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Assessing the self healing capacity of cementitious composites

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ABSTRACT

The project detailed in this paper aims at a thorough characterization of the self-healing capacity of the cementitious composites, i.e. their capacity to completely or partially re-seal cracks and, in case, also exhibit recovery of mechanical properties. In this paper the problem will be investigated with reference a normal strength concrete. Different types of exposure, including accelerated temperature cycles, at constant relative humidity, in climate chamber, air exposure and water immersion. The effects of proprietary additives to engineer the self healing capacity will also be investigated. With reference to 3-point bending tests performed up to controlled crack opening and up to failure, respectively before and after exposure/conditioning recovery of stiffness and stress bearing capacity will be evaluated to assess the self healing capacity. Ultrasonic Pulse Velocity tests, optical microscopy observations and Scanning Electron Microscope analyses at selected "crack healed" locations will complement the investigation.

Keywords. Self healing, crystalline admixtures, strength and stiffness recovery, damage.

INTRODUCTION

Worldwide increasing consciousness for sustainable use of natural resources has made "overcoming the apparent contradictory requirements of low cost and high performance a challenging task" and a major concern in the Civil Engineering community. The availability of self-healing construction materials and technologies, by controlling and repairing "early-stage cracks in concrete structures were possible", 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 (Mihashi and Nishiwaki, 2012).

Since pioneer investigations, dating back to as early as one hundred years ago (Abrams, 1913, Loving, 1936, Turner, 1937) consensus has been achieved about the engineering significance of the problem: "if the mechanism of the [self healing] action is understood, and means can be found for accelerating it, a great stride will have been made in effectively retarding the rate of the disintegration of concrete, which […] is one of the major problems of the concrete field […]" (Lauer and Slate, 1956). The RILEM TC-221-SHC "Self healing phenomena in cement based materials" distinguishes:

- based on the result of the action, between *self-closing* and *self-healing*, whether only closure of the cracks or also restoring of the properties is observed;
- based on the process of the action, between "*autogenic*" (or *natural*) and "*autonomic*" (or *engineered*), whether the crack closure or restoration of material properties is due to either own concrete material or some engineered addition.

Mechanisms of self-healing can be categorized as follows (Ramm and Biscoping, 1998):
(1) further reaction of un-hydrated cement;
(2) expansion of the concrete in the crack flanks;
(3) crystallization of calcium carbonate;
(4) closing of the cracks by solid matters in the water flowing through the crack, in case;
(5) closing of the cracks by spalling-off of loose concrete particles upon cracking.

Several variables, besides the presence of water and, in case, of carbon dioxide, may affect the aforementioned mechanisms and phenomena, such as:

- the mix constituents (Dhir et al., 1973);
- the stress state along the cracks (Ngab et al., 1971);
- the temperature of the water (Reinhardt and Joos, 2003);
- the alternation between water saturated conditions and exposure to air, which reduced "the strength developed by a marked degree" (Lauer and Slate, 1956).

In the very last decade a huge amount of research work has been dedicated to "engineered" self healing, along three main fields of research: self healing engineered with fibre reinforcement, mineral-producing bacteria and proprietary chemical admixtures. The self healing action in the latter case is mainly due to expansion effects and to re-crystallization. The supply of water or at least moisture is essential, but "since most infrastructures are exposed to rain or underground water, usually this is an easily satisfiable requirement". The use of the so-called "crystalline additives" is well known with reference to the reduction of concrete porosity and of water permeability of concrete, and hence to the improvement of the tightness and waterproof properties of structural elements, when required. These additives contain substances which react with cement constituents and form calcium silicate hydrates. The reaction propagates through the concrete mass because of osmosis, Brownian motion and progressive involvement of anhydrous cement particles. The reaction products tend to fill the capillary voids, thus resulting in a system impervious to water and other environment born aggressive substances. The reaction consumes the moisture inside the concrete but can also undergo a delayed activation, whenever the material comes back into contact with water and/or environment moisture, as when cracking happens even at later ages.

The quantitative assessment of crack sealing/healing on the recovery of engineering properties of concrete still needs a much deeper investigation. Most of the surveyed studies (Hearn and Morley, 1997; Hearn, 1998; Edvardsen, 1999) focused on the variation of water permeability and only very few among them (Lauer and Slate, 1956; Dhir et al., 1973) analyzed the effects on recovery of mechanical properties, in case evaluated by means of non-destructive techniques; it was anyway pointed out that recovery in signal transmission was not as spectacular as that in permeability (Aldea et al., 2000).

