An overview on the research on self-healing concrete at Politecnico di Milano

Liberato Ferrara[1,*]

[1]Department of Civil and Environmental Engineering, Politecnico di Milano, Italy

Abstract. Self-healing cement based materials, by controlling and repairing cracks, could prevent "permeation of driving factors for deterioration", thus extending the structure service life, and even provide partial recovery of engineering properties relevant to the application. The author's group has undertaken a comprehensive investigation focusing on both experimental characterization and numerical modelling of the self-healing capacity of a broad category of cementitious composites, including high performance cementitious composites reinforced with different kinds of fibres. Both autogenous healing has been considered and self-healing engineered techniques, including the use of pre-saturated natural fibres and of crystalline admixtures. Tailored methodologies have been employed to characterize the healing capacity under different exposure conditions and for different time spans, ranging up to two years. The healing capacity has been quantified by means of suitably defined "healing indices", based on the recovery of mechanical properties correlated to the amount of crack closure, measured by means of optical microscopy. A predictive modelling approach, based on modified micro-plane model, has been formulated. The whole investigation represents a step towards the reliable and consistent incorporation of self-healing concepts and effects into a durability-based design framework for engineering applications made of or retrofitted with self- healing concrete and cementitious composites.

1 Introduction

The porous structure of concrete is one of the main causes of its being prone to degradation; even if it is generally accepted that a well-proportioned and properly cured concrete, produced using a low water-to-cement (w/c) ratio, leads to obtain a finished product with good durability performance, no concrete material and structure can be made absolutely waterproof. Because of the porous structure of concrete, water can penetrate through pores and micro-cracks, due to either capillary absorption and/or hydrostatic pressure. Concrete professionals typically adopt the term "permeability" to describe the resistance of concrete to water ingress and/or passage under actual service conditions. This includes the overall permeability issue of concrete including possible existing cracks, due, e.g., to restrained drying shrinkage, thermal deformations as well as sustained service loads.

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[*] Corresponding author: liberato.ferrara@polimi.it

In view of the aforesaid statements, it would be highly desirable and advantageous to be able not only to "design" and cast a concrete as compact (and impervious) as possible, but also to be capable to provide the material some capacity to reduce its permeability in the cracked state. In this framework, (autogenous or engineering triggered) self-healing cement-based construction materials, able to control and repairing early stage cracks, would represent an exceptional asset to the 21st Century Civil Engineering. Self-sealing/healing of cracks could prevent permeation of driving factors for deterioration and even provide a partial recovery of the engineering properties of the material relevant to the intended applications, thus contributing to extend the service life of the engineering works [1,2].

Discovered as early as in 1836 by the French Academy of Science, and attributed to either delayed hydration of cement/binder, carbonation or the combination of both, many researchers demonstrated that concrete, under favourable conditions, can heal naturally without any particular additives [1,2]. As a matter of fact, because such a capacity turned out to be quite randomly scattered and thus neither reliable nor predictable in an engineering application perspective, a paramount effort is currently challenging the concrete research community of investigating "engineering" self-healing capacity of concrete and cement-based construction materials. Several different techniques, along different main directions of investigation, have been conceived and explored: i.e., self-healing engineered with fibre reinforcements, mineral-producing bacteria, super absorbent polymers, healing agents contained in shell and tubular capsules and other proprietary chemical admixtures or even mineral additions already traditionally employed as cement substitutes. The supply of water (moisture) is essential, especially in the case of addition of chemical agents able to promote the deposition of crystals inside the crack, but since most infrastructures are exposed to rain or underground water, usually this is an easily satisfied requirement. Several other variables can affect the phenomenon of self-healing, such as the mix proportions, the stress state along the cracks, thermo-hygrometric conditions etc. [1, 2].

