



Coupling of attrition and accelerated carbonation for CO₂ sequestration in recycled concrete aggregates

Glaydson S. Dos Reis^{a,b}, Bogdan Cazacliu^{a,*}, Riccardo Artoni^a, Jean-Michel Torrenti^c, Carlos S. Hoffmann^{b,d}, Eder C. Lima^b

^a Université Gustave Eiffel, MAST, GPEM, Bouguenais, France

^b Graduate Program in Metallurgical, Mine and Materials Engineering (PPGE3M). School of Engineering, Federal University of Rio Grande do Sul, Center of Technology, Mineral Processing Laboratory, Porto Alegre, Brazil

^c Université Gustave Eiffel, MAST, Marne-la-Vallée, France

^d Departament d'Enginyeria Minera, Industrial i TIC (EMIT), Escola Politècnica Superior d'Enginyeria de Manresa (EPSEM), Universitat Politècnica de Catalunya (UPC), Av. Bases de Manresa 61–63, 08242 Manresa, Spain

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ABSTRACT

An accelerated carbonation method was applied to improve the ability of recycled aggregates to store CO₂ through the rolling carbonation process; moreover, recycled concrete aggregates' physicochemical properties to produce secondary aggregates for civil engineering applications were explored and evaluated. The carbonation tests were carried out in a laboratory-scale air-proof rotating drum equipped with a CO₂ feeding system. Performing carbonation in a rotating drum promotes selective attrition of concrete aggregates. The process accelerates the carbonation by removing the carbonated zones on the surface of coarse aggregates and then facilitating the access of CO₂ to un-carbonated zones. The tests were performed at normal atmospheric conditions, with a partial pressure of CO₂ kept at about 0.75 bar. The optimal initial moisture of the concrete aggregate was found. The main tests were conducted on 10/20 mm aggregate, but it was shown that the carbonation process was 20% faster for smaller size aggregate (5/8 mm). The rolling speed, initial moisture content, and mass amount played an important role in CO₂ sequestration. The most efficient rotating carbonation condition was reached at 100 rpm, aggregate moisture around 8%, and 500 g of aggregates in a 5 L reactor at any partial pressure.

The dynamic process highly increased the carbonation kinetics compared with static carbonation of the same duration. The CO₂ sequestered was up to 80 mg/g of aggregate, estimated to be around 60% of the potential of CO₂ capture of the tested recycled concrete aggregate. The number of revolutions appears to be a relevant process parameter concerning the carbonation kinetics. The carbonation duration acts in a complementary way as in a static process.

Some physical properties of the recycled concrete aggregate were determined. As a result, the carbonation process reduced water absorption and enhanced the density of the aggregates.

1. Introduction

The capture of carbon dioxide by mineral (Rahman et al., 2017) carbonation is an emerging, simple-to-perform technology by which carbon can be permanently stored (Sanna et al., 2014). The degree of carbonation depends on the material used for sequestering the CO₂, and calcium or magnesium-containing primary minerals are recognized as the most promising materials for the permanent and safe storage of carbon dioxide. Besides, suitable alkaline solid residues can be a promising alternative (Huijgen et al., 2005), combining the recovery of

solid and gaseous waste. In this perspective, cementitious/concrete materials have been widely used to sequester CO₂ (Bertos et al., 2004). Cementitious/concrete materials are also one central part of the construction and demolition waste generated in most of the world, and so, their availability is high. Such waste materials can be potentially reusable in the production of high-quality materials in the construction industries. However, their mechanical properties should be improved (Silva et al., 2014) and the effect of the carbonation on the pore structure of hydrated cement pastes (Bier 1987) has a positive effect on the recycled aggregates' quality (Zhao et al., 2018) and for the quality of the concrete

* Corresponding author.

E-mail address: bogdan.cazacliu@univ-eiffel.fr (B. Cazacliu).

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Table 1
Comparison of several studies and their results on CO₂ uptake by using different aggregates and carbonation systems.