In this paper an experimental methodology will be proposed to evaluate the self healing capacity of concrete and cementitious composites, both autogenic and engineered with the addition of crystalline admixture. Validation of the methodology, which includes both non-destructive (Ultrasonic Pulse Velocity) and destructive (3-point bending) tests, will be performed by means of a thorough test programme, including, as testing variables, the influence of crack opening and exposure, natural or artificially controlled, conditions.

EXPERIMENTAL PROCEDURE

As remarked above in this paper the self healing capacity will be investigated of concrete with and without crystalline admixtures with reference to mix-design compositions shown in Table 1. Slabs 1m x 0.5 m and 50 mm thick were casted, from which prismatic beam specimens 500 mm long and 100 mm wide were cut. Specimens were cured in a chamber at 20°C and 95% Relative Humidity (RH).

The proposed methodology is based on pre-cracking prismatic beam specimens by means of a COD-controlled 3-point bending set-up (450 mm span - Figure 1), up to different levels of crack opening (100 and 200 μm respectively). Specimens were pre-cracked after 35 to 42 days curing and then,, together with un-cracked reference specimens, were submitted to different exposure conditions, *i.e.* immersion in water at 20°C, open air (un-cracked and pre-cracked at 200 μm, in both cases) and T-RH cycles simulating typical summer climate in Northern Italy (un-cracked and pre-cracked at both 100 and 200 μm). Charts of temperature and RH along the exposure period, as well as of the controlled T-RH cycles are shown in Figure 2a-b. At the end of scheduled exposure times, three point bending tests were performed and results between pre-cracked and virgin specimens were compared. Load and stiffness recovery capacity was evaluated, and related “self-healing” indices to be defined.

Moreover, Ultrasonic Pulse Velocity (UPV) tests were performed according to the set-up shown in Figure 3 (distance between emitter and receiver equal to 90 mm); tests were performed before any cracking and environmental conditioning, after the pre-cracking but before the environmental conditioning, and, finally after the scheduled exposure times.

Figure 1. Three-point bending test set-up

Table 1. Mix composition of investigated concretes

Constituent	Without additive	With additive
Cement type II 42.5	300	300
Water	190	190
Superplasticizer (lt/m^3)	3	3
Fine aggregate 0-4 mm	1078	1080
Coarse aggregate 4-16 mm	880	880
Crystalline additive	=	3

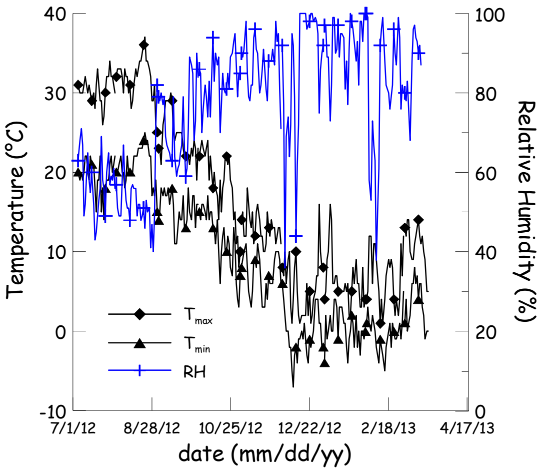

Figure 2a. T and RH recorded along the specimens exposure period.

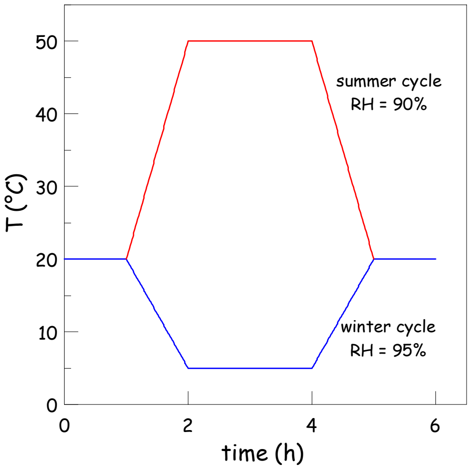

Figure 2b. T-RH cycles for climate chamber specimen conditioning.