The authors' research groups have undertaken for about a 5 years a collaborative comprehensive research project, focusing on both experimental characterization and numerical modelling of the self-healing capacity of a broad category of cementitious composites, ranging from normal strength concrete to high performance cementitious composites reinforced with both steel and natural fibres. Both autogenous healing capacity and self-healing engineered techniques have been investigated, including the use of pre-saturated natural fibres as well as of crystalline admixtures, including their synergistic effects. Tailored methodologies have been employed to characterize the healing capacity of different investigated cement-based materials, which are based on a comparative evaluation of the mechanical performance measured through 3- or 4- point bending tests or indirect tension tests. Tests have been performed on pre-cracked specimens (to a target value of the crack opening), after scheduled conditioning times to selected exposure conditions (e.g., water immersion, wet and dry cycles, humid exposing and dry climates). The healing capacity has been quantified by means of suitably defined "healing indices", based on the recovery of the permeability and of the mechanical properties, and correlated to the amount of crack closure, measured by means of optical microscopy and also "estimated" through suitable indirect methodology. Chemical characterization of the healing products has been performed to understand the different mechanisms governing the observed phenomena. As a further step, predictive modelling approaches, based on either a poro-plasticity and fracture approach or on a modified micro-plane model, have been formulated. On the one hand, a zero-thickness interface model has been proposed to include the effects of crack healing, through suitably defined porosity-based functions which affect the time evolution of mechanical properties of concrete. On the other hand, a micro-plane model incorporates the self-healing effects through a delayed cement hydration rule and the effects of cracking on the diffusivity and the opposite repairing effect of the self-healing.

2 Experimental programme: materials and methodology

Employed mix-designs are summarized in Table 1.

For Normal Strength Concrete (NSC) the self-healing evaluation methodology employed unnotched prismatic beam specimens (500 mm × 100 mm × 50 mm) pre-cracked in 3-point bending up to two different crack opening levels, respectively equal to 0.15 and 0.30 mm, and then either stored in water or left exposed to open air up to 12 months. 3-point bending tests were repeated up to failure after scheduled conditioning periods. Self-healing was thus evaluated by comparing the peak-load bearing capacity exhibited by the specimens after conditioning to the residual one the same specimens featured upon pre-cracking when unloaded at the prescribed crack opening [3].

For steel- and steel-sisal fibre reinforced High Performance Cementitious Composites [4-8] beam specimens of 500 mm x 100 mm x 30 mm were employed, cut from larger cast slabs (1m × 0.5 m). Slabs were cast taking advantage of the self-compacting features of the fresh mixtures, also in order to achieve a tailored alignment of the fibres, and beam specimens were cut with their axis either parallel or orthogonal to the casting flow direction (i.e. the preferential fibre alignment), in order to discriminate the effect of flow inducing fibre alignment on the performance of the HPFRCC. Specimens were pre-cracked in 4-point bending (4pb) and the Crack opening Displacement (COD), was measured by means of two LVDT transducers over a gauge length equal to 200 mm. Specimens featuring a deflection softening response (i.e. with fibres perpendicular to the beam axis) were pre-cracked up to a COD value equals to 0.5 mm. On the other hand specimens with fibres parallel to the axis, featuring a deflection hardening response, were pre-cracked in the post-peak regime equals to (COD_{peak} + 0.5 mm), where COD_{peak} denotes the measured value of the COD in correspondence of the peak stress, under the assumption of the same opening of the localized crack. After, pre-cracking specimens were "conditioned" in four different "environments": (i) immersion in water; (ii) dry (20°C and 50% RH); (iii) moist (20°C and 90% RH); (iv) wet and dry cycles, alternating one day in water, at T = 20 °C, and one day of exposure in dry environment. At the end of the scheduled conditioning period (1, 3, 6 and 24 months), specimens were tested up to failure according to the same 4pb set-up employed for pre-cracking. "Superposition" between pre-cracking and post-conditioning stress-crack opening curves allowed the self-healing capacity to be evaluated.