Material used	Carbonation system	Experimental Conditions	CO ₂ uptake (mg/g)	Ref.
Bottom ash pH: 13.1 Particles size: 5 & 10 mm Moisture: 22–33%	Fixed bed reactor, 27 L Gas flow: 24 NL/h	Number of layers: 1-3 Pressure: 1 bar CO ₂ molar ratio: 100% Temperature: ambient Treatment time: 8–14 h	22	Lombardi et al. (2016)
Bottom ash 5.14% of Ca(OH) ₂ Particles size: 11 mm Moisture: 15%	Fixed bed reactors, 1 L manual pressure regulation	Number of layers: 6 Pressure: 1 bar CO ₂ molar ratio: 100% Temperature: ambient Treatment time: 240 min	15	Brück et al. (2018a)
Recycled concrete aggregates 11–25% of mortar Particles size: 5–10 & 10–20 mm	Fixed bed reactor, 100 L automatic pressure regulation	Pressure: 1.1 & 6.0 bar CO ₂ molar ratio: 100% Temperature: 25 °C Relative Humidity: 50% Treatment time: 24 h	8	Xuan et al. (2016)
Bottom ash pH: 13.1 Particles size: 10 mm Moisture: 22–33%	Rotating drum, 18 L Gas flow of 24 NL/h Filling ratio: 10–30%	Rotating speed: 2.5–5 rpm Pressure: 1 bar CO ₂ molar ratio: 100% Temperature: ambient Treatment time: 8–14 h	36	Lombardi et al. (2016)
Bottom ash 5.14% of Ca(OH) ₂ Particles size: 11 mm Moisture: 15%	Rotating drum, 71 L automatic pressure regulation Filling ratio: <5%	Rotation speed: 1.5 rpm Pressure: 1 bar CO ₂ molar ratio: 100% Temperature: ambient Treatment time: 120 min	26	Brück et al. (2018a)
Bottom ash 2.4–4.5% of Ca(OH) ₂ Particles size: 11 mm Moisture 15–27%	Rotating drum batch reactor automatic pressure regulation Filling ratio: 7–45%	Rotation speed: 1.2 rpm Pressure: 1 bar CO ₂ molar ratio: 15–100% Temperature: ambient Treatment time: 120 min	24	Brück et al. (2018b)
Recycled concrete aggregate 38.5% of CaO Particles size: >2 mm Grinded to 32 µm	Aqueous dynamic reactor Model 4560 from Parr No gas flow liquid/solid = 10 (w/w) gas/liquid = 2.4 (v/v)	Stirrer speed: n. c. Pressure: 10.7 bar CO ₂ molar ratio: 18.2% Temperature: ambient Treatment time: 10 min	~25 ^a	Ghacham et al. (2017)
Fine aggregates (recycled sand) Particles size: up to 6 mm	Rotating drum, 5 L CO ₂ cylinder and a mass flow controller	Rotating speed: 100 rpm Pressure: 1.4 bar Filling ratio: 10% CO ₂ molar ratio: 100%	47.5	Dos Reis et al. (2020)

Table 1 (continued)

Material used	Carbonation system	Experimental Conditions	CO ₂ uptake (mg/g)	Ref.
Water absorption of 7.9%		Temperature: ambient Treatment time: 7 h		

^a Sum of CO₂ dissolved in the liquid and converted in the solid.

incorporating carbonated recycled concrete aggregates (Xuan et al., 2016). This procedure enhances the idea of using waste cementitious/concrete materials for CO₂ sequestration purposes.

Direct mineral carbonation is an approach in which the alkaline metal oxides are made to react with CO₂ in a single step, which may be through the gas-solid route (Mun et al., 2017). This technology also has the advantage of being an exothermal process; due to this, energy consumption and the costs involved can be reduced compared to other technologies (Sanna et al., 2014). Carbonation already occurs naturally in buildings, concrete structures, etc., at atmospheric CO₂ (Baroghel-Bouny, 2004). However, under natural ambient conditions, carbonation of recycled aggregate is slow and limited by the access of CO₂, requiring storage periods of up to several months or even years (Thiery et al., 2013). Then, firstly proposed to study carbonation in laboratory (Daimon et al., 1971), accelerated carbonation has been further proposed as an efficient technique to improve the recycled aggregates' (Ku et al., 2014) eventually in more complex processes, as higher pressure (Cazacliu et al., 2016).

Therefore, over the last years, researchers dealing with accelerated carbonation have been focused on assessing and maximizing the capture of CO₂ by optimizing the operating conditions, which include handling the temperature, pressure, gas humidity, particle size, liquid-to-solid ratio, gas flow rate, etc. (Bertos, 2014).

Different accelerated carbonation process configurations were proposed in recent years, e.g., aqueous reactor configurations (Ghacham et al., 2017), or static reactor systems by exposing the material to CO₂ partial-pressures or passing CO₂ rich gas streams over aggregates/wastes in climate chamber systems (Lombardi et al., 2016) and immersion of RCA in acetic acid solution (Kazmi et al., 2019) and accelerated carbonation, etc.

Junyong et al. (2018) used the carbonation method to improve carbonated recycled concrete aggregate (CRCA) properties. The carbonation process reduced the water absorption ratio of RCA. The microstructural analysis demonstrates that CRAC has a much denser old ITZ than the uncarbonated RAC because of the chemical reaction between CO₂ and RCA's hydration products. It was concluded that the ITZ microstructure of RAC could be efficiently modified by carbonation treatment. Kazmi et al. (2020) used the carbonation method to improve the RA's physicochemical properties and found that the carbonation positively influenced the RCA mechanical properties. Zhan et al. (2019) used the carbonation process to improve the old attached mortar's RCA properties and durability. Also, the ion transportation of the cement mortars was evaluated. It was observed that the water absorption and sorptivity, total charge passed, and Cl ions diffusion coefficient and the bulk electrical conductivity of the cement mortar decreased after the mortar samples were subjected to the accelerated carbonation treatment. It was observed that the carbonation resulted in a decrease of the transport properties due to the decrease in porosity at the surface layer of the cement mortars.

Another carbonation method that has earned attention is the use of dynamic reactors, e.g., rotating drums (Lombardi et al., 2016) resulting in higher carbonation rates than static systems (Brück et al., 2018a). In the rotating drums, the sub-centimeter-sized bottom ash particles are mixed to improve the mass transfer between the gas and the solid, yielding comparably higher CO₂ uptake rates. Table 1 highlights several

Table 2
Mix-design of the parent concrete.

