

Self-Healing Phenomenon Evaluation in Cementitious Matrix with Different Water/Cement Ratios and Crack Opening Age

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Abstract—Concrete elements are subject to cracking, which can be an access point for deleterious agents that can trigger pathological manifestations reducing the service life of these structures. Finding ways to minimize or eliminate the effects of these aggressive agents' penetration, such as the sealing of these cracks, is a manner of contributing to the durability of these structures. The cementitious self-healing phenomenon can be classified in two different processes. The autogenous self-healing that can be defined as a natural process in which the sealing of these cracks occurs without the stimulation of external agents, meaning, without different materials being added to the mixture, while on the other hand, the autonomous seal-healing phenomenon depends on the insertion of a specific engineered material added to the cement matrix in order to promote its recovery. This work aims to evaluate the autogenous self-healing of concretes produced with different water/cement ratios and exposed to wet/dry cycles, considering two ages of crack openings, 3 days and 28 days. The self-healing phenomenon was evaluated using two techniques: crack healing measurement using ultrasonic waves and image analysis performed with an optical microscope. It is possible to observe that by both methods, it is possible to observe the self-healing phenomenon of the cracks. For young ages of crack openings and lower water/cement ratios, the self-healing capacity is higher when compared to advanced ages of crack openings and higher water/cement ratios. Regardless of the crack opening age, these concretes were found to stabilize the self-healing processes after 80 days or 90 days.

Keywords—Self-healing, autogenous, water/cement ratio, curing cycles, test methods.

I. INTRODUCTION

THE self-healing phenomenon can occur in an autogenous or autonomous way, being the last one through the addition of a specific material in the cement matrix with this purpose. Autogenous self-healing can be defined as a natural process in which the sealing or healing of the cracks occurs without any stimulation of external factors and/or efforts [1]. The hydration of anhydrous cement in the cured matrix may be an example of autogenous self-healing. Thus, the composition of the cement, such as the presence of fly ash and slag can contribute to the self-healing process due to the improvement in the cement matrix considering that there is a greater amount of hydrates created [2]-[5], as well as the presence of ions in the environmental in which the samples are placed.

Characteristics associated to the environment can

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significantly influence the self-healing process [6]-[12]. Yang et al. [6] verified total crack sealing under-50 micrometers, after 4 to 5 cycles of immersion in water for 24 hours and drying for 24 hours. The analysis proposed by [7], in wet and dry cycles at 12 hours in each situation, cracks under 15 micrometers were completely sealed after seven cycles, and for submersed curing specimens, this same phenomenon in the same period, was not occurred. Some studies report the phenomenon of self-healing at young ages, with satisfactory results after three consecutive cycles [6] or four to five cycles [13]. It is important to note that these are specific conditions of cracks under 0.15 mm, cement composites mortar, and the use of the cycles without renewal water the environment exposure.

The temperature of the environment is another relevant feature in the formation of the healing compounds [13] and also in the morphology of the formed crystals [14]-[16]. Reinhardt and Jooss [17] compared the permeability of mortars with cracks width of 5 μm , 10 μm and 15 μm , exposed to three different temperatures of 20°C, 50°C and 80°C, concluding that an exponential growth of permeability can be obtained for all temperature ranges in according to the crack width.

As for the phenomenon measurements, some techniques can be used to monitor or determine the self-healing rate in the cementitious matrix, such as optical microscopy, X-ray diffraction, Raman spectroscopy [18], ultrasonic pulse velocity propagation [19], and others. The growth of the healing products tends to densify a cracked area, for example, causing a greater densification of the matrix, and consequently, it increases the ultrasonic pulse velocity.

The purpose of this study is to evaluate the autogenous self-healing phenomenon in concretes produced with different water/cement ratios, exposed to wet/dry cycles, with two ages of cracks openings, 3 days and 28 days.

II. MATERIALS AND METHODS

A. Materials

The cement used in this study was a high early strength Portland cement (CPV), which may contain 0% to 10% of carbonaceous material according to NBR 16697 [20], this cement is similar to the Type III according to ASTM C 595 [21]. This cement was chosen because it is considered a pure cement, since it may contain only limestone filler but any other supplementary cementitious materials such as pozzolans

and slag. In its chemical composition, it was obtained in mass percentage, 12.80% silicon dioxide (SiO₂), 3.73% aluminum oxide (Al₂O₃); 4.89% iron oxide (Fe₂O₃), 68.85% calcium oxide (CaO), 1.06% magnesium oxide (MgO), 4.69% sulfur trioxide (SO₃), 1.47% potassium (K₂O), 1.36% carbonic anhydride (CO₂) and 3.09% loss on ignition. The average particle diameter is 11.35 micrometers, obtained through laser particle size analysis. The specific weight is 3.12 g/cm³ according to NBR 16605 [22], while the specific area is 6.232 m²/g performed using the B.E.T. method, with a compressive strength [23] of 29.3 MPa at 3 days, 35.2 MPa at 7 days and 42.1 MPa at 28 days.

The fine aggregate used is a natural sand of quartz origin, with fineness modulus of 1.89, calculated according to NBR 7211 [24], with a maximum particle size of 2.36 mm. The specific weight of this dry aggregate is 2.54 g/cm³, bulk weight of 1.49 g/cm³ and water absorption of 1.30%. The coarse aggregate is a basalt, with fineness modulus of 6.10, maximum particle size of 12.5 mm; the specific weight of this dry aggregate is 3.01 g/cm³, bulk weight of 1.51 g/cm³ and water absorption of 0.20%.

