# EFFECT OF CARBONATED RECYCLED COARSE AGGREGATES ON THE MECHANICAL AND DURABILITY PROPERTIES OF CONCRETE

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#### Abstract

The carbonation treatment seems to be an efficient process to increase the quality of recycled concrete aggregate (RCA) and to improve the hardened properties of concrete, especially the mechanical ones, as already demonstrated in several studies. However, research on durability properties of concrete obtained with carbonated RCA is still limited and do not allow to properly optimize the concrete mix design to effectively gain environmental advantages. In this work, mechanical and durability tests were performed on three different concretes, made with a water/cement ratio equal to 0.5, and either fully natural aggregates, or with a partial replacement with RCA or carbonated RCA (cRCA). RCA and cRCA were obtained by crushing respectively old alkaline and old carbonated concretes, and a further carbonation treatment was also performed on a fraction of cRCA. From results, a limited improvement in compressive strength, electrical resistivity, water absorption and sorptivity was observed in concrete made with cRCA with respect to concrete with RCA, whilst a significant decrease of the chloride diffusion coefficient was obtained. Despite the improvements obtained with respect to concrete with untreated RCA, the

mechanical and durability performances of cRCA concrete remained on average lower than concrete made with natural aggregate.

#### Highlights

RCA and cRCA were obtained by crushing old alkaline and carbonated concretes About 30% of natural aggregate was replaced with RCA and cRCA to cast new concretes Several mechanical and durability tests were performed after different curing times The hardened properties of concrete improved when cRCA was used instead of RCA The hardened performances of concrete with cRCA remained lower than the Ref concrete

## Keywords

RCA, carbonation treatment, mechanical properties, concrete durability performances

## 1. Introduction

In an attempt to promote the circular economy also in the construction industry, the efficient re-use of recycled concrete aggregates (RCA) for the production of new concrete is a challenge that needs to be addressed and promoted in order to reduce demolition waste and achieve environmental benefits. RCA are obtained by crushing old concrete and, due to the old, adhered mortar and the previous interfacial transition zones (ITZ), RCA exhibit lower density and higher water absorption with respect to natural aggregates. Consequently, new concretes made with total or partial replacement of natural aggregates with RCA show lower mechanical and durability performances when compared to ordinary ones [1,2].

The carbonation treatment revealed to be an efficient process to enhance the properties of recycled concrete aggregates, and, as a further benefit, it allows the permanent sequestration of carbon dioxide (CO<sub>2</sub>). In this context, it has been shown that one ton of cement attached in RCA could theoretically absorb approximately 0.5 ton of CO<sub>2</sub> [3]. During the carbonation process, CO<sub>2</sub> reacts with calcium hydroxide to form calcium carbonate that, due to its higher hardness and solid-phase volume than Ca(OH)<sub>2</sub>, leads to a decrease in porosity and crack filling, which contribute to the quality improvement of carbonated RCA (cRCA) with respect to untreated RCA.

This has led to an increasing attention towards this method, as shown by the increasing number of papers on this topic and the reviews published on this emerging technology in the last years [4-8]. In general, the improvement extent of RCA was influenced by carbonation conditions, i.e. CO<sub>2</sub> concentration, duration, temperature and gas flow rate of the carbonation treatment, but also by the characteristics of RCA as particle size, compressive strength of parent materials, water content, and additional calcium source [4,9].

Many studies verified that the water absorption of RCA after carbonation could be reduced of approximately 20%-30% [10-15], although some Authors found a reduction even of the order of 60-70%, obtained adding an extra calcium source or prolonging the carbonation time [16-17]. The water absorption reduction was related to a decrease of the cumulative porosity of carbonated RCAs than that of non-carbonated RCA, due to the precipitation of calcium carbonate that occurs in the pores. The higher density of calcium carbonate with respect to portlandite led also to an increase, although limited, of the apparent density of cRCA, that was included in the range of about 0.65% and 5.62% [9-11, 13, 16].

With regard to the hardened properties of concrete incorporating cRCA, the compressive strength was generally superior to that of RCA concrete, although negligible increments were observed when the replacement percentage of carbonated RCA was between 20 and 30%, whilst relevant improvements were observed by replacing 100% of the aggregate [15, 18-20]. The compressive strength of cRCA concretes remained however always lower than that of the reference concrete, made with natural aggregates [15, 19, 21], expect some cases in which the carbonation period was prolonged [17]. This behavior was often ascribed to the densification of RCA, but also to a local densification of the interfacial transition zone, ITZ, between the carbonated RCA and the new cement mortar/paste, with an improvement of its microhardness [22]. The local densification might be ascribed to the reaction of the calcite, precipitated on the surface of carbonated RCA, with the aluminate species of the new cement to form the monocarbonate and to act as nucleation sites for the grow of C-S-H on the surface of carbonated RCA [22].