Figure 3. UPV test set-up

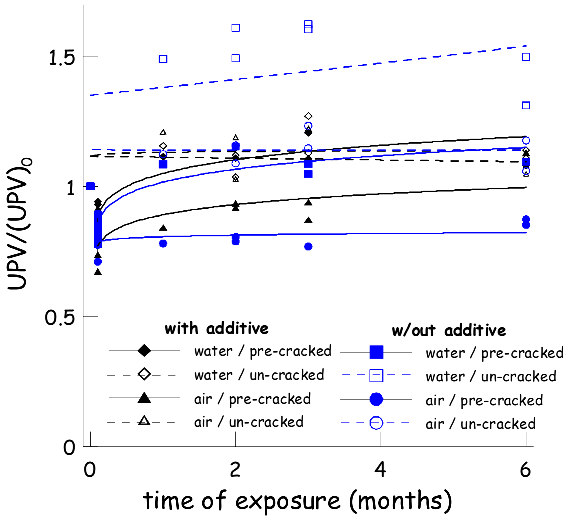

Figure 4a. Relative UPV for water and air conditioned specimens

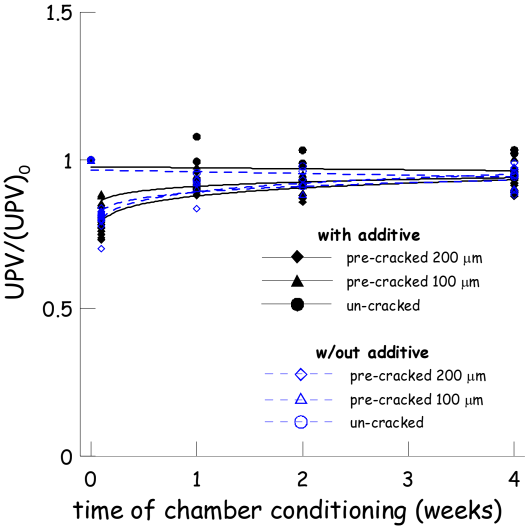

Figure 4b. Relative UPV for chamber conditioned specimens/summer cycles

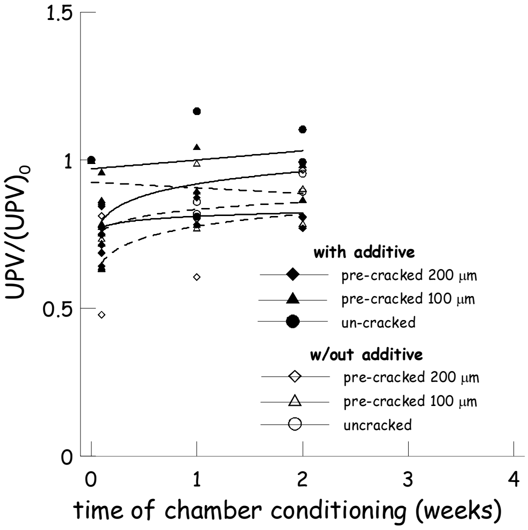

Figure 4c. Relative UPV for chamber conditioned specimens/winter cycles

EXPERIMENTAL RESULTS

Ultrasonic Pulse Velocity tests

From measured transit time, between the emitter and receiver units of the UPV test apparatus, wave speed was calculated with reference to distance between the units (90 mm, as said above). The velocity measured for each specimen before both pre-cracking and environmental conditioning was assumed as a reference, and denoted as $(UPV)_0$. Values of velocities measured after cracking and after conditioning, dimensionless to the aforementioned reference value, have been plotted as a function of the exposure time in Figures 4a-c, respectively for air exposure and water immersion as well as for accelerated conditioning in climate chamber representative of either summer or winter climate. Results for concrete both without and with the crystalline admixture have been reported.

The effectiveness of the crack repairing phenomena, either autogenic or cathalyzed by the presence of the crystalline additive, clearly appears from the trend of the plotted relative ultrasonic pulse velocity for pre-cracked specimens. The higher effectiveness of a continuous water immersion is also evident: specimens in water, after only one month, are able not only to recover their pristine features but even show an improvement with respect to the values of ultrasonic pulse velocity measured before any cracking and conditioning process. Concrete containing the crystalline additive behaves slightly better than concrete without it. On the other hand, specimens exposed to air, recovery more slowly the pristine level of performance. It has furthermore to be noted that only for specimens made with concrete containing the crystalline additive, after six months exposure to natural weather conditions (as in Figure 2a) a value of the ultrasonic pulse velocity as for virgin specimens was gained back. Specimens made with concrete not containing any additive showed, with reference to UPV tests, an almost constant performance all along the exposure time.