The use of a superplasticizing admixture was necessary to obtain a established workability of 220 ± 20mm, according to NBR NM 67 [25], common to all water/cement ratios used. This admixture is a polycarboxylate base, with specific weight between 1.080 g/cm³ and 1.120 g/cm³ and 40.28% solids; its recommended dosage is between 0.3 and 2.0% on the cement content.

B. Experimental Program

The experimental program developed for this research aims to verifying the influence of different crack opening ages and different water/cement ratios on concrete autogenous self-healing, using one single type of cement (Fig. 1).

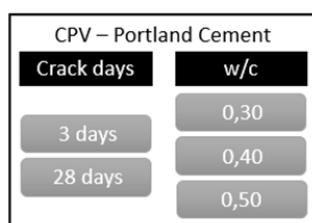


Fig. 1 Proposed experimental program

The mixes used in this study are presented in Table I, in every mix, the slump and the mortar proposition where fixed.

TABLE I
MIXES DEVELOPED AND CEMENT CONTENT

Water/cement ratio	m	Unit mix design	SP (%)	Slump (mm)	Cement content (kg/m ³)
0.30	2.56	1:0.85:1.71	0.23	222	640.74
0.40	3.55	1:1.37:2.18	0.15	215	481.25
0.50	4.54	1:1.88:2.66	0.14	235	390.01

The mixing of the materials was done using a 150-liter mixer with vertical axis in which the material mixing order

and the operator remained the same through the entire study. The samples used where prismatic with dimension of 100x100x50 mm. To cast the samples, a vibrating table was used; the procedure was done in three equal layers and vibrated for 15 seconds each. The process was repeated for the cylindrical specimens (100x200 mm) casted for mechanical characterization of the concretes in the study.

After casting the samples, the specimens were conditioned at the laboratory room conditions (23 ± 2°C) for the first 24 hours, covered with a plastic membrane. Subsequently, they were removed from the mold and stored in a curing room at a temperature of 23 ± 2°C and humidity above 95%, where they remained until the test dates, according to NBR 5738 [26].

By the age of crack opening, at 3 days and 28 days, the specimens were subjected to a compression load to open the cracks by traction, and a restriction of the movement on the sides of the specimen was used. This way, the cracks width was limited to 0.4 mm or less. Although for autogenous healing, cracks under 0.15 mm are recommended, even preferably under 0.05 mm, in order to highlight the phenomenon of self-healing [7], it was decided to consider cracks with larger dimensions, limited to 0.4 mm, taking as a parameter this value considered to be the maximum recommended in the NBR 6118 [27] and ACI 318 [28]. The complexity of a cementitious matrix containing a coarse aggregate causes the crack not to dissipate in a regular and homogeneous line, therefore, in the specimens studied, the cracks width varies along its length.

After the cracks were opened, the specimens were conditioned in wet/dry cycles, with a two-day immersion period, followed by twelve days in a dry condition, the dry condition were set in a curing room at a temperature of 30 ± 2°C and a relative humidity of 75 ± 5%. The water used in the cycles was from the city public supply, and was renewed for each cycle; the purpose of this approach was to more closely replicate the usual conditions of natural daily cycles. The specimens were stored with the cracks in the vertical position in the cycles, with the larger width of the crack directed downwards, since the lixiviation products tend to follow the gravitational flow and, therefore, did not favor the closure of the smaller width of the crack.

C. Methods

For the mechanical characterization of these concretes, a compressive strength test was performed at 3 days and 28 days, same ages used for the crack openings, the test was performed as recommended in the NBR 5739 [29]. The self-healing phenomenon was evaluated using two techniques, chosen to check the water permeability recovery, through the determination of ultrasonic pulse velocity and a qualitative analysis by monitoring the cracks through optical microscopy techniques, based on study developed by [30]. This research describes the main techniques of monitoring the phenomenon of self-healing in to cement matrix.

The water permeability recovery was evaluated by the test that determines the velocity of the ultrasonic pulse propagation in a solid body. The method consists in

determining the time required for the propagation of an ultrasonic frequency pulse emitted by one transducer and received by another positioned on opposite face of the element to be measured. The characteristic ultrasonic pulse velocity is the ratio of the distance between the transducers to the propagation time obtained, with an accuracy of ± 0.1 microsecond [31]. This test is an important technique to detect the occurrence of cracks and the degree of damage caused [32], helping on the self-healing verification [33], [34]. The existence of voids present in the damaged cementitious matrix can decrease the velocity of the ultrasonic pulse, due to the increase of the time necessary in the propagation path of the pulse inside the specimen. This is evidenced in the method of ultrasonic pulse transmission [32].

Nine specimens (100x100x50 mm) were casted to perform this test. The method was used to measure the ultrasonic pulse in the specimens before opening the cracks and after, followed by 91 days in wet/dry cycle. The direct reading method was chosen, with three measurements per specimens, aligned and perpendicular to the direction of the crack. A test apparatus was developed to lock the specimens and ensure that the readings were always performed at the same points. The specimens were inserted in this apparatus and received a mark before the first reading, so that the direction was kept the same for the other readings during the study (Fig. 2).