The densification of cRCA and, hence, the lower water absorption with respect to RCA, as previously described, may have direct influence on concrete durability. At this regard, chloride permeability was reduced significantly when RCA was treated by accelerated carbonation [17, 21, 23-26] and concrete obtained with carbonated RCA showed higher resistance to the penetration of carbonation with respect to RCA concrete [18, 21, 27]. In general, however, as observed for mechanical properties, the durability performances in relation to the resistance to chloride and carbonation penetration of cRCA concrete remained lower with respect to ordinary concrete made with natural aggregates [17, 21, 24, 26].

To summarize, previous studies highlighted that the carbonation treatment was an effective and economic solution to improve the quality of RCA and concrete products. Major attention has been focused on the properties of RCA after the carbonation treatment and on the mechanical properties of concrete incorporating cRCA, whilst little attention has been paid to the durability performances of cRCA concrete or cRCA mortar. To better understand if this solution is promising to produce structural concrete with comparable durability properties with respect to ordinary concrete the durability performances of cRCA concretes should be further investigated.

This paper reports the results of a preliminary experimental campaign aimed at evaluating the enhancement of performances of carbonated RCA and of hardened concrete made with carbonated RCA to confirm the potential of CO<sub>2</sub> treatment. Three types of concrete were made, with w/c ratio of 0.5, and either fully natural aggregates, untreated RCA, or carbonated RCA. The latter were obtained with the substitution of the two coarsest natural aggregate fractions with RCA from alkaline concrete, and with RCA from carbonated concrete that were subjected to a further carbonation treatment on the coarsest fraction, respectively. Major attention was devoted to the investigation of hardened properties of concretes related to mechanical strength and durability performances such as the resistance to the penetration of aggressive agents, i.e. water, carbonation and chlorides. In particular, compressive strength and concrete electrical resistivity after different

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times of moist curing, water absorption, sorptivity and resistance to chloride and carbonatation penetration were evaluated on the three types of concrete.

#### 2. Materials and methods

## 2.1 Materials and mixes

Three concretes, with a water/cement ratio of 0.5, were prepared with 420 kg/m<sup>3</sup> of a limestone Portland cement designated according to European standard EN 197 as CEMII/A-LL 42.5R, 210 kg/m<sup>3</sup> of fresh water and 1677 kg/m<sup>3</sup> of aggregate, divided in six different classes - two sands (0-2 mm and 0-4 mm) and four gravels (2.36-4.75 mm, 4.75-8 mm, 7.5-9 mm and 9-16 mm). An acrylic high range water reducing superplasticizer (according to EN 934-2 standard) (0.5% in mass vs cem) was added to the mixes in order to achieve a class of consistency S5 according to EN 206-1 standard.

As reference mix (Ref), natural crushed limestone aggregate (NA), was used, whilst in RCA and cRCA mixes the two coarsest aggregate fractions, i.e. 7.5-9 mm gravel and 9-16 mm gravel, that corresponded to the 29% of the total amount of aggregate, were replaced with recycled concrete aggregates (RCA), respectively in alkaline and carbonated conditions, maintaining the fine NA. To obtain the RCA, old concrete specimens (stored from previous researches and made with Portland and Portland-limestone cement, with w/c ratio ranging from 0.46 to 0.61 and crushed limestone aggregate), were crushed to obtain a fraction passing between 7.5 mm and 16 mm (corresponding to the minimum and maximum dimensions of the two coarsest fractions of natural aggregate). The majority of old specimens were exposed indoor for about 15 years, which promoted the carbonation of the outermost layer up to a depth of 40 mm. During crushing, phenolphthalein was sprayed on the surface of the crushed aggregate to separate the alkaline (RCA) from the carbonated (nat-cRCA) ones and after separation both RCA and the nat-cRCA were sieved to obtain the two fractions. Figure 1 shows the results of the phenolphthalein test carried out on a bunch of RCA aggregate and a pile of alkaline and carbonated RCA aggregate obtained after separation. Faded pink areas can be observed on nat-cRCA, suggesting that carbonation could be only partial. Hence, nat-cRCA

aggregates, with dimension between 7.5 and 9 mm, were exposed to a further carbonation treatment before mix, in a carbonation chamber with a flux of 100% CO<sub>2</sub> ( $T = 20^{\circ}C$  and R.H. = 65%) for approximately one month (100-cRCA). For casting cRCA concrete, nat-cRCA and 100-cRCA were mixed together.