Un-cracked specimens, on their hand, either immersed in water or exposed to air, featured almost constant performance. Specimens not containing the crystalline additive and immersed in air stand as the only visible exception to the aforementioned statement, for which value of UPV 50% higher than the reference one was in fact measured as soon as after one month immersion; after then the values held almost constant. This may be explained considering very low absolute values of the UPV measured for virgin specimens made with concrete not containing the crystalline additive, as low as 3.5 km/s, compared to average velocity values of 4 km/s measured for specimens made with the concrete with it. This discrepancy can be explained by the significant plastic shrinkage that specimens without the additive experienced, even if cured under moist towels before demoulding, as witnessed by diffused observed surface cracking. The crystalline admixture, with its action, could have been effective also in counteracting this phenomenon.

With reference to climate chamber conditioning recovery of pristine level of performance, as measured for virgin specimens, was observed after four weeks for summer type cycles, whereas it appears that, at least for specimens made with concrete containing the crystalline additive, two weeks were enough for the smaller crack opening (100 μm). It is furthermore evident that, whereas in conditioning meant as representative of autumn/winter exposure, the performance of concrete containing the crystalline additive was significantly better than for concrete without it, differences were less evident, if almost scant, in the case of conditioning representative of spring/summer exposure. This could be most likely attributed to the effect of the higher temperatures attained, together with the repetition of the warming/cooling cycles. Anyway, for both winter and summer temperature cycles, similarly to what happened for “natural” exposure conditions discussed in the previous paragraph, uncracked specimens did not exhibit any significant variation of the measured properties. This evidence strongly supports the assumption that the measured recovery, in terms of ultrasonic pulse velocity has to be attributed not to a bulky continuing hydration but rightly to a self healing, either autogenic or engineered by the addition of the crystalline additive, of the cracks, due to the reaction with water or atmospheric humidity of potentially reactive material exposed upon cracking, being it un-hydrated cement or ready to react hydrophilic admixture particles.

Three point bending tests

The same set-up employed for pre-cracking specimens up to selected crack-opening values (Figure 1), was employed to test up to failure the same specimens after scheduled times of exposure to the different environment conditions, namely water immersion, open air and climate chamber temperature cycles meant as representative of winter and summer. In Figure 5 results of typical tests, in terms of nominal stress σ_N vs. COD curves, are shown: it is worth remarking that figures are built up in such a way that the curves pertaining respectively to the pre-cracking test and to the post-conditioning up-to-failure test *for the same specimens* are compared. Through the analysis of the data garnered through the wide test program, the following remarks can be highlighted:

- a significant recovery of the stiffness has been generally observed when testing specimens after environmental conditioning, with respect to the unloading stiffness measured upon pre-cracking;
- a recovery of the load bearing capacity has been also observed, but only for selected exposure conditions (e.g. winter climate chamber) and/or prolonged exposure times (e.g. specimens immersed in water after three to six months or summer climate chamber after four weeks); recovery of load bearing capacity is evaluated by comparing the maximum load in the post-conditioning 3pb test with reference to the load attained in correspondence of the unloading during the pre-cracking 3pb test.

The analysis of data allowed “indices of healing” to be defined and evaluated as follows:

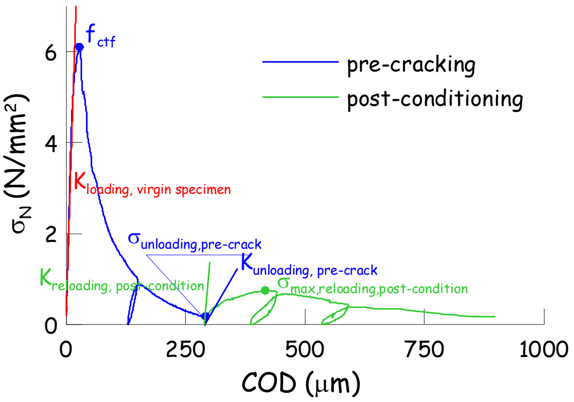

Figure 5. Example of stress vs. COD curves for specimens submitted to pre-cracking and post-conditioning 3pb tests; definition of quantities for calculation