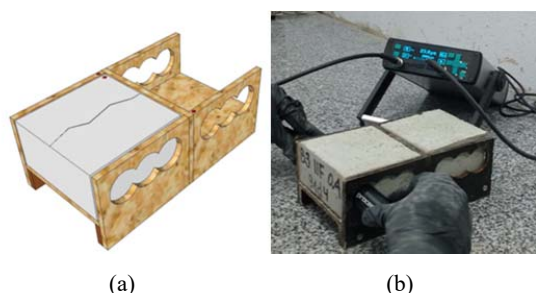


Fig. 2 Device used to ensure the correct alignment of the readings during the test (a), and (b) test equipment

The velocity of the ultrasonic pulse (V_{t0}), prior to the opening of the crack, was defined as the initial reference. The opening of a crack in the matrix causes the decrease of the ultrasonic pulse velocity (V_{ti}) due to the increase in the distance required for its propagation, with “i” being the date for each measurement. Thus, through (1) the velocity recovery rate can be obtained. This way, P_{t0} represents the damage caused by the opening of the crack. The calculation that expresses the ultrasonic pulse velocity rate compared to the initial measurement is presented in (1).

$$P_{t0}(\%) = \frac{V_{ti}}{V_{t0}} \quad (1)$$

P_{t0} = relative ultrasonic pulse (%); V_{ti} = ultrasonic pulse velocity after crack opening - at every reading time; V_{t0} = ultrasonic pulse velocity before crack opening.

An optical microscopy analysis, using a Zeiss Stemi 508 optical microscope, with a magnification of 2x and 250x, from

the “Laboratório de Materiais e Tecnologia do Ambiente Construído” (LAMTAC / NORIE / UFRGS) was performed in two of the nine specimens used in the test of ultrasonic pulse velocity. Through this procedure, a more detailed verification can be done, since often they can be determinants for the understanding of the results [35]. This test aims to qualitatively analyze, over time, the healing of the crack. In addition to the self-healing analysis, the optical microscope was also used to measure the width of the cracks. Some research demonstrates this method to be suitable for qualitative analysis, and quantitative measure of this phenomena [7].

The images were obtained with a magnification of 2x at a resolution of 0.65x, presenting a scale of 0.1 mm in the images. A grid, divided into five parts, was inserted to measure the width of the cracks in order to obtain four readings points with similar dimensions. When there were pores or unusual imperfections, such as air voids in the area where the measurement was taken, the displacement of the reading grid was necessary. In these cases, the reading was performed to the left of that point, at the closest area available for the analysis (Fig. 3). This methodology allowed to obtain the average widths of the cracks for each combination, in addition to the maximum and minimum widths that will allow the analysis of the results obtained to explain the occurrence of the self-healing phenomenon.

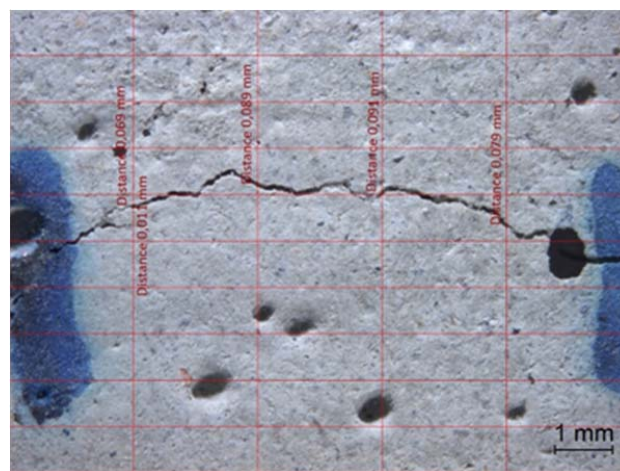


Fig. 3 Example of crack width measurements performed with optical microscopy

The proposed visual analysis procedure, aims to create a correlation between the widths of the cracks and the speed of the ultrasonic pulse velocity, considering that, for small width cracks, there is a greater probability of self-healing. Although a limitation of this technique is that the observation is only possible in the surface of the crack, the association with the ultrasonic pulse test allows the interior of the cementitious matrix to be contemplated in the analyzes. The composition of every image of each crack, was made through a mosaic of eight images, spaced each other every 8 mm, as shown in Figs. 4, 10 and 11.

A qualitative analysis was performed by visual verification

of possible healing points at the age of 14 days and 91 days, the time defined as the end of the analysis for this study.

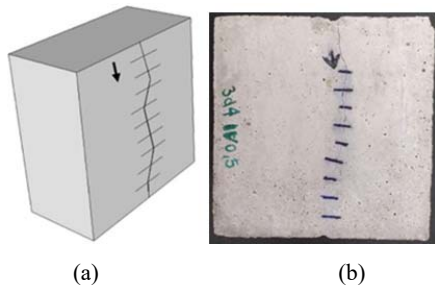


Fig. 4 Methodology for self-healing monitoring by optical microscopy (a) schematic representation; and (b) photographic record of a sample used in the optical microscopy

III. RESULTS AND DISCUSSIONS

The mechanical characterization of the concrete was performed at same ages of cracks openings, 3 days and 28 days. The results of the compressive strength are shown in Table II and Fig. 5.

Crack opening age (days)	a/c ratio	Compressive Strength (MPa)	SD (MPa)	VC (%)
3	0.30	46.40	0.74	1.60
	0.40	32.53	1.97	6.06
	0.50	21.37	1.48	6.92
28	0.30	51.53	2.71	5.26
	0.40	31.38	1.02	3.25
	0.50	27.55	0.53	1.92

SD: Standard deviation; VC: Variation coefficient.