For ordinary Fibre Reinforced Concrete (FRC), double edge notched 150 mm side square specimens were employed and pre-cracked, according to the "Double Edge Wedge Splitting (DEWS)" test methodology [9], up to a crack opening value equal to 0.25mm; specimens were then subjected to different exposure conditions: immersion in tap water, open air exposure and wet/dry cycles (4 days in water and 3 days exposed to open air). After one-month exposure to the different conditioning environments, specimens were tested again up to or further a crack opening of 0.25mm, and, after that, subjected again to the same exposure conditions for two further months and finally, they were re-tested again and pre-cracked up to further 0.25mm. The following cases were also considered:

- specimens continuously healed for three months after pre-cracking at 0.25 mm, re-cracked up to further 0.25 mm, further healed for one month and then re-cracked again up to further 0.25 mm;
- specimens continuously healed for six months after pre-cracking at 0.25 mm and then re-cracked up to further 0.25 mm.

Reference specimens were monotonically tested as well up to a crack opening width equal to 2.5mm. The healing efficiency was evaluated by comparing the crack-cycling curves with the monotone curve for a similar reference specimen.

Correlation with crack closure, evaluated through image analysis of optical microscopy garnered crack images, was also always performed.

Table 1. Mix designs of investigated cementitious composites (in kg/m^3).

Constituent	**NSC**		**FRC**		**HPFRCC (PoliMI/UFRJ)**		
Cement CEM II 42.5 A/L	300	300	360	360			
CEM I 52.5 R					600	600	600
Slag					500	500	500
Crystalline admixture		3		3		3	
Water	180	180	180	180	200	200	200
	w/c = 0.6		w/c = 0.50		w/(c+s) = 0.18		
Superplasticizer	3	3	3.5	3.5	33	33	33
fine aggr. 0-2 mm					982	982	982
fine aggr. 0-4 mm	1080	1077	814	811			
coarse aggr. 4-16 mm	880	880	1081	1077			
Steel fibers Dramix 5D 65/60BG			20	20			
Steel fibers Bekaert OL 13/0.16					100	100	50
Sisal fibers							7

3 Experimental results: analysis and discussion

3.1 Normal strength concrete

In Figure 1a the results of a typical test, in terms of load vs. COD curves are shown: the graphs are built up to compare the curves pertaining respectively to the pre-cracking test and to the post-conditioning up-to-failure test for the same specimens. From the values of nominal bending stresses an Index of Load Recovery (ILR) has been defined:

$$ILR = \frac{P_{max\ reloading,} - P_{unloading}}{P_{max,uncracked} - P_{unloading}} \tag{1}$$

An estimation of the crack closure due to the self-healing can be provided, by operating a “backward” shifting along the COD axis, of the stress-COD curve representative of the behaviour of each pre-cracked specimen after t conditioning, until the stress-COD curve of the same specimen, as measured during the pre-cracking test on the virgin undamaged sample is met. The new value of the “origin” COD can then estimated and an index of crack healing (ICH) was defined as:

$$ICH = \frac{COD_{pre-cracking} - COD_{post-conditioning}}{COD_{pre-cracking}} \tag{2}$$

Figures 2 a-c show the trend of the ISR and ICH vs. the exposure time for different exposure conditions and the related correlation:

- specimens immersed in water and made with concrete containing the crystalline additive exhibited an almost immediate and quite significant recovery, which even upon prolonged exposure, showed continuing improvement of the recovered performance; on the other hand, specimens made with plain concrete and immersed in water showed a more gradual recovery, which anyway, even after six months, barely attained half the level achieved by concrete with the crystalline additive;
- specimens exposed to open air and made with concrete containing the crystalline additive showed a gradual recovery capacity, as high as the one exhibited by plain concrete specimens in water; on the contrary a scant recovery capacity at all was exhibited by specimens without the additive.

The results highlighted in Figures 3 a-d and the SEM images with the related EDS analysis in Figures 4 a-b confirm the aforementioned statements and are coherent with the EDS spectrum of the admixture [3].

