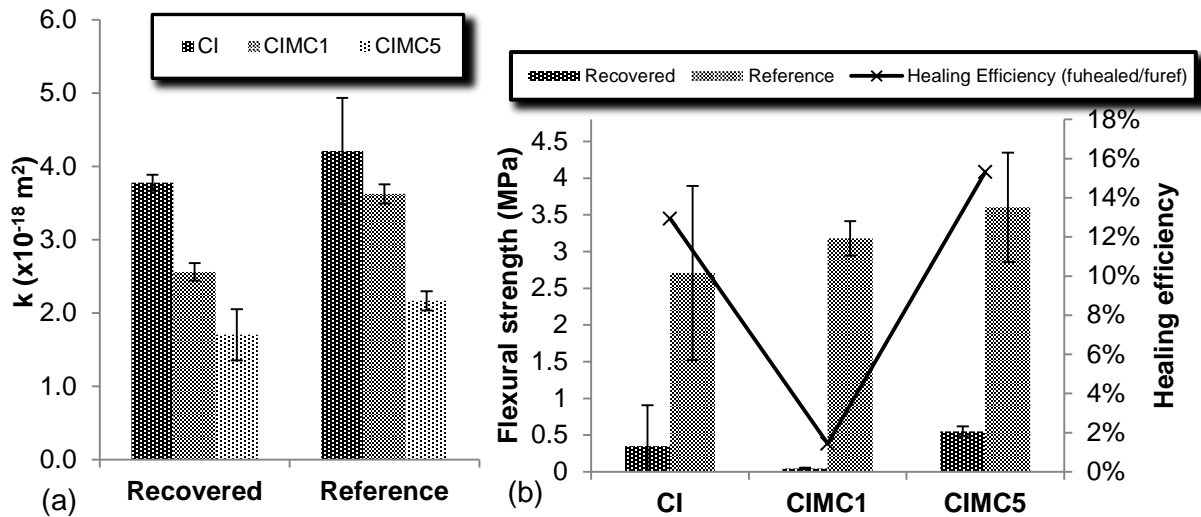


Figure 2: (a) Permeability coefficients of cement composites for cracked (reference) and healed specimens (recovered). (b) Mean values and standard deviations for peak flexural strength for virgin (reference) and healed specimens (recovered).

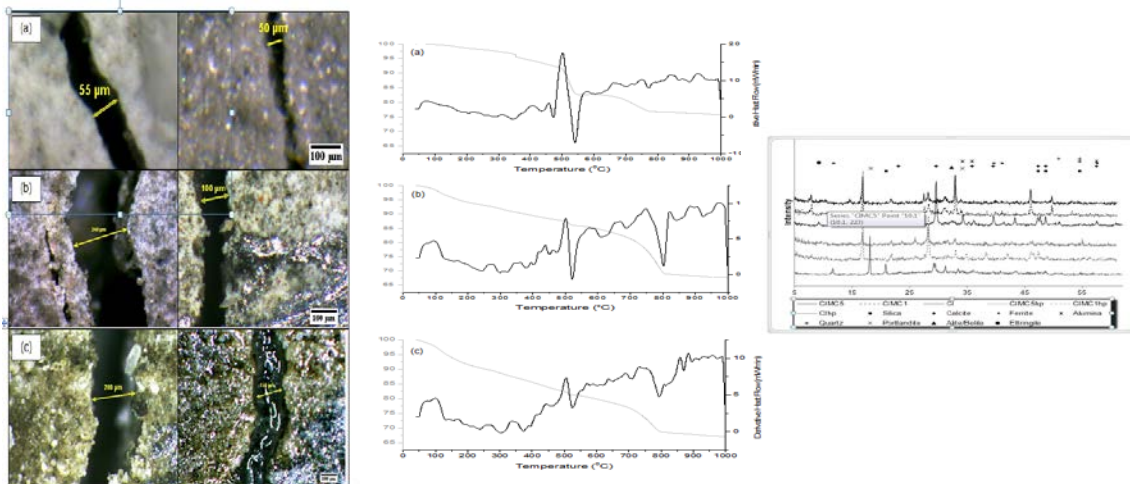


Figure 3: Healing efficiency and healing products produced at the crack opening in (a) CI without MCs, (b) CIMC1 with 1 m% of MCs and (c) CIMC5 with 5 m% of MCs. Corresponding TGA and XRD characterisation of the healing material.