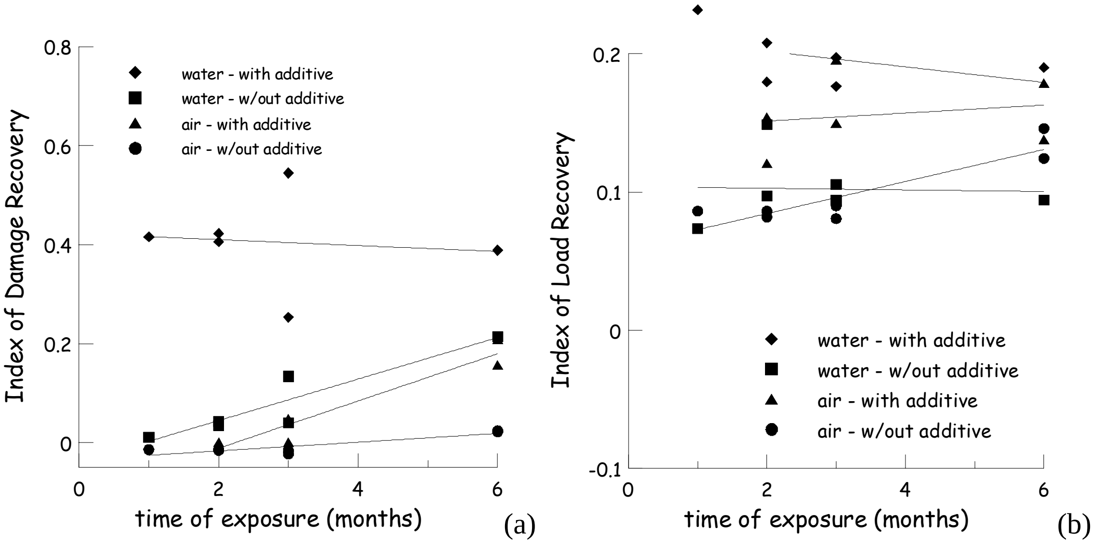

Figure 6. Indices of Damage (a) and Load (b) Recovery for water/air exposure

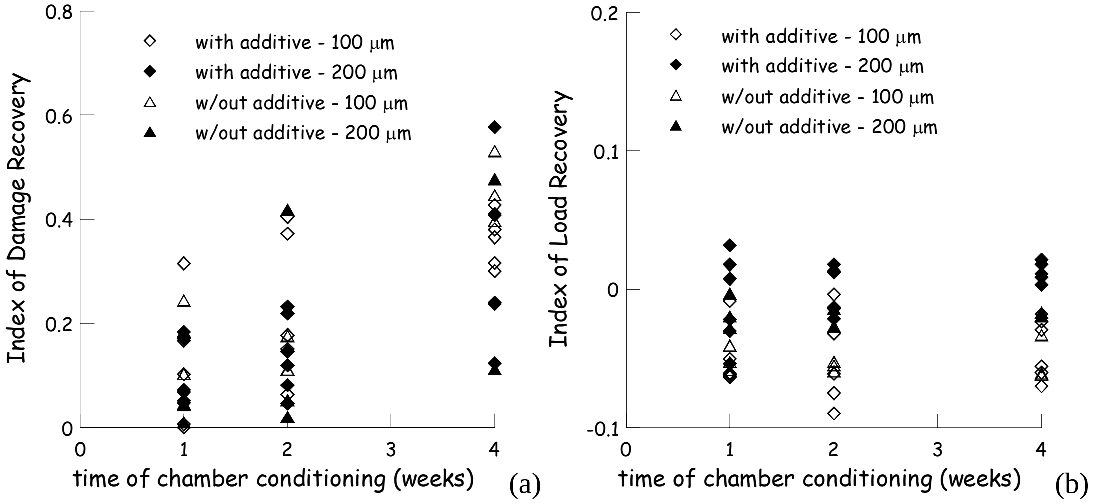

Figure 7. Indices of Damage (a) and Load (b) Recovery - summer cycles

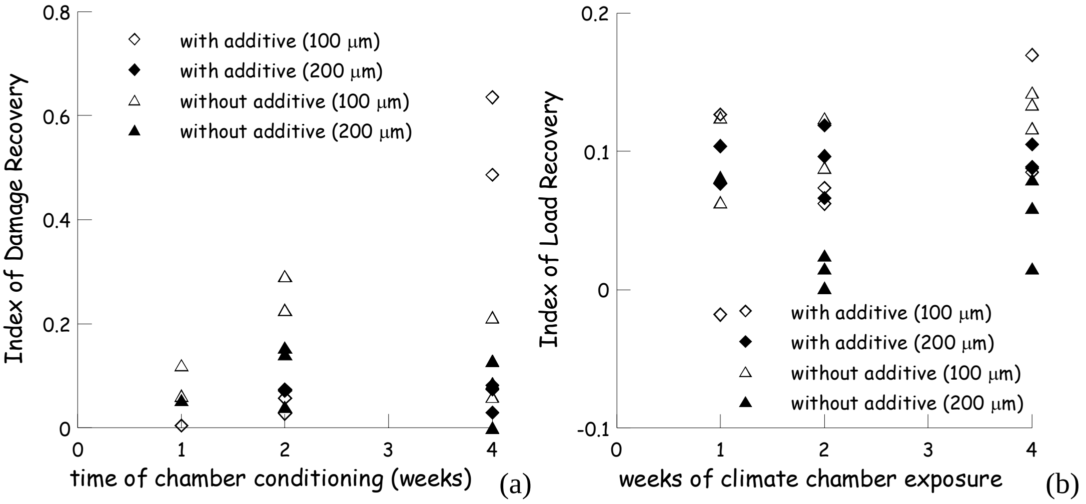

Figure 8. Index of Damage (a) and Load (b) Recovery - winter cycles.

- *index of damage recovery*

$$\mathrm{IDR} = \frac{K_{\mathrm{reloading,\,post\text{-}conditioning}} - K_{\mathrm{unloading,pre-crack}}}{K_{\mathrm{loading,virginspecimen}}} \qquad (1)$$

the significance of stiffness K notations being self evident (see also Figure 5).

Figure 6a, 7a and 8a show the trend of the Index of Damage Recovery, computed as above, vs. the exposure time for different exposure conditions. The following remarks hold:

- specimens immersed in water and made with concrete containing the crystalline additive exhibited an almost immediate and quite significant recovery, which held constant afterwards; on the other hand specimens made with plain concrete and immersed in water showed a 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 immersed in water; on the contrary no recovery capacity at all;

- climate chamber conditioning, representative of both winter and summer climates, induced gradual recovery of stiffness; higher temperatures (summer climate) are likely to favour, in general, a more systematic recovery of the stiffness. The catalyst role of the crystalline additive in promoting self healing reactions is confirmed.

- *index of load recovery*

$$\mathrm{ILR} = \frac{\sigma_{\mathrm{N,max\,reloading,\,post\text{-}conditioning}} - \sigma_{\mathrm{unloading,pre-crack}}}{f_{\mathrm{ctf}}} \qquad (2)$$