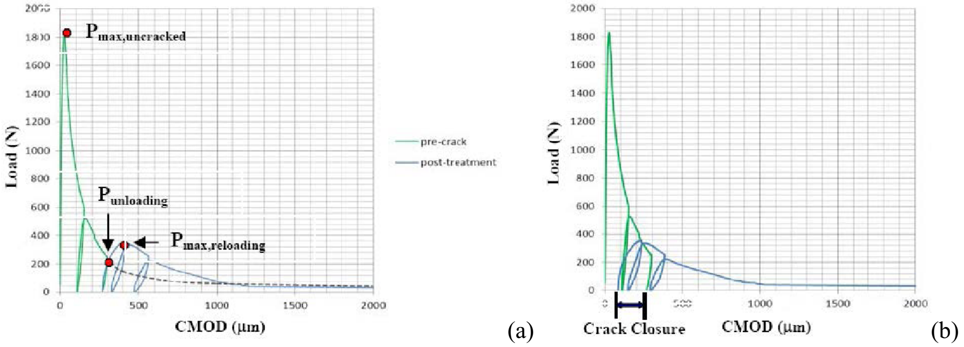

Fig. 1. example of load-COD curve obtained from 3pb tests on the same specimen before and after T-RH conditioning (a); proposal of a procedure to evaluate crack closure (b).

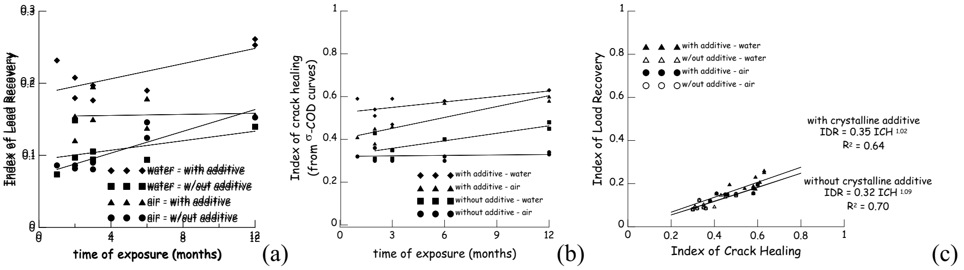

Fig. 2. Indices of Load Recovery (a) and of Crack Healing (b) and related correlation (c).

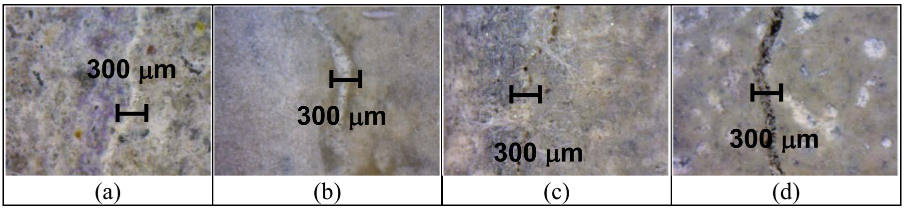

Fig. 3. Healed/healing cracks for specimens with (a,c) and without (b,d) crystalline additive after six months of immersion in water (a,b) and exposure to air (c,d).

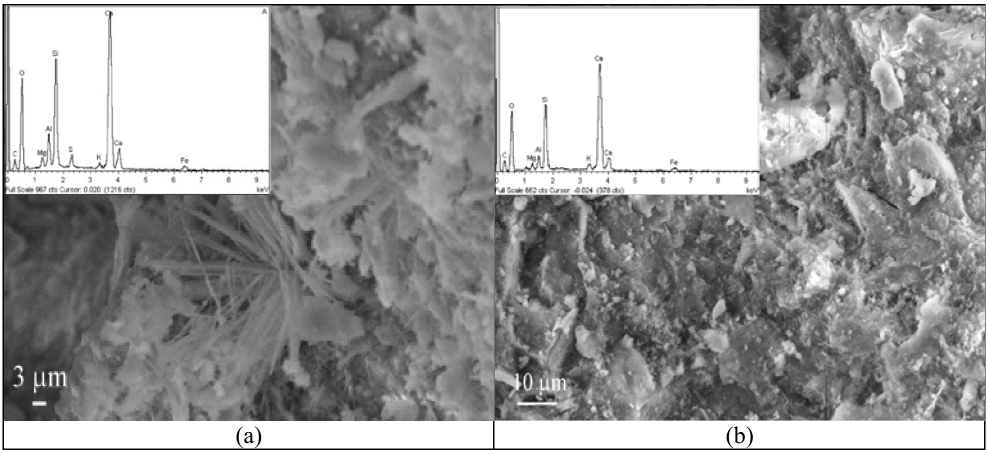

Fig. 4. SEM images and EDS analyses for specimens with (a,c) and without (b,c) crystalline additive after three months of immersion in water.