Constituent	proportion
crushed coarse siliceous aggregate 5.6/11.2 (Bréfauchet)	728 kg/m ³
alluvial sand 0/4 (Cheviré)	728 kg/m ³
cement CEM I 52.5 N (Saint-Pierre la Cour)	613 kg/m ³
Water	209 l/m ³
HRWR	9.2 l/m ³

published works that reported various carbonation system configurations applied to coarse waste particles with different calcium contents. The carbonation efficiencies in terms of CO₂ sequestration varied widely. Nevertheless, the rotating drum process applied to bottom ash seems more effective than static or fixed bed systems. It is also possible to infer that the rotating drum reactor can be considered a more cost-effective process than an aqueous carbonation process. Indeed, aqueous processes use vast amounts of water that need to be appropriately managed and treated at the industrial scale, and that might lead to a very costly process (Mun and Cho 2013).

This study focuses on coarse recycled concrete aggregates (RCA), i.e., centimeter-sized particles obtained from crushed concrete. These are recycled materials that could easily reenter the concrete production process after sieving. However, their use is nowadays limited because of their characteristics, which make the concrete production process challenging to control: their strength, which is relatively lower than natural aggregates, their high water absorption potential (Djerbi Tegguer 2012), but also for their propensity to attrition during mixing (Moreno et al., 2016). Besides the CO₂ sequestration objective, accelerated carbonation also seems a possible solution to strengthen the particles and reduce their water absorption. However, for such coarse materials, the overall carbonation kinetics is slow because of the predominance of internal diffusion over the reaction and the other mass transfer mechanisms. A possible path for accelerating the process is to couple the carbonation reaction with an attrition process, which reduces the surface layer depth of calcium carbonate formed by the reaction (Juilland et al., 2014). This attrition process can also be seen as beneficial for the mechanical properties of the resulting aggregates because it can be thought to quite selectively remove the mortar phase from the natural aggregates in the particles. This step means that both the selective removal of mortar and the carbonation reaction may strengthen

the coarse RCA and reduce water absorption.

The purpose of the present research is to investigate such a process coupling attrition and carbonation for CO₂ sequestration and to improve coarse RCA. In particular, this paper is focused on a laboratory scale rotating drum process. The evolution of the CO₂ sequestration efficiency with 4 main process parameters (initial moisture and size of the aggregate, drum rotation speed, and duration) is presented, followed by a characterization of the recycled aggregate after the carbonation by the rolling system (water absorption, density, particle size and paste content). The results are discussed in the light of a phenomenological model, combining attrition and carbonation. The CO₂ uptake potential and kinetics are then more attentively discussed.

The RA accelerated carbonation through rotating drums is still unexplored in the current literature. Table 1 shows the works found in the international literature being only one using RCA (dos Reis et al., 2020). This work's novelty lies in using an unexplored accelerated carbonation method, using rotating drums, to improve the RCA physicochemical properties. The method is based on both pressure drop in the gas phase induced by the CO₂-uptake of aggregates and attrition among the aggregates, which is also responsible for increasing CO₂ uptake. With this method, the mass-transfer phenomenon between CO₂ gas and aggregates interface inside the reactor is significantly improved, resulting in a high conversion within a short period.

2. Materials and methods

2.1. Materials

The recycled concrete aggregate was obtained by crushing concrete produced in the laboratory. It consisted of cubic samples of 1 dm³, which had been stored for 2 years at ambient conditions. The parent concrete's mix design, a high-strength – self-compacting concrete mixture with a high clinker content, are given in Table 2. It is important to note that the natural aggregates of the parent concrete were siliceous. This result means that CO₂ was meant to react only with the cement matrix. The cubic samples were crushed by using a jaw crusher; afterward, the aggregates were separated by different sizes, 5/8 mm, 8/10 mm, and 10/20 mm, by manual sieving.

After sieving, the concrete aggregates were heated in an oven at 70 °C for about two days to eliminate moisture. The objective was to reduce the amount of carbonation occurring naturally in the particles

