Evaluation of the self-sealing efficiency of cracks in mortar by bioprecipitation or application of hydrogels

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ABSTRACT

Within the EU-FP7 project HEALCON, smart concrete with self-healing properties is developed in order to guarantee liquid tightness of concrete structures. For repair of early age cracks in concrete, non-elastic repair materials, such as calcium carbonate precipitated by bacteria, or new cement hydrates / calcium carbonate of which the formation is stimulated by the presence of hydrogels, are proposed.

Thanks to the expertise of the consortium in the field of self-healing concrete, a thoughtful selection and the development of promising techniques was possible. The healing agents presented in this paper include (i) newly developed superabsorbent polymers (SAPs) with a high swelling and pH-sensitiveness which were developed in order to minimize the uptake of mixing water during concrete manufacturing and maximize the sealing efficiency of cracks in concrete, (ii) commercial superabsorbent polymers, as a reference to judge the performance of the newly developed SAPs, (iii) micro-encapsulated *Bacillus sphaericus* spores which have the ability to precipitate calcium carbonate in the crack and (iv) a mixed ureolytic culture (MUC) which is more cost-effective than using pure cultures and can close cracks in concrete.

A comparison of the regain in liquid tightness of mortar specimens containing the different healing agents is presented. The evaluation of the self-sealing performance of the agents occurred through a water permeability test via water flow, a test method developed within HEALCON.

1. INTRODUCTION

One of the objectives within the EU-FP7 project HEALCON was to develop efficient self-healing techniques that enable concrete to regain liquid-tightness via bioprecipitation by suitable micro-organisms or application of hydrogels. In this paper, the healing products themselves and the test methods are described, together with some results, out of a larger testing program, showing the self-sealing efficiency of mortars.

2. HEALING AGENTS

At first instance, commercially available superabsorbent polymers (cross-linked copolymers of acrylamide and acrylate, called SAP in this paper) were tested within the HEALCON project to seal cracks in concrete. This is possible as superabsorbent polymers, present at the crack planes, can, upon crack formation and ingress of water, swell and block the crack. Later on, they can release their water and provide it to the surrounding cement matrix to stimulate further hydration and $CaCO_3$ precipitation. Although the results were satisfying and small cracks could be sealed quite well with these commercial products [1], the HEALCON consortium strived to obtain optimal self-sealing capacities. Therefore, a new generation of formulation of superabsorbent polymers having an improved swelling and pH sensitiveness has been developed and produced via bulk polymerization (particle size: 400 μm - 600 μm) by CEINNMAT (called SAP G in this paper). These superabsorbent polymers should (1) reduce the uptake of mixing water during concrete mixing (high pH) and thus limit macro pore formation in the hardened mortar and (2) enhance the sealing and healing capability of mortar due to an increased water absorption capacity at neutral pH.

In order to compare the swelling behaviour of the different types of superabsorbent polymers, a filtration test was performed in cement filtrate (100 g of OPC mixed with 1 L of demineralized water) and in demineralized water. These two solutions simulate respectively the situation in a concrete environment (pH ~ 13) and the situation in a crack when fresh water (pH ~7) intrudes. The results are as follows: commercial superabsorbent polymers have a swelling capacity of 308 g/g SAP in demineralized water and 38 g/g SAP in cement filtrate, while the newly developed superabsorbent polymers have a swelling capacity of 424 g/g SAP G in demineralized water and 36 g/g SAP G in cement filtrate. The absorption of water by the superabsorbent polymers from the fresh mix is thus only slightly reduced with the newly developed product, but the pH sensitiveness is enlarged. This will lead to a better sealing efficiency for the cracks, as shown in the next sections.

Besides the studies related to the development of superabsorbent polymers suitable for self-sealing, research was done on bacterial healing agents as earlier research has shown beneficial effects on self-healing by incorporation of bacterial spores [2]. First, the production of pure axenic cultures of the bacterial strain *Bacillus sphaericus* was upscaled by VTT and the micro-encapsulation process, previously developed, was fine-tuned by Devan. The final product is a slurry of spores encapsulated by a melamine formaldehyde shell (called MEBS in this paper). However, as the cost price of this product is quite high, further efforts were made by Avecom to produce non-axenic mixed cultures [3]. These mixed cultures (called MUC^+ in this paper) have ureolytic properties and are capable to precipitate $CaCO_3$ in the crack.