3.2 Fibre reinforced concrete and effect of cracking/healing cycles

As a first proof of the occurred healing, the maximum crack opening was measured before and after each healing period with a digital optical microscope at different locations along the crack for both concretes. An Index of Crack Healing was calculated as above (Eq. (2)).

The results are shown in Figures 5 a-b, first comparing the effect of a one cycle healing duration (1, 3 and 6 months) and then comparing the performance after the repetition of cracking-healing cycles (1+2 months vs. 3 months).

The following statements hold:

- prolonged exposure to healing conditions, whatever it is, results in an improved crack healing; the presence of crystalline admixture in the mix results in a faster crack sealing, mostly under less favourable exposure conditions (wet/dry cycles and even exposure to open air) (Figure 5a);
- the performance after the second cracking/healing cycle, is generally comparable with that after the corresponding first cracking-healing cycle, and somewhat better when a crystalline admixture is added to the mix; the longer the duration of the first healing period (3 vs. 1 month), the better the performance after the second cracking/healing cycle (Figure 5 b);
- for the same cumulative healing period, an intermediate cracking/healing cycles (1+2 months vs. 3 months continuing exposure) results in a slightly worse healing performance (Figure 5 b).

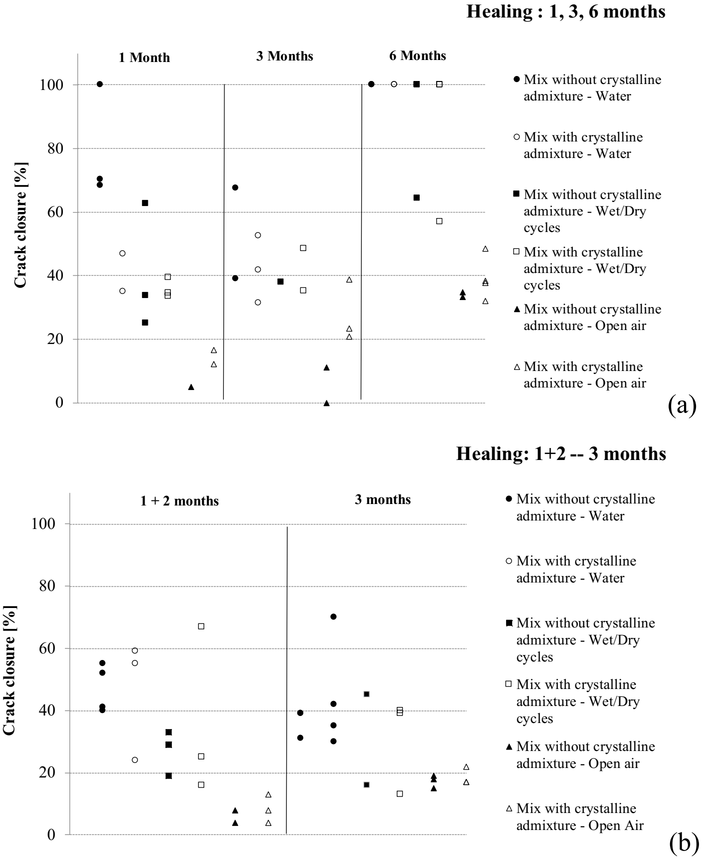

Fig.5. Index of Crack Healing (crack closure %) after different healing and cracking/healing exposures for ordinary Fibre Reinforced Concrete.