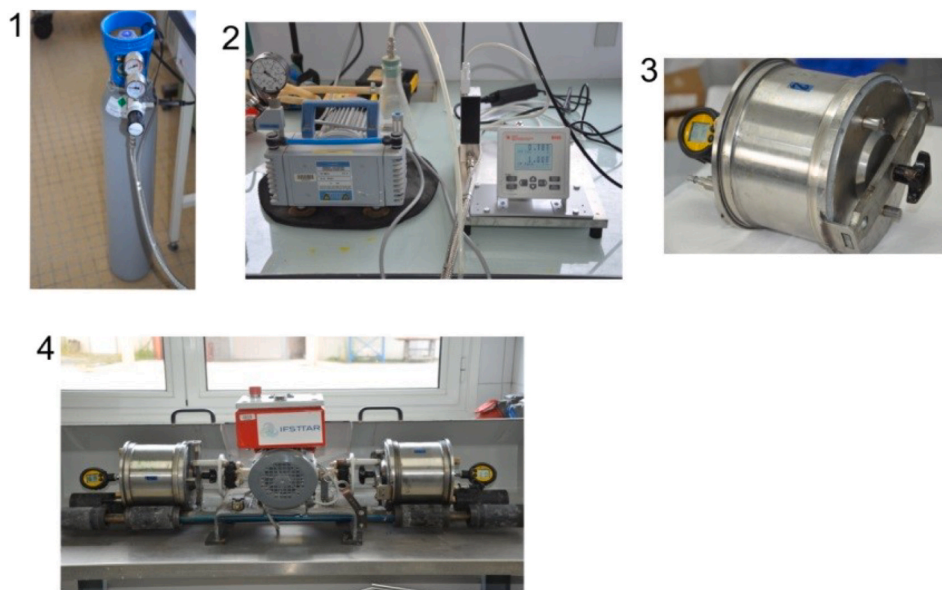


Fig. 1. Set-up of the laboratory-scale rolling carbonation experiments. (1) CO₂ gas cylinder; (2) vacuum pump and CO₂ flow automated controller; (3) hermetic vessel (with pressure sensor) for placing the concrete aggregates; (4) rolling machine.

and therefore standardize the materials' initial condition. A limit of 70 °C for the drying temperature was chosen to keep the stability of the ettringite.

2.2. Experimental set-up and procedure

As shown in Fig. 1, the system consisted of a CO₂ cylinder, a vacuum pump, a controlled injector of CO₂ with a maximum capacity of 2 L of CO₂ per minute (in normal conditions, i.e., p = 1 bar, T = 273.15 K) and horizontal drum vessels of 5.0 L of volume, totally air-proof. The metal covers were adequately sealed by using grease (petroleum jelly) to improve air-tightness. The jar was a classical Micro-Deval equipment jar (EN 1097-1), which internal dimensions are 200 mm in diameter and 154 mm in length. The angular velocity could be increased from 10 to 100 rpm.

Before the test, a mass of 500 g of concrete aggregates, filling about 5% of the vessel volume, was placed into the drum. Dry concrete aggregates were simply immersed in water for a given time to prepare samples with different initial moisture values. The water content was determined from the difference between the weighed mass after immersion (and eventually partial re-drying) and the dry initial mass. The concrete aggregates were placed in the heating chamber at 70 °C for some minutes until reaching the desired moisture to obtain moisture values below the water absorption capacity.

Before the beginning of each test, leak tests were performed using a vacuum of about 0.15 bar and monitoring the pressure for 5 min. The vessel was then filled with CO₂ up to the atmospheric pressure, 1.014 bar.

The rotation speed's effect on the carbonation process was studied, and different reaction times, from 1 to 7 days. It is important to stress that, during the tests, as the reaction progressed, the reactor's internal pressure decreased. The vessel's pressure was then manually restored to the atmospheric pressure by inserting quantities of CO₂ several times during the test, i.e., 2 or 3 times in each 24 h of the test. The pressure drop between successive CO₂ additions was recorded, and the mean CO₂ partial pressure during a test could be henceforth estimated.

At the end of the test, the vessel was opened. The concrete aggregates were stored in plastic bags (air-proof) for further analysis.

All experiments were performed at room temperature, around 22 °C.

2.3. Analysis and characterization techniques

2.3.1. CO₂ uptake

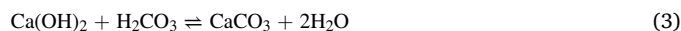
In the cement matrix, CO₂ reacts with portlandite (CH) and with calcium silicate hydrates (C-S-H). It is not easy to precisely estimate the CO₂ uptake, given that the stoichiometry for C-S-H is not clear. Besides, the reactions produce water, which contributes to the sample's mass increase, but whose fate in the drum (partial evaporation, sticking at the walls) makes it hard to quantify precisely its amount. Given these complications, as it is often done in the Literature, it was decided to simply analyze the CO₂ capture performance through the difference (increase) in the dried mass before ($m_{\text{ini dried}}$) and after ($m_{\text{final dried}}$) the tests.

$$\text{CO}_2 \text{ uptake} = \frac{m_{\text{final dried}} - m_{\text{ini dried}}}{m_{\text{ini dried}}} \quad (1)$$

The recycled concrete aggregates were dried in a heat chamber, and the samples were weighed for nearly two days to do it, before and after carbonation. The concrete aggregates were considered dry at "constant mass," namely when the mass changed less than 0.05% between two successive weighings, spaced 1 h apart. The drying temperature was fixed at 70 °C to avoid the ettringite dehydration between 75 and 120 °C (Zhou Q, Glasser 2001) as recommended by Quattrone et al. (2016).