Mortar mixes containing the different healing agents were tested with regard to their possible regain in liquid-tightness after crack formation and healing. The mortar mixes were standard mortars according to EN 196-1, with the healing agent added on top (1 wt.% relative to the cement weight for the superabsorbent polymers and 3 wt.% relative to the cement weight for the bacterial healing agents). In table 1, the exact mortar composition can be found. The specimens (40 x 40 x 160 mm prisms) were stored in plastic foil for 28 days and subsequently, a three-point bending crack was created (aimed crack mouth width of ~ 150 μm). Healing occurred under wet-dry conditions (12 h wet / 12 h dry) for 28 days.

Table 1: Mortar mix composition (g)

	SAP	SAP G	MEBS	MUC $^+$
Cement	450	450	450	450
Water	225 + 90*	225 + 80*	196	214
Sand	1350	1350	1350	1350
Healing agent	4.5	4.5	32**	4.5
Precipitation precursor (urea)	/	/	18	18
Nutrient (yeast)	/	/	3.84	/
Ca-source ($Ca(NO_3)_2.4H_2O$)	/	/	36	36

* extra water to compensate for the uptake of mixing water by the SAP/SAP G

** 32 g of slurry corresponds with 13 g of dried microcapsules (~ 3% relative to the cement weight)

3. WATER PERMEABILITY TESTS

Two test methods to assess the sealing efficiency were proposed within the HEALCON project. The first test method evaluates the absorption of water by the cement matrix in the presence of a healed crack, and compares the sorptivity with the sorptivity of a sound specimen (best case scenario) and the sorptivity of a cracked and unhealed specimen (worst case scenario) in order to calculate the sealing efficiency. The test procedure is based on the standard method according to EN 13057, except that only a small zone of 40 mm wide surrounding the crack is exposed to the water. The second method judges whether a water flow under pressure (0.05 bar to 2 bar), supplied via an inner hole in the specimen crossing the crack, can be withstood by the products sealing the crack. In case the sealing is not perfect, water will flow out of the specimen and the amount will be measured in function of time. Comparing the water flow of the unhealed and healed crack allows to define a sealing efficiency. In this paper, only results obtained with the second test method (and pressures up to 0.05 bar) are presented. More details about this procedure can be found in [4].

4. SELF-SEALING EFFICIENCY

In Table 2, the water flow through the unhealed and healed cracks is tabulated. Water flow tests were running for 15 minutes and the average values obtained from testing 3-5 specimens with comparable crack width are reported. For the specimens with micro-encapsulated *B. sphaericus*, no specimens were available with a crack width around 150 μm and the values of the water flow are thus reported for a small crack (~115 μm) and a wider crack (~280 μm). These results clearly show that the crack width is one of the most important parameters determining the value of the water flow. Nevertheless, incorporation of *B. sphaericus* leads to an almost complete healing after 28 wet-dry cycles, even for cracks up to 280 μm wide.

Comparing then the results of the SAP, SAP G and MUC$^+$ specimens, having comparable crack widths, shows that:

(1) the water flow measured on unhealed specimens is much higher for the MUC$^+$ than for the SAP and SAP G. This is not surprising as the superabsorbent polymers available at the crack faces can absorb water and swell and thus lead to an immediate (partially) blocking and sealing of the crack. The immediate

sealing effect is, for the measurements reported here, slightly better for the SAP than for the SAP G.

(2) the water flow after healing has been decreased a lot in comparison to the values before healing, leading to a quite high healing efficiency. These results correspond quite well with the microscopic analyses done on the specimens, showing (almost complete) closure of most of the cracks.

Finally, as reported in [5], the autogenous healing capacity of mortars is, for crack widths of around 160 μm, quite small (~ 50%), indicating the beneficial effect of the incorporation of healing agents to seal wider cracks.

Table 2: Results of the water flow tests (average values of 3-5 specimens)

	Crack width (μm)	UNHEALED Water flow (15 min) (g)	HEALED Water flow (15 min) (g)	Healing efficiency (%)
SAP	~ 140	3.9**	0.5**	87**
SAP G	~ 150	16.4	0.3	98
MEBS	~ 115*	2.0	0	100
	~ 280*	612.7	22.9	96
MUC+	~ 145	50.2	1.3	97

* result of only one specimen due to a large variation in crack width between the specimens
** correction

5. CONCLUSIONS

Within this paper, some results of a larger testing program were reported, showing that the healing products developed and optimized within the HEALCON project performed well related to the regain of liquid tightness of specimens containing cracks of ~ 150 μm.

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