Figure 1– Phenolphtalein test on a bunch of RCA (a) and a pile of alkaline (b) and naturally carbonated (c) RCA

In the mixes, aggregates were combined to fit the Fuller's grading curve. Figure 2 shows the grading curve of each fraction as well as Fuller's grading curve and the obtained combination (minimum and maximum dimension of each fraction were determined graphically and they were not reported). For both coarsest fractions, i.e. 7.5-9 mm and 9-16 mm, the grading curves of RCA, both alkaline and carbonated, were quite close to those of natural aggregate. The 9-16 mm RCA gravel had a higher passing for each opening, suggesting that the particles were slightly smaller than those of the natural aggregate and that the minimum diameter was lower, whilst the maximum diameter was comparable. However, these differences were neglected, and the optimal combination of aggregate defined for natural aggregate, was considered suitable also for RCA.



Figure 2 – Grading curve of each fraction of natural aggregate and Fuller's and combination grading curves (pink and grey symbols represent respectively alkaline and carbonated RCA)

Before the mixing, the coarse aggregate (NA, RCA, and cRCA) was pre-wetted to reach the saturated surface dry condition to avoid effects on the w/c ratio due to a higher water absorption of RCA. The aggregates and the cementitious materials were firstly introduced in the mixer and mixed for about 30 s to homogenize the materials. Then, the water was added and mixed for about 3 min. Concrete was poured into molds on a vibrating table with a vibration time useful to fully compact it. The moulds were covered with a plastic sheet and stored in laboratory at 20°C. After 24 hours the specimens were demoulded and curing continued, in a curing chamber, at 20°C and 95% relative humidity, until specimens were tested (duration of curing depended on the performed type of test and will be specified later).

#### 2.2 Testing

## 2.2.1 Determination of the properties of aggregate

The water absorption of RCA and carbonated RCA was tested according to EN 1097-6 [28]. The water absorption,  $A_{agg}$ , was evaluated from the difference between the saturated surface dry mass ( $M_{ssd}$ ) after vacuum saturation with distilled water and oven-dry mass ( $M_d$ ) after 24 h drying at 105°C, as:

$$A_{agg} = (M_{ssd} - M_d) / M_d \cdot 100 \,(\%) \tag{1}$$

The water absorption test was carried out on three bunches of about 0.5 kg each one. For cRCA, the test was carried out on the nat-cRCA and after the carbonation pre-treatment on 100-cRCA.

#### 2.2.2 Compressive strength of concrete

Compressive strength of concrete was measured on two replicate cubic specimens (100 mm side length) after 7, 28 and 90 days of moist curing, according to EN 12390-3 [29]. On the same specimens, before the compressive test, density was measured as the ratio between the mass, evaluated just after the removal from curing chamber, and the volume.

## 2.2.3 Electrical resistivity of concrete

On the same specimens used for compressive strength and density evaluation, measurements of the bulk electrical resistivity, through the uniaxial method, were carried out with weekly frequency. Concrete specimens were placed between two metallic and parallel plates (electrodes), with the interposition of two wet sponges to guarantee the electrical contact between the electrodes and the concrete specimen, and the conductance was then measured with a conductimeter. Then, from conductance, electrical resistivity was evaluated using as cell constant the ratio between the side length of the specimen and the surface area in contact with the electrodes, which was equal to 10 m<sup>-1</sup>.