A more detailed survey of the carbonation schemes could clarify the relation between dry mass increase and real CO₂ uptake. Carbonation of hydrated Portland cement converts Portlandite (Eq. (3)) and calcium

silicate hydrate (Eq. (4)) into carbonate:



By considering that the cement paste contains only portlandite (CH) and that, therefore, for each mole of CO₂ captured, a mole of H₂O is liberated, the stoichiometry implies that the mass of CO₂ captured is 1.69 times the mass increase of the sample. However, the most important hydration product is by far the CaO-SiO₂-H₂O-like gel phase (C-S-H) and not the CH. The exact stoichiometric coefficients in Eq. (4) are not clearly defined. However, Morandau et al. (2014) demonstrated that for CEM I cement paste, C-S-H carbonation consists only in decalcification, and the reaction mechanisms are likely not produced evaporable water ($z \approx yt$). This result suggests that the mass of CO₂ captured by C-S-H carbonation is roughly equal to the sample's corresponding mass increase. For a classical proportion of 0 to 25% of CH in the hydrated Portland cement, one can conclude that the mass of CO₂ uptake during the process was 1 to 1.17 times the dry mass increase of the samples before and after carbonation. This range was validated by analyzing the pressure loss during the reaction, which was conducted to mass CO₂ uptake estimated to about 1.05 times the dry sample mass increase.

Consequently, Eq. (1) implies that the amount of captured CO₂ is slightly underestimated in this paper.

2.4. Characterization of recycled concrete aggregates

Several techniques were employed to characterize the changes in the aggregates' properties occurring due to the rotating drum process. We detail them here for the properties analyzed:

- **Particle size and shape.** The particle size distribution evolution was determined by sieving, while a VDG40 video grader determined the evolution of the particle shape according to the standard French XP P 18-566-2002 (AFNOR 2002). The shape was characterized by the angularity parameter, which is related to the mean value of the most sharpened outgoing edges between two adjacent straight lines (Descantes et al., 2006a).
- **Chemical composition.** The progress of the carbonation front was determined by spraying a 1% phenolphthalein solution on the surface in the broken carbonated and non-carbonated concrete aggregates, according to Rilem criteria (Rilem 1994). Phenolphthalein was prepared as a 1% solution in 70% ethyl alcohol and 30% of deionized water. The color changes from colorless to purple-red in the pH range from 8.3 to 10. The aggregates' alkaline nature was observed due to the change in the aggregates' color provoked by the phenolphthalein indicator. Therefore the progress of carbonation can be qualitatively estimated by the depth of the zone, which does not change its color after the treatment. EDX analysis was performed on non-carbonated and carbonated samples as detected by the color changes to reinforce the conclusions made on phenolphthalein tests' bases. The EDX analysis was obtained through a scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (TESCAN 3, Sweden). The coarse grains were ground at the maximum particle size of 500 μm. For each material, the test was performed on two samples, and the mean of these two measures was considered in comparing the non-carbonated and carbonated sample's elemental composition.
- **Physical properties.** Particle density and water absorption potential (which characterizes the maximum amount of water that can be stored in the particle's internal accessible pores, expressed by mass ratio) change due to carbonation gave that carbonates occupy more volume than the original components. Before and after rolling

Table 3
Experimental plan description for the rolling carbonation tests.

Test Duration (days)	Particle Size (mm)	Initial Moisture (%)	Rotation Speed (rpm)	CO ₂ partial pressure mean each day (bar)
Effect of rolling speed				
3 ^a	10/20	8.2	100	0.70 / 0.68 / 0.73
3	10/20	7.6	60	0.74 / 0.76 / 0.71
3	10/20	7.7	20	0.73 / 0.79 / 0.77
3	10/20	7.5	0	0.78 / 0.78 / 0.78
Effect of moisture				
3	10/20	4.1	100	0.75 / 0.78 / 0.82
3	10/20	5.9	100	0.72 / 0.75 / 0.83
3	10/20	7.1	100	not measured
3	10/20	10.0	100	0.72 / 0.73 / 0.75
Effect of aggregates size				
3	8/10	7.9	100	0.65 / 0.69 / 0.75
3	5/8	8.1	100	0.67 / 0.73 / 0.78
Effect of carbonation duration				
4 ^b	10/20	7.6	100	0.65 / 0.64 / 0.70 / 0.71
7 ^b	10/20	7.4	100	0.54 / 0.64 / 0.72 / 0.74 / 0.78 / 0.79 / 0.80
Static mode				
4**	10/20	7.3	0	0.81 / 0.81 / 0.82 / 0.82

^a This test was repeated 3 times.

^b For these tests, the aggregates were characterized after each day of the test than inserted again in the drum.

carbonation, the concrete aggregates' density and water absorption were determined according to NF EN 1097-6.

- **Amount of mortar.** The amount of hydrated cement in the total mass of the coarse aggregates is essential because it is related to the potential of sequestration of the RCA. Also, its evolution during the process may give insights into the selective nature of attrition. Therefore, the paste content of the recycled concrete aggregates was estimated as proposed by Nagataki et al. (2004). More recently discussed in the literature (Yang et al., 2018), this method was adapted to the concrete aggregates' characterization in the present study. Some representative samples were dried at 70 °C for three days until constant weight. Then, 10 g of the dried samples were immersed in 100 ml of a 0.1 M HCl solution for 24 h. Afterward, the solid fractions were filtered on a glass filter (Pyrex No. 4, pores: 10–16 μm) and washed four times using tap water. The solid fractions were then dried in the oven at 70 °C until constant weight loss (see Section 2.3.1 for details) to calculate the solid fraction dissolved by the hydrochloride acid (CPC) of each sample:

$$CPC = \frac{m_1 - m_2}{m_1} \quad (5)$$

where m_1 is the mass of dried material before dissolution and m_2 is the mass of dried filtrate.