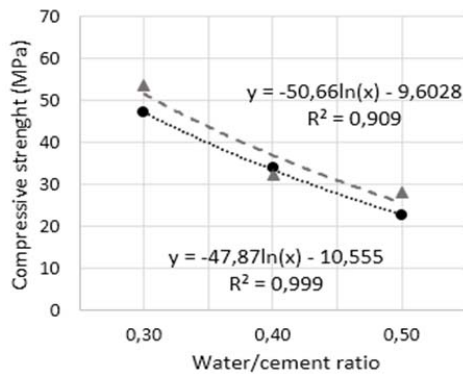


Fig. 5 Concrete compressive strength at 3 days and 28 days

A. Water Permeability Test through Ultrasonic Pulse Velocity Test

A small percentage of crack healing by the autogenous process may already be representative to extend the service life of concrete structures [7]. After opening the crack at 3 days and 28 days, ultrasonic pulse propagation velocity was monitored until 91 days. Three measurements were performed on each of the samples (Fig. 2), for a total of 27 reading points (9 samples); however, each of the points was analyzed separately, as shown in Fig. 6. In these graphs, the vertical

axis refers to the percentage of ultrasonic pulse velocity relative to the initial intact specimen, calculated according to Equation 1, and the age of each reading. The “0” day reading was performed on the same day of the crack opening, before and after the cracking process. The first reading point in each graph refers to the percentage of the crack sample that maintained its integrity in relation to the pre-cracking sample.

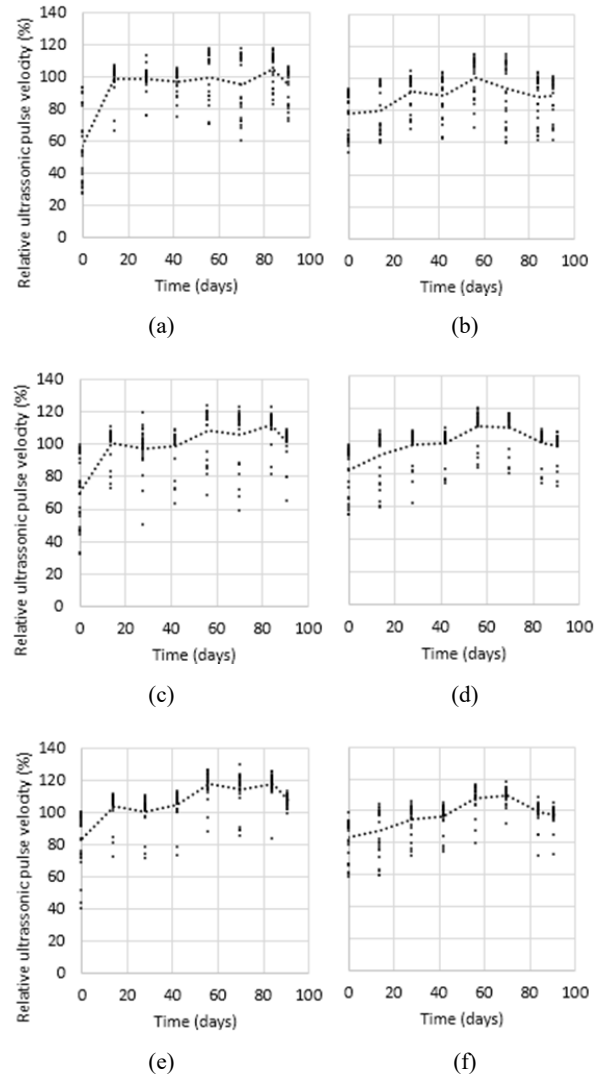
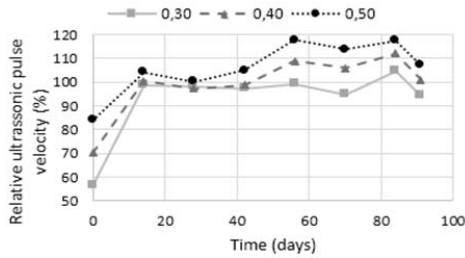


Fig. 6 Ultrasonic pulse velocity percentage of specimens with different water/cement ratios over time for two cracking ages (a) 0.30 / 3 days, (b) 0.30 / 28 days, (c) 0.40/3 days, (d) 0.40 / 28 days, (e) 0.50 / 3 days and (f) 0.50 / 28 days

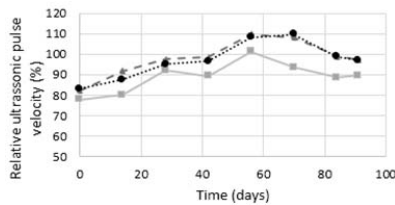
The damage caused by cracking the samples with the water/cement ratio of (a/c) 0.30, at the age of 3 days, is higher than the other a/c ratios, with greater variations, justified by the fact that this concrete presents greater toughness when compared to the others concretes (a/c ratios of 0.40 and 0.50), and therefore, great energy dissipation occurs at the moment of the load application, causing the energy accumulated during loading to dissipated abruptly and producing greater damage to the specimen. Thus, P_{t0} (ultrasonic pulse velocity relative to the initial intact specimen) is lower for smaller water cement

ratios, with thicker cracks, and is increased for concretes with higher a/c ratios.

Although the initial damage is greater for younger ages, the recovery of the crack occurs with greater intensity in this situation, which can be observed by the graphs shown in Fig. 7. This fact is due to the greater amount of anhydrous cement present in the matrix, which tends to densify and create more hydrated products due to continuous hydration. This phenomenon happens with lower intensity in the cracks opened at the age of 28 days.