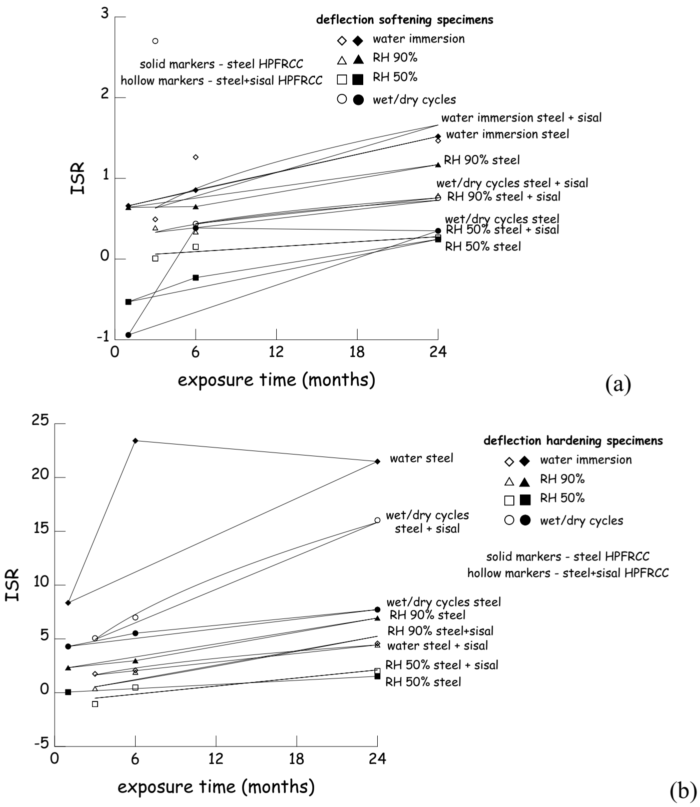

Fig.6. Index of Strength Recovery for deflection softening (a) and hardening (b) specimens made with steel only and hybrid steel+sisal HPFRCCs.

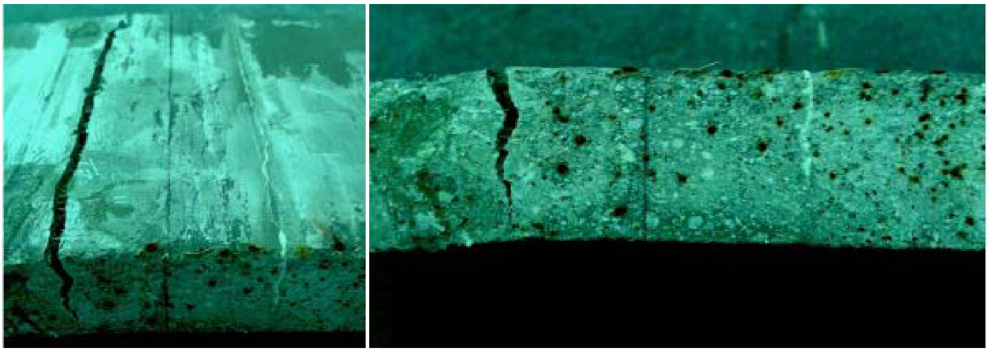

Fig. 7. Healed crack and new crack formed in post-healing tests in sisal + steel HPFRCC.

3.3 High performance fibre reinforced cementitious composites

Quantitative evaluation of the effects of self-healing has been performed once again calculating the Index of Stress Recovery through Equation (1) from the experimental results. Indices, plotted in Figure 6 for steel-only and steel+sisal HPFRCC specimens, highlight, on the one hand, the easily predicable effect of the exposure conditions and, on the other, the effects of conditioning duration.

The effectiveness of natural sisal fibres as facilitators and enhancers of the healing capacity also clearly appears, mainly with reference to the performance of specimens exposed to wet/dry cycles, which actually represents the most challenging condition in this respect. As a matter of fact, the natural fibres at a cracked site absorb the water in the wet stages of the cycles and transport it throughout the bulk material in the vicinity of the same crack. This contributes to an enhancement of the delayed hydration reactions, which are, together with $CaCO_3$ precipitation under wet and dry cycles, responsible of self-healing, thanks to the higher and longer availability of water, as retained by the fibres all along the drying stage of each cycle. This may also like to contribute to "shadow" the negative effect of calcium carbonate dissolution, precipitating during the same drying stages of each cycle, which is also responsible for self-healing and which may dissolve upon prolonged exposure, mainly in wider cracks, as observed.