The measures were done in duplicate and, their respective deviations are presented below. It should be noted that the coarse aggregate of the original concrete did not contain calcareous aggregates, but the alluvial sand contained a proportion of calcareous elements. CPC is then an overestimation of the cement paste content.

2.5. Experimental plan

The experimental program is summarized in Table 3. The main process parameters which were investigated in this study were: (1) initial moisture of the aggregates, (2) rotating speed, (3) test duration, and (4) initial particle size.

The base case corresponds to initial particle size in the range 10/20 mm, a rotational speed of 100 rpm, initial moisture of the grains of 8.2%, and three days of rolling carbonation. This test was repeated three times. Five initial moisture levels, three particle sizes, and four rotational speeds were investigated by varying the central parameter set

Table 4
Experimental plan description for the attrition tests.

Test Duration (days)	Particle Size (mm)	Initial Moisture (%)	Rotation Speed (rpm)	Mass of aggregate (kg)
Effect of Moisture				
3	10/20	0	100	0.5
3	10/20	3.0	100	0.5
3	10/20	7.6	100	0.5
Effect of rolling speed				
3	10/20	7.5	20	0.5
3	10/20	7.7	60	0.5
3	10/20	7.8	100	0.5
Effect of the filling ratio				
3	10/20	7.4	100	0.5
3	10/20	7.9	100	1.0
3	10/20	8.1	100	2.0

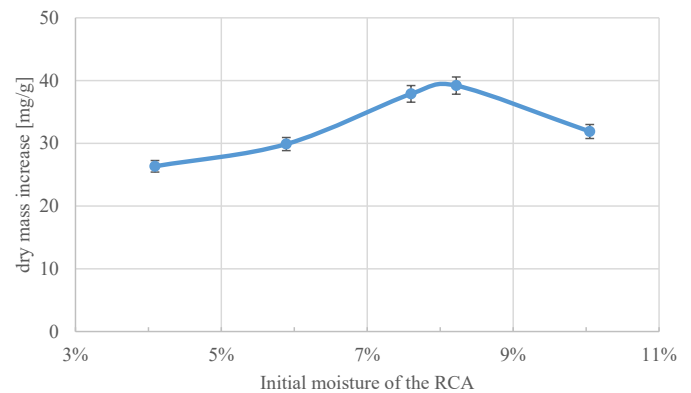


Fig. 2. Effect of the moisture on the CO₂ sequestration (100 rpm, 3 days, and initial size of the particles 10/20 mm). Bar error corresponding to the mean partial CO₂ pressure incertitude.

independently. The same central parameters were tested in a seven-day test. Finally, one particular experiment tested this set of parameters by analyzing the aggregate after each day of the test for four days. In this particular test, the same concrete aggregate was first pre-wetted to the initial reference moisture and afterward introduced in the jar for a new day of the test.

Table 3 also gives the calculated mean partial pressure of CO₂ in the jar for each test configuration during each day of the test.

The normalized standard deviation of the mass increases after carbonation was determined based on the repeated tests. This level of normalized standard deviation was confirmed by analyzing the pressure lost in the drum after carbonation at different carbonation durations.

As a parallel evaluation, the particle size distribution evolution due to the attrition induced by bulk shear and collisions was tested independently from the carbonation process. Table 4 presents the rotating drum tests made to characterize attrition, which was made without the CO₂; besides the rolling speed, the initial moisture of the aggregates, and the initial particle size, in these tests also the effect of the filling level of the drum was characterized.

3. Results

3.1. CO₂ sequestration

The CO₂ sequestration performance of the process is evaluated for the process parameters detailed above. As it was already discussed, the CO₂ uptake is characterized by the dry mass increase of the aggregates.

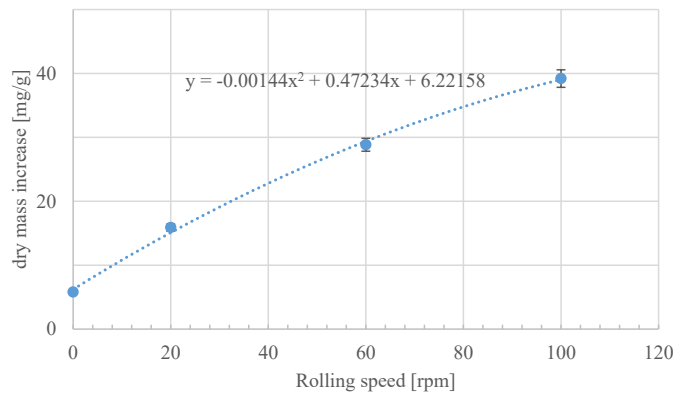


Fig. 3. Effect of the rolling speed on the CO₂ sequestration at 3 days of duration (moisture ranging from 7.5 to 8.2%). Bar error corresponding to the mean partial CO₂ pressure incertitude.