## 2.2.4 Absorption and sorptivity of concrete

Sorptivity test was performed according to EN 13057 standard [30] on two replicate cylindrical specimens for each concrete mix, with a diameter of 100 mm and cured 28 days. Initially, the specimens were dried in an oven at approximately 40 °C until constant mass, M<sub>d</sub>, then they were placed in a tray with the bottom surfaces in contact with water for a height of approximately 2 mm. After different time intervals up to 24 h, the specimens were removed from the tray and weighed to evaluate mass gain. The water absorption per unit surface was then plotted versus the square root of time and the slope of the best-fit line was defined as the sorptivity coefficient of concrete. The

specimens were then saturated to evaluate the saturated mass, M<sub>s</sub>, and the water absorption, A, was evaluated according to:

$$A = (M_s - M_d) / M_d \cdot 100 \,(\%) \tag{2}$$

#### 2.2.5 Resistance to chloride penetration of concrete

To evaluate the resistance to chloride penetration, cylindrical specimens with diameter of 100 mm and cured 28 days, were exposed to the immersion test according to EN 12390-11 [31] and to the rapid chloride migration test, according to NT-BUILT 492 standard [32]. Each test was performed on two replicate specimens of the same concrete type. To perform the immersion test, after the preparation of the specimens according to the standard, the specimens were immersed in a 3% by mass sodium chloride solution, for 90 days. To assure the unidirectional penetration of chlorides, before the exposure, the lateral and the bottom surfaces were sealed with epoxy resin. At the end of the exposure time, powder was collected at eight different depths, up to a depth of 25 mm, by means of grinding and the chloride concentration was evaluated through chemical titration. The chloride profiles were then interpolated (excluding the first point) to evaluate the chloride diffusion coefficient, D<sub>app</sub>, and the surface chloride concentration, C<sub>s</sub>, through the Fick's second law for chloride diffusion:

$$C(x,t) = C_s \left[ 1 - erf\left(\frac{x}{2\sqrt{D_{titr} \cdot t}}\right) \right]$$
(3)

Where C(x,t) is chloride concentration at depth x and time t, and erf is the error function. To perform the rapid chloride migration test, after the saturation in a Ca(OH)<sub>2</sub> solution, one surface of the specimens was exposed to a catholyte solution, 10% NaCl, while the upward face was exposed to an anolyte solution, 0.3 M NaOH and connected to the power supply. The voltage and the duration of the test were set in accordance to the method prescriptions. At the end of the exposure, a colorimetric test was performed on the split surface, to evaluate the chloride penetration profile. A 0.1 M silver nitrate solution was sprayed on freshly split surfaces, and the measurement of chloride penetration depth was performed in seven different points. Chloride penetration depth measurements were then used to evaluate the non-steady state migration coefficient,  $D_{RCM}$  (m<sup>2</sup>/s), according to the following correlation:

$$D_{RCM} = \frac{RT}{zFE} \cdot \frac{x_m - \alpha \sqrt{x_m}}{t}$$
(4)

where R is the gas constant (8.314 J/K mol), T is the average value of the temperatures in the anolyte solution (K), z is the absolute value of ion valence (1 for chloride ions), F is the Faraday constant (9.648×104 J/V mol), x is the average value of the penetration depth measurements (m), t is the duration of the test (s), E is the actual potential difference across the specimen, U the external applied potential between the two electrodes (V), L is the concrete specimen thickness (m). The  $\alpha$  parameter in (4) is calculated as:

$$\alpha = 2 \sqrt{\frac{RT}{zFE}} erf^{-1} \left( 1 - \frac{2c_d}{c_0} \right)$$
<sup>(5)</sup>

where  $c_d$  is the chloride concentration at which the colour change is observed (assumed equal to 0.07 N) and  $c_0$  the chloride concentration of the test solution (2 N).

## 2.2.6 Resistance to carbonation penetration of concrete

Concrete resistance to the penetration of carbonation was evaluated on prismatic specimens,  $100 \times 50 \times 200$  mm, both in natural and accelerated conditions. Regarding the natural condition specimens were cured 7 days, and then two replicate specimens for each mix, were exposed indoor  $(T = 20^{\circ}C \text{ and R.H.} = 55\text{-}65\%)$ , one specimen for each mix was exposed to outdoor conditions, sheltered from rain and sun, and one specimen for each mix in unsheltered conditions on the roof of the Department of Chemistry, Materials and Chemical Engineering of Politecnico di Milano. The four lateral faces of the prismatic specimens were masked with epoxy, so that carbonation was allowed to penetrate only from two opposite faces (the cast and a mould surfaces). During exposure,

the unmasked faces were oriented vertically. After about 300 days of exposure, the specimens were split, and the carbonation depth on the fracture surface was highlighted by spraying a solution with phenolphthalein. Carbonation depth measurements were taken from both the exposed surfaces (cast and mould) in nine different points spaced 10 mm from each other, and then the average value was determined.