For a deflection hardening specimen immersed in water for six months, in the post-conditioning test, the sealed crack did not reopen and a new crack was formed elsewhere (Figure 7). As a matter of fact, natural fibres also promote a better and more thorough healing at the fibre-matrix interface level, which acted synergistically with the reconstructed through-crack material continuity justifying the observed results.

Specimens immersed in water and containing also the crystalline admixture showed an even better performance (Figure 8). This may lead to hypothesize that, because of the expansion caused by the crystalline admixture reactions, some kind of "internal chemical pre-stressing" of fibres may have occurred [6]. SEM analyses of the fractured healed surfaces in specimens both without and with the crystalline admixture showed, in the latter case, a rather amorphous structure of the products covering the aggregate grains (Figure 9a) with typical composition of cement hydration products, thus confirming that healing is mainly due to delayed hydration of un-hydrated binder particles. In the former, the presence of crystals is observed (Figure 9b) with chemical composition compatible with the composition of the admixture. Deposition of healing products on the surface of the fibres is also evident (Figure 9c). The observed fibre rupture is attributable to an improved bond rightly resulting from the deposition of the aforementioned products, confirming the hypothesis about chemical pre-stressing resulting from the admixture action.

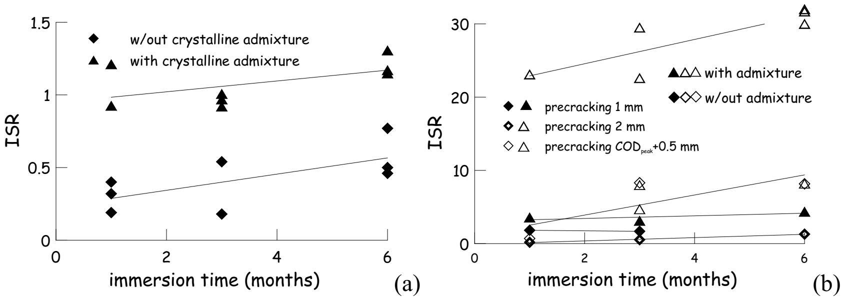

Fig. 8. Index Strength Recovery: deflection softening (a) and hardening (b) steel only HPFRCC specimens without and with the crystalline admixture immersed in water.

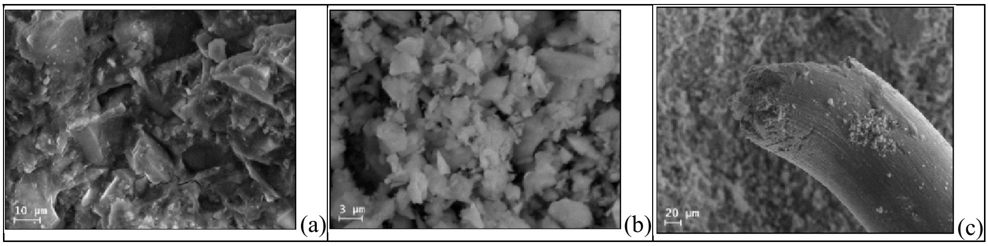

Fig. 9. Microscope image of healed fractured surfaces of specimens without (a) and with (b) the crystalline admixture; healed products on a ruptured fiber in HPFRCC with crystalline admixture (c).

4 Numerical modelling

Two modelling approaches have been proposed by the authors' group. A first one, based on micro-mechanics, incorporates the self-healing effects olf modeling the delayed cement hydration, as well as the effects of cracking on the diffusivity and the opposite repairing effect of the self-healing on the proposed micro-plane model constitutive laws [10].

The second one is a discontinuous-based porosity model and represents an extension of a fracture energy elastic-plastic interface formulation which now includes porosity evolution induced by self-healing mechanisms. The formulation accounts for the characterization of concrete failure behaviour in mode I and II fracture types. The post-cracking response is considered by means of specific work softening rules in terms of work spent and porosity evolution [11].