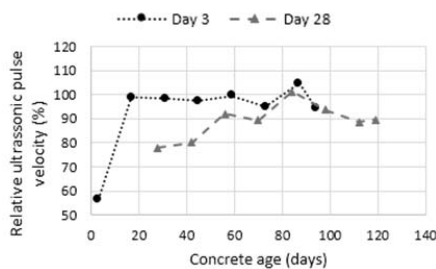
(a)



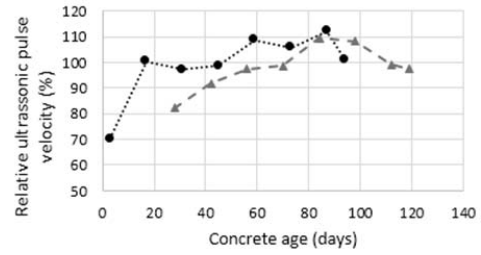
(b)

Fig. 7 Percentage of ultrasonic pulse velocity relative to the intact specimen for different w/c ratios and crack opening at (a) 3 days and (b) 28 days

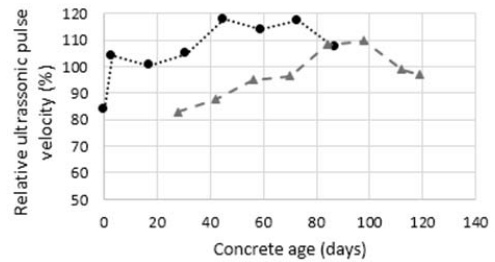
Similar behaviors were observed for cracks opened at 3 days and 28 days for different water/cement ratios, up until 80 days and 90 days the test specimens demonstrated a self-healing capability, possibly resulting from the continuous hydration of the cement; demonstrating after this age, a reduction of the percentage of ultrasonic pulse if compared to the initial intact specimen, which is possibly due to the increase in the porosity or in the damage itself. This fact can be correlated to the hydrolysis processes of the calcium hydroxide, considering that the samples were subjected to wet/dry cycles and, therefore, even with possible visual closure by the precipitation of calcium carbonate, there was a reduction in the propagation velocity of the ultrasonic pulse.



(a)



(b)



(c)

Fig. 8 Percentage of ultrasonic pulse velocity relative to the intact specimen for different ages and w/c ratios (a) 0.30, (b) 0.40 and (c) 0.50

Also, as shown in Fig. 8, it is important to explain the reason why some results overpass a 100%, meaning that the ultrasonic pulse velocity measured in these points after the cracking, results in higher readings than those before the crack opening. This can be explained by the fact that the concrete has increased its compressive strength if compared to the time of the crack opening, occurring the decrease of the capillary voids by the hydration of the cement paste over time [36]. Therefore, the ultrasonic pulse velocity increases when compared to the readings at the initial intact situation even without the complete healing of the crack. The variables inserted in this process are the condition of mass constancy, meaning the saturation of the specimen at the time of the test and the compressive strength at the same time, due to the process of hydration and the loss of this property because of the lixiviation process.

B. Cracks Visualization through Optical Microscopy Image Analysis

The cracks average width as well as the minimum and maximum readings obtained for the two crack opening ages, 3 days and 28 days for all three water/cement ratios are shown in Fig. 9. It is noticed that at 28 days, due to the continued hydration of the binder and consequent compressive strength increase, the cracks width variation becomes more evident, reducing the coefficient of determination (R^2) from 0.978 to 0.75.

This width variation may occur because these concretes have higher compressive strengths even at a young age, due to the use of high early strength cement. Larger width cracks tend to occur in concretes with higher compressive strengths when submitted to mechanical stresses, this type of fracture occurs in a fragile and abrupt way, as discussed previously.

Concretes with lower compressive strengths tend to present an inelastic deformation before its rupture, resulting in smaller width cracks than concretes with higher compressive strength [36].

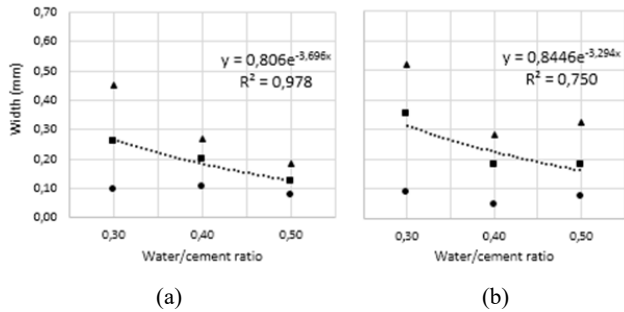


Fig. 9 Cracks width for three water/cement ratio, opened at (a) 3 days and (b) 28 days

Concerning the self-healing phenomenon, for higher water/cement ratios, which represent smaller width cracks, its healing may occur faster. Maes et al. [7] demonstrate that the percentage of crack healing is related to the initial crack width. Thus, the hypothesis of the efficiency in the self-healing of concretes with low water/cement ratios and a higher presence of anhydrous cement may not be confirmed, because even if there is not hydrated cement in the hardened matrix, the self-healing phenomenon may not occur if the cracks width is not capable of generating its own sealing. For large width cracks the gain can be obtained due to the sealing of the crack through self-healing, not overcome by the possible loss of the matrix compressive strength due to the increase of the porosity by lixiviation. Table III shows the average cracks width for both crack opening ages, at 3 days and 28 days, for all three water/cement ratios, obtained by the average of 32 readings in nine specimens.

TABLE III
 AVERAGE CRACK WIDTH OPENED AT 3 AND 28 DAYS

Water/cement ratio	Average width (mm) at 3 days	Average width (mm) at 28 days
0.30	0.2576	0.3510
0.40	0.1959	0.1816
0.50	0.1230	0.1814

The images were gathered, as described in the methodology, in eight previously demarcated zones, using a 2x magnification at the resolution of 0.65x, at the age of registration after opening the crack (0 days), reading of the first cycle (14 days) and end-of-analysis reading, after the 91 days.