Two replicate specimens for each type of concrete, cured 28 days and subsequently dried for about 14 days at laboratory conditions, were exposed to an accelerated carbonation chamber ( $CO_2 = 3\%$ , T = 20°C, R.H. = 65%), after masking the lateral surfaces with epoxy. Interruption and disruption occurred during the emergency due to COVID-19 pandemic situation, and, hence, the duration of the exposure period, as well as the effective CO<sub>2</sub> concentration, were not clear. Anyway, specimens were split and the carbonation depth was measured on nine points, spaced 10 mm from each other, of the fracture surface, considering that the specimens were exposed to the same exposure conditions of the behavior in relation to carbonation resistance could be anyway determined.

## 3. Results and discussion

#### 3.1 Properties of aggregate

RCA was obtained by crushing old concrete specimens made with different types of cement and water/cement ratio, with very different mechanical and durability properties. On those concretes, compressive strengths measured after 28 days of wet curing varied between 40 and 90 MPa, whilst water sorptivity between about 0.35 and 1.5 kg/m<sup>2</sup>·h<sup>1/2</sup> [33]. The parent materials used to produce RCA were, then, quite variable and might affect their properties.

For each type of aggregate, i.e. alkaline (RCA), naturally carbonated (nat-cRCA), and the naturally carbonated after the pre-treatment with accelerated carbonation (100-cRCA), only the water absorption was evaluated. Figure 3 shows the results obtained for the two fractions on three bunches. It can be observed that, for both the fractions, the water absorption of nat-cRCA was slightly lower than that of RCA. For instance, for 7.5-9 mm gravel the water absorption decreased

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of only 3% when the aggregate was carbonated, confirming that un-treated cRCA could not have been fully carbonated. After the accelerated carbonation treatment, the water absorption of 100cRCA was reduced of approximately 20% in comparison to RCA, in accordance with values obtained by other Authors [10-15]. A lower water absorption might be associated to a decrease in porosity of 100-cRCA in comparison to RCA, occurred during the carbonation treatment due to the precipitation of calcium carbonate in the pores and also in the cracks formed after crushing. On 9-16 mm gravel fraction, the water absorption of nat-cRCA was reduced of about 12%. This confirmed that for this gravel fraction most of the aggregates were effectively carbonated.



Figure 3 – Water absorption of RCA

## 3.2 Hardened properties of concrete

#### **3.2.1** Compressive strength

Figure 4 shows the evolution in time of density and compressive strength evaluated on two replicate specimens for the three concretes. An increase of compressive strength with time can be observed, as expected. It can be observed that the density of RCA concrete was the lowest at any curing time, suggesting a higher porosity, whilst the density of cRCA concrete resulted slightly higher than that of RCA concrete, but lower than that of Ref concrete. Regarding compressive strength, on 7-day cured concretes comparable values, about 44 MPa, were measured on Ref and cRCA concretes, whilst on RCA concrete a slightly lower value, about 40 MPa, was determined. After 28 and 90

days of curing RCA concrete showed the lowest compressive strength. These results confirmed, as stated in the literature [15, 18-20], a lower strength of concrete made with recycled concrete aggregates in comparison to concrete made with natural aggregates. However, slight differences between Ref and RCA concretes were observed, and, in accordance with Xuan et al. [18], this might be due to the limited amount of natural aggregate replaced with RCA, that corresponded to about the 30% of total aggregate and 50% of the gravel, i.e. coarse aggregate. The observed tendency probably depended also on the parent materials used to produce the RCA and, in the literature, concretes with different compositions were used that led to a different trend of that observed in this work. When RCA aggregates were carbonated comparable compressive strength after 28 days of moist curing with respect to the reference concrete was obtained, about 47 MPa and 48.5 MPa, respectively. The improvement in performances of cRCA in comparison to RCA could be ascribed to the lower porosity and to an improvement of the ITZ as suggested by Zhan et al. [22].