the significance of stresses and strength notations being once again self evident (Figure 5).

Figures 6b, 7b and 8b show the trend of the Index of Damage Recovery, computed as above, vs. the exposure time. With reference to natural exposure conditions, i.e. immersion in water and exposure to open air, the same trends as previously detected and analyzed with reference to the index of damage recovery appear to be confirmed. On the other hand, with reference to climate chamber conditioning, a scant recovery of the load bearing capacity, as computed with reference to the flexural tensile strength of the virgin undamaged material, is achieved when specimens are exposed to warmer temperatures. However, specimens with larger crack opening and presence of crystalline additive in the mix-design perform better than others, most likely because of the larger cracked surfaces which is likely to expose to moisture larger cluster of self healing catalysts. Load recovery capacity under cooler temperatures appears to be more significant, the effect of the additive being also confirmed. The detected differences between specimens exposed to summer or winter temperature cycles could be attributed to a sort drying effect of the higher temperatures in the former case, which may have counteracted and jeopardized the build up of self healing products and their stronger interpenetration within the structure of the bulk original material.

Comparison among the different indices, evaluated from either bending or UPV tests, provided consistent results, not shown herein for the sake of space limitation, which anyway supports the reliability

Figures 9 and 10 show healing and healed cracks for different exposure conditions.