3.2. Effect of the initial moisture of the aggregates

It is well recognized that the concrete aggregates' moisture content plays a critical role in the CO₂ sequestration (Verbeck 1958; Drouet et al., 2015). Sufficient water must be present for carbonation to occur, but not as much as to fill pores and slow down the access of CO₂ that occurs by diffusion (Brück 2018b; Chang 2012). Tests of CO₂ sequestration were made at different initial moisture of the concrete aggregates to identify the moisture optimum in the proposed process, as shown in Fig. 2.

At moisture of 4.0%, the CO₂ sequestration was the lowest; as the moisture in the aggregates was increased from 4.0 to 7.6%, the CO₂ sequestration increased by 40%, while around 8% the CO₂ sequestration reached a maximum (around 39.5 mg/g). When the moisture exceeded the value of 8.2%, the CO₂ uptake decreased.

These findings are consistent with the results found by Li et al. (2018), and the excess of intergranular water likely limited the diffusion of CO₂ (Brück et al., 2018b). However, the optimum moisture value, between 7 and 8%, looks high compared to the water absorption of the present concrete aggregates (which is around 5.5%). Indeed, one could expect optimum moisture in the humidity range corresponding to unsaturated concrete aggregates. The hypothesis here is that a part of the water is present in the form of capillary bridges, or it is squeezed out of the aggregates by centrifugation during the drum rolling. Indeed, when the drum was opened after the carbonation, a significant amount of droplets of free water were observed on the internal drum walls. So, the water amount within the grains could be fairly lower than the overall moisture, explaining why the overall moisture's optimal value is larger than the water absorption value.

The drops of free water inside the drum could also suggest a saturation of the vapor pressure inside the drum. As the drum's total pressure is not constant during the test, the vapor pressure level could also vary. For this reason, we did not consider the measure of total pressure during the test to estimate the amount of CO₂ consumed during the carbonation.

3.3. Rotating speed of the drum

Fig. 3 displays how the rotation speed influenced the overall carbonation kinetics. The difference in the sequestered CO₂ amounts was remarkable; for an initial mass of 0.5 kg, with about 7.5% initial moisture, the static mode sequestered only 6 mg/g in 3 days; a rolling speed of 20 rpm sequestered 16 mg/g; 60 rpm sequestered 29 mg/g, and at 100 rpm the sequestration was about 39.5 mg/g, more than 6.5 times compared with the static-mode.

These results, therefore, suggest that for the present geometry and filling ratio, the carbonation rate was (and probably could still be

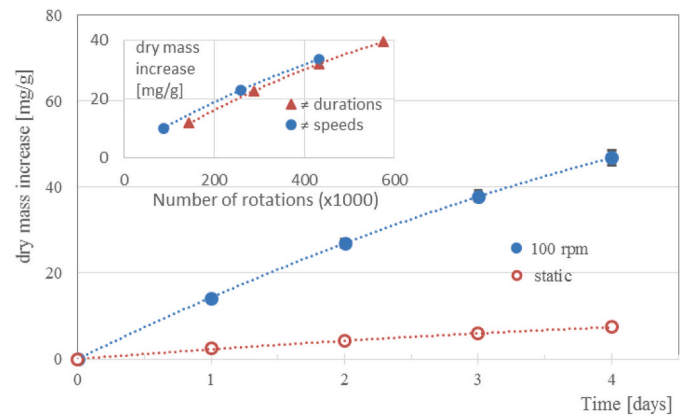


Fig. 4. Kinetic of CO₂ sequestration under static mode and at 100 rpm (moisture ranging from 7.3 to 7.6%); in the inset: Kinetics of CO₂ sequestration with the number of drum revolutions; only the contribution of the dynamics is considered (the difference between the carbonation in dynamic and static conditions, for the same process duration); the number of drum revolutions represented on the figure was calculated for the tests with different rotation speeds ("≠ speeds") and the tests with different process durations ("≠ durations").

further) significantly improved by increasing the rotation speed.

Given the coarse size of the aggregates, it is expected that the controlling mechanism is the diffusion within particles; therefore, the intensification of the process is not imputable to the mixing of the grains or the gas phase induced by the rotating drum geometry. The most likely reason for this performance improvement lies probably within the fact that the motion of the solid provided attrition of the aggregates, liberating carbonated zones and increasing the specific surface exposed to carbonation. This hypothesis will be reviewed in the Discussion section.

3.4. Rolling carbonation duration

The time evolution of the CO₂ uptake kinetics, during four days, in static mode and at 100 rpm was evaluated, and the results are shown in Fig. 4. The figure confirms the trend shown in Fig. 3, i.e., that rotation significantly improved the carbonation kinetics compared with the static-mode. The improvement ratio seemed almost independent of time (about 6) at 1, 2, 3, and 4 days of carbonation, which suggests that the mixing and attrition phenomena in the rotating drum were mostly linear in time for the test duration range. From Fig. 4, it is also possible to infer that the kinetic did not reach an equilibrium.