Figure 4 - Trend in time of compressive strength (circles) and density (crosses) of the three

#### concretes

## 3.2.2 Electrical resistivity

The average bulk electrical resistivity of concrete prepared with the non-carbonated and the carbonated RCA, measured after different days of moist curing, is shown in Figure 5. It was not possible to measure resistivity on Ref concrete specimens after 7 days of curing. It can be observed,

in general, an increase of resistivity increasing the curing time, especially in the first days, due to a refinement of pore structure for the proceeding of cement hydration. Being the electrical resistivity correlated to the interconnectivity of pores network, it gives a direct indication of the resistance to ionic transport. After one day of moist curing, resistivities of RCA and cRCA concretes were significantly lower than that in Ref concrete (about 20  $\Omega$ m versus 35  $\Omega$ m), conversely with the proceeding of hydration the differences among the concretes decreased, in accordance with results obtained in Xuan et al. [24] for percentage replacement of aggregate between 20 and 40%. cRCA concrete always showed higher resistivity than those of RCA concrete approaching about 60-70  $\Omega$ m after 90 days of curing. Recycled concrete aggregate was partially constituted by cement paste, from crushed old concrete, and hence a major amount of cement paste was present in RCA concrete, leading to a higher current flow than in Ref concrete (therefore to a lower electrical resistivity). The decrease of porosity in cRCA led to a decrease of the current that flows through the recycled aggregate, and therefore to an increase of resistivity with respect to RCA concrete.



Figure 5 – Electrical resistivity measured after different times of moist curing on the three concretes **3.2.3 Absorption and sorptivity** 

Figure 6 reports the results of absorption and sorptivity test, carried out after 28 days of moist curing on the three concretes. The reference concrete showed the slowest water uptake, with an average sorptivity coefficient around  $1.16 \text{ kg/m}^2 \cdot h^{1/2}$ , whilst the concretes made with recycled

aggregates showed higher sorptivity coefficients, around 1.46 kg/m<sup>2</sup>·h<sup>1/2</sup> for RCA and 1.34 kg/m<sup>2</sup>·h<sup>1/2</sup> for cRCA. An improvement of water uptake was, indeed, observed employing carbonated RCA that may be linked to the pore structure densification of the RCA resulting in improved durability performances.

An increase of water absorption, of the order of 30%, was observed on the two concretes with recycled aggregates with respect to the Ref concrete, similar to those obtained in Kazmi et al. [21]. However, the values were comparable and around 6.6% for both RCA and cRCA concretes, without showing any improvement due to the carbonation treatment.



Figure 6 – Absorption, A (grey bars), and sorptivity coefficient, S (blue bars), evaluated on 28-day cured specimens made with NA and RCA aggregate

## 3.2.4 Resistance to chloride penetration

Corrosion in the reinforced concrete structures due to chloride penetration is considered as one of the most severe degradation processes. In order to evaluate the resistance to chloride penetration two different types of tests were performed on 28-day cured specimens: a uniaxial diffusion test and an accelerated test, i.e. the rapid chloride migration test. Figure 7 reports the apparent diffusion coefficient (D<sub>app</sub>) and the chloride surface concentration (C<sub>s</sub>) evaluated by fitting through equation 3 the chloride profiles obtained after the diffusion test, and the rapid chloride migration coefficient (D<sub>RCM</sub>) evaluated after the migration test with the colorimetric method. Compared with the reference concrete, the apparent diffusion coefficient increased by replacing the natural aggregate with RCA. The reference concrete showed a  $D_{app}$  of about 23 m<sup>2</sup>/s that increased approximately to 37 m<sup>2</sup>/s and 28 m<sup>2</sup>/s in RCA and cRCA concretes, respectively. The  $D_{app}$  was therefore 1.6 and 1.2 times higher for RCA and cRCA concretes with respect to Ref concrete, respectively. No significant difference in terms of chloride surface concentration was observed among the three concretes. From the results of rapid chloride migration test, a  $D_{RCM}$  of about 22.2 $\cdot 10^{-12}$  m<sup>2</sup>/s was obtained for the reference concrete, significantly higher for the RCA concrete (around  $38 \cdot 10^{-12}$  m<sup>2</sup>/s), while just slightly higher than reference for cRCA concrete, around  $24 \cdot 10^{-12}$  m<sup>2</sup>/s. Similarly to the results of the diffusion test, the  $D_{RCM}$  resulted therefore 1.7 and 1.1 times higher in RCA and cRCA concrete with respect to Ref concrete. The increase of the chloride diffusion coefficient in RCA concrete obtained through both tests could be ascribed to the higher porosity and water absorption of RCA aggregate that promoted the penetration of aggressive agents. The use of carbonated RCA enhanced the resistance to chloride-ion penetration in comparison to untreated recycled concrete aggregate.