Fig. 10 shows the complete image of the crack for the sample with 0.30 water/cement ratio and crack opening at 3 days, showing an average crack width of 0.1830 mm. Fig. 11 highlights a specific area from this specimen. It is possible to observe in the images recorded over time that there is a crack healing between zone 1 and part of zone 3, and the healing occurs in the smaller crack width side. The numbers presented over the image's presentation, refer to the identification of the

image collecting zone that goes from 1 to 8.

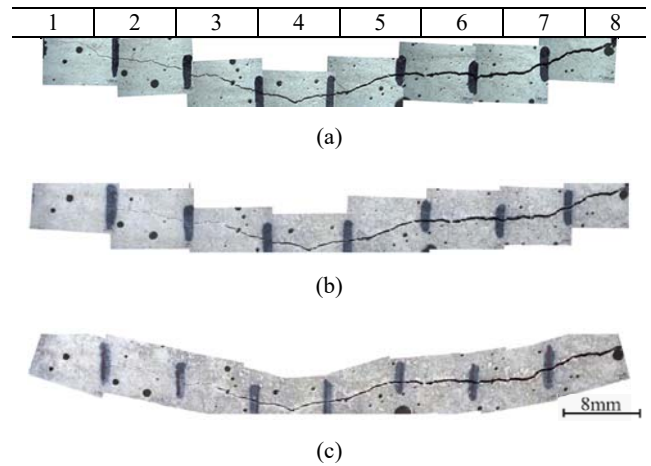


Fig. 10 Microscopy image at (a) 0 days, (b) 14 days and (c) 91 days with a/c ratio of 0.30, for cracks opened at 3 days

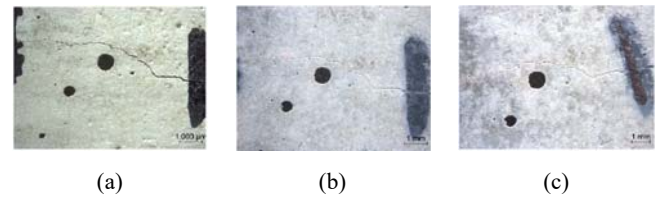


Fig. 11 Microscopy image at zone 1 for the concrete with a/c of 0.30, for cracks opened at 3 days, image taken at (a) 0 day; (b) 14 days and (c) 91 days

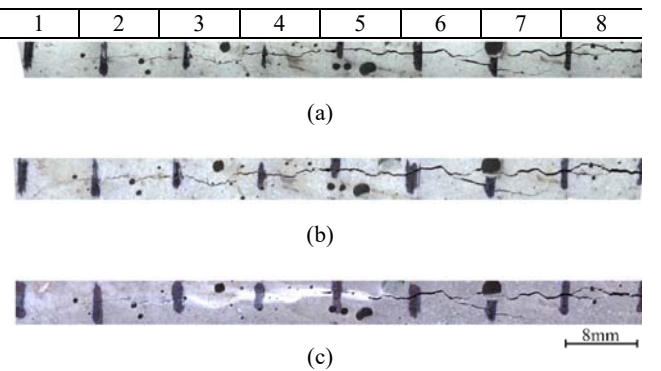


Fig. 12 Microscopy image at (a) 0 days, (b) 14 days and (c) 91 days for an a/c ratio of 0.30, for cracks opened at 28 days

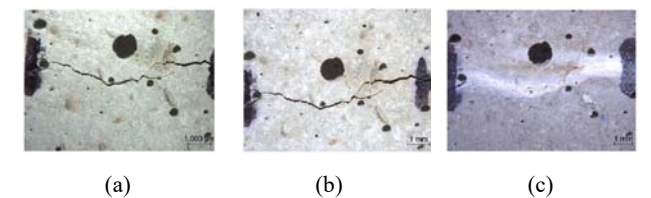


Fig. 13 Microscopy image at zone 1 for the concrete with a/c of 0.30, for cracks opened at 28 days, image taken at (a) 0 days, (b) 14 days and (c) 91 days

Fig. 12 shows the combination for the sample with water/cement ratio of 0.30 and crack opening at 28 days,

showing an average crack width of 0.1314 mm. Fig. 13 highlights a specific area from this specimen. Zone 1 to zone 3 show crack healing as early as 14 days, but at 91 days, a greater number of areas are healed, even zone 7 and zone 8, which have larger width cracks.

In autogenous self-healing, the main product created in the crack healing is the calcium carbonate (CaCO_3) from the lixiviation of the calcium hydroxide (Ca(OH)_2) [7]-[9] and hydrated calcium silicate (C-S-H) [37], [38]. The healing demonstrated by optical microscopy images is mainly related to the formation of calcium carbonate due to the lixiviation process of calcium hydroxide [39]. The precipitation of the calcium carbonate depends on the temperature, the pH and the concentration of the reagents; it is considered to be the product with greater contribution to the self-healing phenomenon [1].

Concretes that are produced with high early strength cement (CPV - NBR/Type III - ASTM), usually have a higher amount of C_3S in its composition and therefore it generates more Ca(OH)_2 [31], [36]. Thus, it is observed in concretes that are produced with this type of cement a large quantity of lixiviated products, because the Ca(OH)_2 is the main resultant product hydrolyzed in the cementitious matrix. Moreover, unlike blended cements with pozzolans, there is no consumption of the calcium hydroxide due to the pozzolanic reaction with these supplementary cementitious materials during the formation of more C-S-H [40].

Regarding the shape of the grains presented as possible particles of calcium carbonate, the morphological variation exhibited by the crystals depends on the atmospheric conditions [16]. According to [14], calcium carbonate has the most stable phase of calcite at regular environment condition, and other phases such as vaterite and aragonite can be transformed into calcite depending on the conditions in which they are exposed. In the work of [15], for example, the morphology of the calcium carbonate was influenced by the increase in temperature, and at higher temperatures the crystals are in the shape of needles, the aragonite and the vaterite, belong to the group of crystals with more hexagonal shape, and the calcite belongs to the hexagonal-cubic shape, and can occur in more than 300 shapes [41].