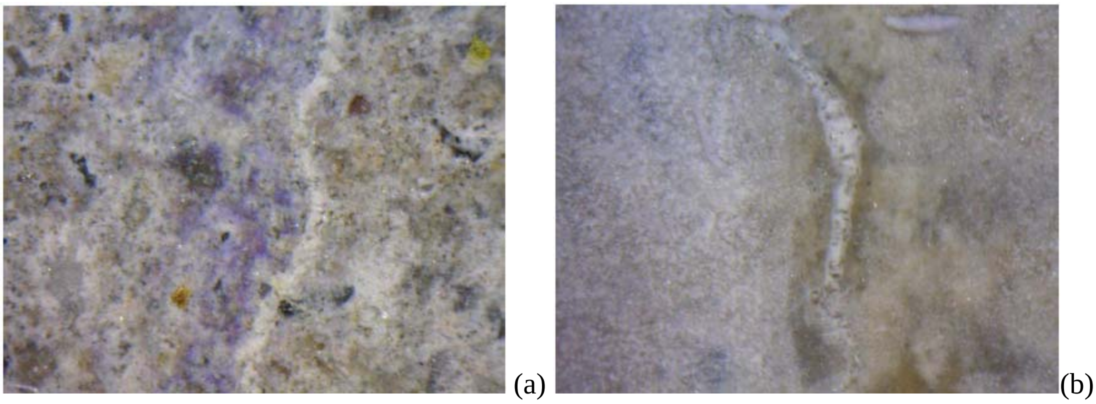

Figure 9. Healed cracks (200 μm) for specimens with (a) and without (b) crystalline additive after six months immersion in water.

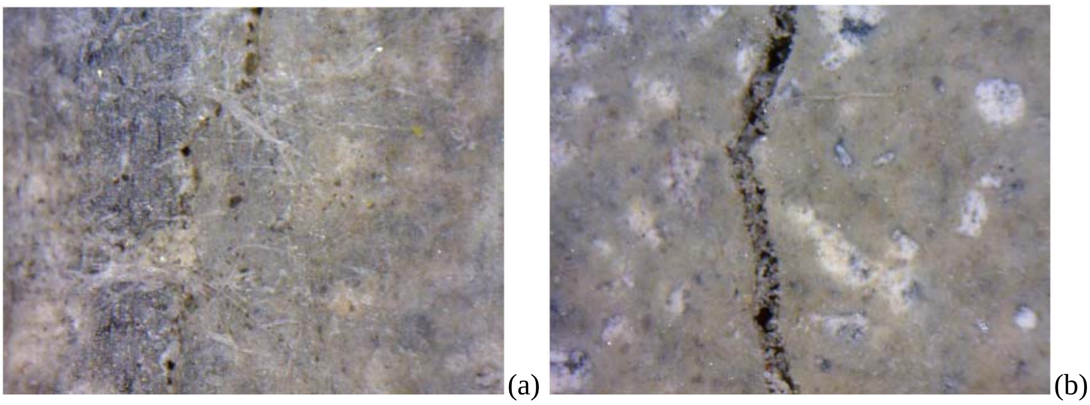

Figure 10. Healing cracks (200 μm) for specimens with (a) and without (b) crystalline additive after six months exposure to open air.

CONCLUDING REMARKS

A methodology has been proposed to quantify the effects of self-healing on the mechanical properties of cement based materials. It is based on pre-cracking beam specimens to prescribed crack-widths, and, after scheduled times to selected exposure conditions, testing them again until failure. Indices of recovery of mechanical properties, such as bending stiffness and post-cracking load bearing capacity have been evaluated.

Cement based materials inherently possess, within an acceptable range, some self-healing capacity, most likely due to continuing hydration favoured by suitable environment conditions, which is anyway randomly scattered. Crystalline admixtures not only enhances the aforementioned self-healing capacity but also makes it more reliable and consistent.

Validation of the proposed methodology with reference to a wider range of exposure conditions, including effects of constant high and low temperatures, as well as to a wider range of concrete compositions, together with characterization of self-healed cracked interfaces through microscopy observation, will be instrumental to gain a stronger confidence in the self-healing phenomenon and its effects on mechanical properties of cementitious composites. This is of the utmost importance in order to consistently include self-healing into durability-based design approaches.

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