Figure 7 – Apparent diffusion coefficient, D<sub>app</sub> (grey bars), chloride surface concentration, C<sub>s</sub> (black circular symbols), and rapid migration coefficient, D<sub>RCM</sub> (blue bars) evaluated on 28-day cured specimens made with NA and RCA aggregates

#### 3.2.5 Resistance to carbonation penetration

Another mechanism that can induce the corrosion of steel reinforcement is the carbonation of concrete that, leading to a decrease of the pH, can cause the passive film on steel surface to become unstable. Even in chloride-bearing environment, carbonation cannot be neglected and need to be properly investigated. The resistance to carbonation penetration was, then, evaluated both in natural and in accelerated conditions. Figure 8 shows the average carbonation depth measured on the mould and cast surfaces of the three concretes exposed in the different exposure environments, with the variation evaluated considering the minimum and maximum values measured on all the specimens. Despite a quite high variability in terms of carbonation depths and slight differences comparing the three concretes, some trends can be observed considering the results referred to the mould and the cast surfaces.

In indoor conditions, higher carbonation depths were measured on RCA concrete in comparison to Ref concrete, whilst cRCA concrete had lower carbonation depths in comparison to RCA concrete and similar to Ref concrete. For instance, on the mould surface, average carbonation depths of 2.7 mm, 2.7 mm and 2.1 mm were measured respectively for Ref, RCA and cRCA concretes. A similar trend was observed when specimens were exposed under sheltered condition and, surprisingly, only the Ref concrete had a highest carbonation depth in sheltered conditions in comparison to indoor one. Under natural unsheltered carbonation, as expected, the lowest carbonation depths, of the order of 2-4 mm after about 300 days of exposure, were measured. In fact, during raining periods, concrete was getting wet, and the partial saturation of concrete pores with water hindered the penetration of carbon dioxide. Under this exposure condition the three concretes behaved differently than in the other exposure conditions and the lowest carbonation depths were detected on cRCA concrete. Values measured in accelerated exposure were in general higher than those under natural carbonation and, also for this exposure condition, the highest values were measured on RCA concrete.

A systematic effect of the replacement of NA with RCA on the carbonation depth was not observed and the behaviour in relation to the penetration of carbonation might depend on a combination of several factors. In RCA concrete a higher amount of cement paste was present, but this has two consequences that have opposed effects on carbonation: a higher amount of portlandite and a higher total porosity of concrete. The former hinders carbonation, the latter promotes it. The final behaviour depends on which factor is prevalent between the two. cRCA concrete had a higher amount of cement paste in comparison to Ref concrete, that, however, did not necessarily mean a higher amount of Portlandite, being them carbonated. Again, a higher total porosity of concrete was expected and, hence, in this case which effect was prevalent cannot be easily detected. Results after longer exposure times are required to better investigate the effect of the use of RCA and cRCA on the resistance to the carbonation penetration. At this regard it is worth of mentioning that the measured carbonation depths, lower than 10 mm, were smaller than the maximum dimension of the recycled concrete aggregates in the concrete. This might affect the behaviour in relation to the penetration of carbonation and, hence, carbonation depth of at least the maximum dimension of aggregate needs to be reached to better understand the behaviour of these concretes in relation to carbonation resistance.





Figure 8 – Carbonation depth evaluated on the mould (a) and cast (b) surfaces of specimens made with NA and RCA aggregate and exposed to different exposure conditions

## **3.3 Effect of the carbonation treatment**

Results obtained in this work, concerning both mechanical and durability properties obtained replacing about 30% of the total amount of natural aggregate with RCA, allow to infer some useful indications on the benefits related to a carbonation treatment of RCA. Firstly, it is useful to observe that the carbonation treatment allows to reduce the water absorption of RCA (Figure 3) and, hence, to improve the quality of the recycled aggregate. The carbonation treatment, with accelerated carbonation carried out on a single fraction (7.5-9 mm gravel), led to further benefits. These are reflected in an improvement of the properties of hardened concrete, even if each property is affected to a different extent. Figure 9 shows the percentage variation (increment or decrement) of each single performance of hardened concrete obtained when cRCA was employed instead of RCA (Figure 9a) or when RCA and cRCA were employed instead of natural aggregate (Figure 9b). For some properties (i.e. compressive strength and electrical resistivity) a positive variation implies an improvement, since they are resistance parameters, whilst for other properties (i.e. water absorption, sorptivity, diffusion coefficient, carbonation depth) a positive variation is associated to a decrease in the durability performances. It can be observed that when RCA were replaced with cRCA the