The agglomerated products seen in the images on the surface of the specimens represent, in many situations, the main factor contributing to the self-healing of the cracks [1], [13], [41]. This is the formation of the CaCO_3 due to the lixiviation of calcium-based products from the matrix. The hydrolysis of this compound depend on the temperature, the pH and the concentration of the reagents [1], and it is more visible with the CPV because of the highest amount of clinker in its composition (> 90%). Other authors have also demonstrated the CaCO_3 as the responsible product for the healing of the cracks, such as [7], [13].

In case of wet/dry cycles, [37] verified that the extension of the autogenous self-healing in cracks, occurs at a depth of 0 mm to 0.8-1 mm per healing with calcium carbonate. Since the crack healing is often confined to the surface of the specimen, the interior of the crack is usually not completely healed due to the higher concentration of CO_3^{2-} near the

surface of the crack, which is the ideal condition for the carbonation of calcium hydroxide.

Cracks of smaller dimension have demonstrated, during the analyzed period, a higher tendency to heal. The reason for this in cracks smaller than 0.1 mm is due to the late hydration associated with other reasons that help the autogenous self-healing [1]. The cracks generated in a concrete structure are one of the causes for its deterioration, including the appearance of pathological manifestations associated with chemical deterioration reactions. The lixiviation of the hydrated compounds, for example, is well noticed in the water of the wet/dry cycles, especially in the initial cycles. This increase in the porosity of the concrete associated with the lixiviation of the components from the hardened cement paste triggers the reduction of the compressive strength turning the concrete more vulnerable to erosion phenomenon [36].

IV. CONCLUSIONS

The widths of the crack created are larger for lower water/cement ratios and older age of crack opening. Both readings performed with the optical microscopy and the ultrasonic pulse velocity propagation, were relevant in the analysis of the self-healing process. The self-healing capacity is higher for younger cracking ages and lower water/cement ratios. Regardless of the cracking age, concretes achieve stability in the processes that can contribute to self-healing, between 80 days and 90 days of age after casting the specimens. The concretes analyzed demonstrated lower recovery by the ultrasonic pulse propagation velocity test but presented considerable amount of product formed when analyzed with the optical microscopy. Thus, under the conditions of this test, the main process of self-healing of cracks, possibly occurred through the lixiviation of the calcium hydroxide, with possible formation of CaCO . The sealing phenomenon is higher for the crack opened at the age of 3 days.

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REFERENCES

- [1] M. de Rooij, K. Van Tittelboom, N. de Belie, E. Schlangen, Self-Healing Phenomena in Cement-Based Materials. v. 11. Dordrecht: Springer Netherlands, 2013.
- [2] K. Van Tittelboom, E. Gruyaert, H. Rahier, N. De Belie, Influence of mix composition on the extent of autogenous crack healing by continued hydration or calcium carbonate formation, *Constr. Build. Mater.* 37 (0) (2012) 349–359.
- [3] P. Termkhajornkit, T. Nawa, Y. Yamashiro, T. Saito, Self-healing ability of fly ash– cement systems, *Cem. Concr. Compos.* 31 (3) (2009) 195–203.
- [4] M. Sahmaran, G. Yildirim, T.K. Erdem, Self-healing capability of cementitious composites incorporating different supplementary