Both models have been calibrated on the NSC self-healing campaign detailed above and the comparison between the experimental and the numerical results (Figures 10a-b) highlights their reliability.

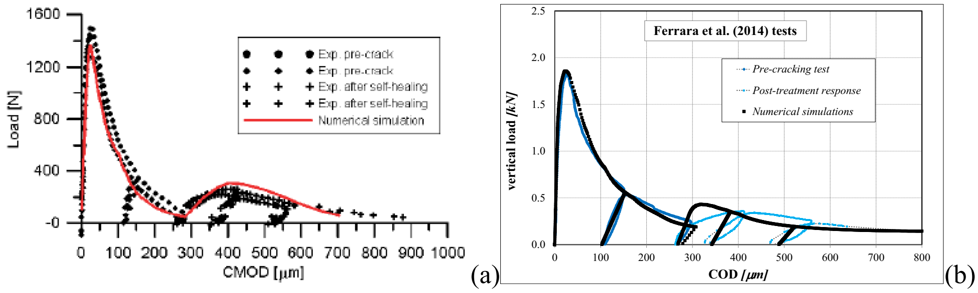

Fig. 10. Experimental vs. numerical results for self-healing capacity of NSC [10 and 11].

5 Conclusions

In this paper, the most significant results have been summarized of a comprehensive investigation on the self-healing capacity of different kinds of cement based materials, currently on-going for more than five years at the author's home institution, also in cooperation with other prominent institutions worldwide. The following statements hold:

- in presence of water NSC possesses a scattered autogenous self-healing capacity;
- tailored "self-healing activating" admixtures, such as crystalline ones, may enhance and make more reliable this capacity, even in case of other less favourable exposure conditions, and for cracks up to a few hundred microns (200-300 µm) wide;
- the same holds also for ordinary FRC, whose residual post-cracking strength can be significantly retained upon healing, also under repeated cracking/healing cycles, and mostly in the presence of crystalline admixtures;
- because of their peculiar composition HPFRCCs exhibit a significant autogenous healing capacity, always favoured by water and high relative humidity, which can lead to completely heal cracks up to several hundred microns (up to 0.5 mm) wide even occurring at quite later ages;
- the use of crystalline admixtures in fibre-reinforced concrete and cementitious composites can result in significant enhancement of the healing performance. This could be caused by some kind of internal "chemical pre-stressing" of fibres, triggered by the expansive reactions of the admixtures, which needs anyway to be further and more systematically investigated;

- the combined use of industrial and natural fibres enhance the aforementioned autogenous healing capacity of HPFRCCs thanks to the porous structure of fibres which can absorb water and promote its diffusion not only on the crack faces but through the bulk matrix of the composite;
- in all fibre reinforced cementitious composites the effects of healing on recovery or conservation of post-cracking mechanical performance upon exposure to different environments is attributable to both reconstruction of through-crack matrix continuity and healing of fibre/matrix bond, which also benefits from the aforementioned factors;

Further fundamental experimental investigations are needed to clarify the nature of self-healing products and their role in the sealing of the cracks and recovery of the material mechanical properties. Nonetheless, the results herein shown and the partially conclusive statements drawn from their analysis encourage to continue with the research on this topic to pave the way for the diffusion of self-healing cement-based materials into engineering practice with reference to both the construction of new buildings and the retrofitting of existing damaged or deteriorating structures. To this aim, it is fundamental in the authors' opinion to orient the investigation towards the characterization of healing capacity of cement based materials under sustained through-crack tensile stresses, as well as towards assessing the effectiveness of healing persistence and repeatability under alternating healing-loading/cracking cycles, as herein shown with reference to preliminary results.

A fundamental gap has to be filled which deals with the availability of reliable modelling tools able to reproduce the self-healing mechanisms and predict their effects on the performance of the materials. In this way, self-healing concepts could be consistently incorporated into durability design frameworks and life-cycle assessment design approaches. In this paper, the main results have been shown with reference to the calibration of two numerical models, which represent a first step in the aforesaid direction.

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