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compressive strength improved and an increase of about 10% was evaluated after 7 days of curing. For longer curing times the increase of the strength was limited to values around 2-4%. Electrical resistivity experienced a significant increase after 28 days of curing, around 13%, whilst after longer curing times a reduction seemed to occur. Concerning the durability properties of concrete, although a benefit of the carbonation treatment was not observed on the water absorption (the reduction was lower than 1%), sorptivity was reduced by 8%, whilst a significant reduction was observed on the diffusion coefficient (37% and 24% respectively for  $D_{RCM}$  and  $D_{app}$ ). The carbonation penetration depth was also reduced of about 13-17% (carbonation depth in unsheltered environment constituted an exception). Hence, the carbonation treatment demonstrated to be an effective mean to improve the quality of RCA and to improve the hardened properties of concrete made with recycled concrete aggregates. However, Figure 9b shows that in comparison to concretes made with NA, the use of RCA and cRCA led to a worsening of all the properties, although limited for several of them. Regarding compressive strength, the reduction was lower than 5% after longer curing times (higher than 28 days). This means that the mechanical performances of ordinary concrete could be easily guaranteed in cRCA concrete with minimum variation of the mix design (for instance a reduction of the water/cement ratio), at least when the percentage of replaced NA by cRCA is around 30% as in this study. A significant increase, around 40%, of the water absorption can be observed, both for RCA and cRCA concretes. A slight lower variation was measured for sorptivity, with values of 25% and 15% for RCA and cRCA concretes respectively. The chloride diffusion coefficient was strongly affected and an increase of 70% and 60% was measured for DRCM and Dapp when NA was replaced with RCA. The use of cRCA, although led to an improvement in comparison to RCA, led to an increase of DRCM and Dapp of about 8% and 20% with respect to the Ref. The effect of the use of RCA and cRCA on the carbonation penetration depth was unclear, since after short-term tests either an increase or a decrease was obtained, depending on the exposure conditions. Although almost all durability parameters experienced a worsening when NA aggregate was replaced with cRCA, the limited variation suggested that an optimization of the mix design, in

terms of water/cement ratio, cement content and percentage of replaced aggregate, as well as a prolonged carbonation treatment, might assure comparable properties to ordinary concrete.



Figure 9 – Variation of different properties when RCA was replaced with cRCA (a) and when NA was replaced with RCA and cRCA (b) (grey bars: cRCA, pink bars: RCA)

## Conclusions

In this study, mechanical and durability tests were carried out on concretes with a water/cement ratio equal to 0.5 and made by replacing the 30% of the total natural aggregates with RCA and cRCA obtained by crushing respectively old alkaline and carbonated concretes, with compressive strength in between 40 and 90 MPa. A carbonation treatment was also performed on a fraction of cRCA. On the basis of the experimental results the following conclusion can be drawn:

- in carbonated recycled concrete aggregate, cRCA, the water absorption decreased in comparison to RCA, likely due to the precipitation of calcium carbonate in the pores. The carbonation treatment of RCA led to a further reduction of the water absorption, maybe due to the filling of the cracks, formed after crushing. An improvement of the quality of aggregate could be gained when the RCA are fully carbonated;

- compressive strength of cRCA concrete was higher in comparison to RCA, gaining 3% after 28 days of curing. In comparison to Ref concrete, a reduction lower than 5% was observed for cRCA concrete for curing times higher than 28 days, whilst RCA experienced a higher reduction that, however, was limited;

- concrete electrical resistivity of Ref concrete was higher than that of RCA and cRCA concrete, of approximately 10% after 90 days of curing. The cement paste incorporated in RCA aggregates, allowing the current flow, might decrease the electrical resistivity;

- water absorption of cRCA concrete was comparable to that of RCA concrete, whilst sorptivity decreased of approximately 8%. An increase in water absorption and sorptivity of about respectively 40% and 15% was observed when NA was replaced with cRCA. A significant decrease of the diffusion coefficient of about 25-50%, and therefore a marked improvement in the resistance to chloride penetration, was observed, depending on the test performed, when carbonated recycled concrete aggregate was used in place of RCA. The performances of cRCA concrete remained anyway lower than that of Ref concrete of approximately 7-15%. The effect of the carbonation treatment on the carbonation depth was unclear and needs to be further investigated at longer exposure times;

- When cRCA are used, comparable mechanical and durability properties with respect to ordinary concrete might be achieved by tailoring the mix design and prolonging the duration of the carbonation treatment.

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