- cementitious materials, *Cem. Concr. Compos.* 35 (1) (2013) 89–101.
- [5] P. van den Heede, M. Maes, N. de Belie. Influence of active crack width control on the chloride penetration resistance and global warming potential of slabs made with fly ash + silica fume concrete, *Construction and Building Materials*. v. 67 pp. 74–80, 2014.
- [6] J. H. Yu, E. H. Yang Microstructure of self-healed PVA engineered cementitious composites under wet-dry cycles. In: *Advances in Applied Ceramics*, v. 109, n. 7, pp.399-404, 2010.
- [7] M. Maes, D. Snoeck, N. de Belie, Chloride penetration in cracked mortar and the influence of autogenous crack healing. *Construction and Building Materials*, v. 115, p. 114–124, jul. 2016.
- [8] S. Z. Qian, J. Zhou, E. Schlangen, Influence of curing condition and precracking time on the self-healing behavior of engineered cementitious composites. *Cement and Concrete Composites*. v. 32, pp. 686-693. 2010.
- [9] H. Huang, G. Ye, Z. Shui, Feasibility of self-healing in cementitious materials – By using capsules or a vascular system? *Construction and Building Materials*. v. 63, p. 108–118, jul. 2014.
- [10] N. ter Heide, E. Schlangen, Self-healing of early age cracks in concrete. In: *Proceedings of the first international conference on self-healing materials*. Noordwijk aan Zee, The Netherlands, 2007.
- [11] K. Sisomphon, O. Copuroglu, E. A. B. Koenders. Effect of exposure conditions on self-healing behavior of strain hardening cementitious composites incorporating various cementitious materials. In: *Construction and Building Materials* v. 42, pp. 217-224. 2013.
- [12] V. G. Cappellesso, N. S. Petry, D. C. C. Dal Molin, A. B. Masuero, Use of crystalline waterproofing to reduce capillary porosity in concrete. *Journal of Building Pathology and Rehabilitation*. 1:9, p. 12. 2016.
- [13] V. C. Li, Y Yang, Self-healing materials: An alternative approach to 20 centuries of materials science. In: S. Van Der Zwaag, p. 161-193. Dordrecht: Springer, 2007.
- [14] Y. S. Han, G. Hadiko, M. Fuji, M. Takahashi. Effect of flow rate and CO₂ content on the phase and morphology of CaCO₃ prepared by bubbling method. *Journal of Crystal Growth*. v.276 pp. 541–548. 2005.
- [15] J. Chen, L. Xiang, Controllable synthesis of calcium carbonate polymorphs at different temperatures. *Powder Technology*. v.189 pp. 64–69. 2009.
- [16] W. Zappa. Pilot-scale experimental work on the production of precipitated calcium carbonate (PCC) from steel slag for CO₂ fixation. Thesis. School of Engineering. Department of Energy Technology. Aalto University. Finland. p.126. 2014.
- [17] H. W. Reinhardt, M Jooss, Permeability and self-healing of cracked concrete as a function of temperature and crack width. *Cement and Concrete Research*, v. 33, n. 7, p. 981–985, 2003.
- [18] O. Çopuroğlu, E. Schlangen, T. Nishiwaki, K. van Tittelbomm, D. Snoeck, N. de Belie, M. R. de Rooij, M.R. Experimental techniques used to verify healing in: *Self-Healing Phenomena in Cement-Based Materials*. v. 11. Dordrecht: Springer Netherlands, 2013.
- [19] E. Tsangouri, D. G. Aggelis, N. de belie, T. Shiotani, D. van Hemelrijck. Experimental Techniques synergy towards the design of a sensing tool for autonomously healed concrete. 18th International conference on experimental mechanics (ICEM18), v. 2, p. 449. Bélgica, 2018.
- [20] Associação Brasileira De Normas Técnicas. NBR 16697: cimento Portland – requisitos. Rio de Janeiro, 2018.
- [21] American Society For Testing And Materials. ASTM C 595: Standard Specification for Blended Hydraulic Cements. West Conshohocken, PA, 2003.
- [22] Associação Brasileira De Normas Técnicas. NBR 16605: cimento Portland e outros materiais em pó - determinação da massa específica. Rio de Janeiro, 2017.
- [23] Associação Brasileira De Normas Técnicas. NBR 7215: cimento Portland - determinação da resistência à compressão. Rio de Janeiro, 1996.
- [24] Associação Brasileira De Normas Técnicas. NBR 7211: agregados para concreto - especificação. Rio de Janeiro, 2009.
- [25] Associação Brasileira De Normas Técnicas. NBR NM 67: concreto - determinação da consistência pelo abatimento do tronco de cone. Rio de Janeiro, 1998.
- [26] Associação Brasileira De Normas Técnicas. NBR 5738: concreto - procedimento para moldagem e cura de corpos de prova. Rio de Janeiro, 2015.
- [27] Associação Brasileira De Normas Técnicas. NBR 6118: Projeto de estruturas de concreto - Procedimento. Rio de Janeiro, 2014.
- [28] American Concrete Institute – ACI 318-14: Building code requirements for structural concrete. 2014.
- [29] Associação Brasileira De Normas Técnicas. NBR 5739: concreto - ensaios de compressão de corpos-de-prova cilíndricos. Rio de Janeiro, 2018.
- [30] K. van Tittelboom, N. de Belie, Self-Healing in Cementitious Materials—A Review. *Materials*, v. 6, n. 6, p. 2182–2217, 27 maio 2013.
- [31] A. M. Neville. Propriedades do concreto. In: CREMONINI, R. A. (Tradução) 5. ed. Porto Alegre: Bookman, 2016.
- [32] W. Zhong, W. Yao, Influence of damage degree on self-healing of concrete. *Construction and Building Materials*. (S.1.). v.22, p.1137-1142. 2008.
- [33] Y. Abdel-Jawad, F. Dehn, Self-healing of self-compacting concrete. In: *Proceedings of SCC 2005*, Orlando, Florida, USA, p. 1023–1029, 2005.
- [34] A. Abd-Elmoaty, Self-healing of polymer modified concrete. *Alexandria Engineering Journal* 50(2), p.171–178. 2011.
- [35] L. Bertolini. *Materiais de construção: Patologia, Reabilitação e Prevenção*. HELENE, P. Oficina de Textos, 2010.
- [36] P. K. Mehta, P. J. M. Monteiro. *Concreto: estrutura, propriedades e materiais*. In: HASPARYK, N. P. (Ed). 2. ed. São Paulo: Ibracon, 2014.
- [37] D. Snoeck, J. Dewanckele, V. Cnudde, N. de Belie, X-ray computed microtomography to study autogenous healing of cementitious materials promoted by superabsorbent polymers, *Cement and Concrete Composites*. v. 65, pp. 83-93, 2016.
- [38] L Kan, H Shi, Investigation of self-healing behavior of engineered cementitious composites (ECC) materials. *Construction and Building Materials*. v.29, pp. 348-356. 2012.
- [39] M. Wu, B. Johannesson, M. Geiker, A review: Self-healing in cementitious materials and engineered cementitious composite as a self-healing material. *Construction and Building Materials* 28. Elsevier. 2012. p.571-583.
- [40] T. –H. Ahn, T. Kishi, Crack Self-healing Behavior of Cementitious Composites Incorporating Various Mineral Admixtures. *Journal of Advanced Concrete Technology*, v. 8, n. 2, p. 171–186, 2010.
- [41] D. Chakraborty, V. K. Agarwal, S. K. Bhatia, J. Bellare, Steady-state transitions and polymorph transformations in continuous precipitation of calcium carbonate. *Industrial & Engineering Chemistry Research*. v.33 pp. 2187-2197. 1994.

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