It is interesting to plot the dry mass increase as a function of the number of drum revolutions to provide additional elements for understanding the role of attrition in the process. This result allows comparing the tests made for different test duration but the same speed with tests made for different speeds but a fixed duration. Therefore, the inset of Fig. 4 shows the evolution of the dry mass increase of the RCA with the number of drum revolutions. The number of drum revolutions represented in the figure was calculated for the tests with different rotation speeds and the tests with different process durations. The static contribution for the same duration is subtracted (given that it does not depend on the speed but only depends on the test's duration) to compare the drum rotation effect. It is evident from the figure that the number of revolutions is the main parameter controlling the intensification of the drum rotation's carbonation. The number of revolutions can be seen as a macroscopic measure of the cumulative mechanical energy injected in the process. This energy is partially used for particle abrasion. More energy introduced into the system (higher rotating speed or most extended duration) implies that more abrasion occurs, removing – as we will underline in the Discussion section – previously carbonated layers, thereby accelerating the process. A secondary effect seems to be present, however, given that, for the same number of revolutions, the longer

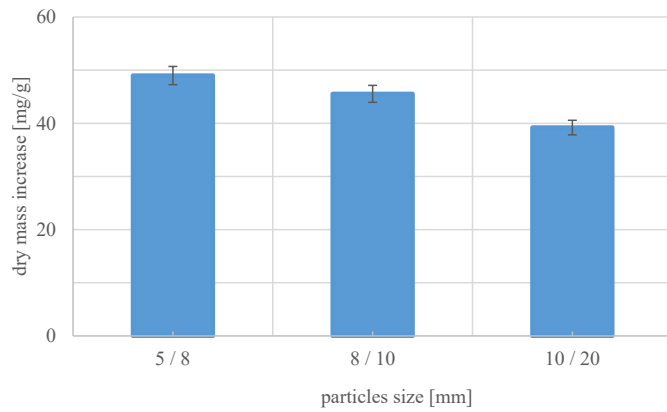


Fig. 5. Effect of the particle size on the CO₂ sequestration, for moisture ranging from 7.9 to 8.2%, and 3 days of the rolling process (100 rpm). Bar error corresponding to the mean partial CO₂ pressure incertitude.

carbonation process (i.e., lower rotational speed) induces a slightly higher overall rate (about 10 to 20%). So, the carbonation time seems to be the second influent parameter of the dynamic process, independently of the number of revolutions.

3.5. Initial particle size

A series of tests were devoted to evaluating the initial particle size distribution effect on the CO₂ sequestration. Different size ranges (5/8, 8/10, and 10/20 mm) of the aggregates were separately subjected to the rotating drum process. The results, displayed in Fig. 5, showed that particle size played an essential role in determining the degree of CO₂ uptake by the aggregates. Smaller aggregates sequestered a higher amount of CO₂. Aggregates sized between 5 and 8 mm sequestered 49 mg/g; between 8 and 10 mm sequestered 46 mg/g and between 10 and 20 mm sequestered 40 mg/g. It seems clear that the carbonation rate is faster in small particles due to the higher surface area available to get in contact with the CO₂ (Thiery et al., 2013), possibly also because of slightly higher attached mortar content. The inter-particle porosity, so the accessibility of the CO₂ gas to the surfaces, is similar for these three aggregate fractions. Evidently, reducing particle size accelerates the reaction, eventually changing the limiting mechanisms: it is probable that, for finer materials such as sand, the kinetics will be way faster and controlled by the mass transfer outside the grains (dos Reis et al., 2020). For CO₂ storage only, fine particles are therefore preferred. However, in our case, the focus is on CO₂ sequestration combined with the production of coarse aggregates for recycling into concrete, so it is also interesting to study the process's performance for coarse grains.

3.6. Characterization of the evolution of the RCA

3.6.1. Chemical changes

The elemental composition determined by energy-dispersive X-ray spectrometry (EDX) analysis highlighted the presence of calcium, oxygen, silicon, and carbon (Table 5) in both non-carbonated and carbonated mixtures. The EDX results showed a significant increase in the carbon peak in the carbonated concrete aggregates sample compared to the non-carbonated sample (16.0% and 7.5% in mean, respectively). The results confirm that the carbonation took place in the mixtures, evidenced by the phenolphthalein test.

3.7. Water absorption and density

Chemical analyses such as EDX are valuable tools for analyzing the aggregates' chemical composition changes in-depth. However, their use may be complex in the case of coarse RCA because of the strong heterogeneity of the material that needs to be powdered, statistically homogenized, etc. The need for finely crushing the samples also makes it impossible to evaluate microstructure effects. Therefore, classical techniques such as water absorption or densitometry are essential tools that

Table 5

Content in main chemical elements in non-carbonated and carbonated samples (two different samples for each configuration).

Test	Carbon	O	Ca	Si
Virgin aggregates	7.9	35.3	21.1	17.7
100 rpm, 8%,	7.1	41.1	25.3	21.9
10/20 mm, 3 days	17.1	33.1	20.3	17.5
	15.2	37.1	23.9	18.2

Table 6

-The influence of the rolling carbonation process on the water absorption and density values.

Size (mm)	Duration (days)	Speed (rpm)	CO ₂ uptake (mg/g)	Water absorption (%)	Enveloped Density (kg/m ³)
				Standard deviation: 0.07	Standard deviation: 17
10/20	0	0	0	5.0	2 229
	4	0	7.5	4.6	2 227
	3	20	15.9	4.3	2 282
	3	60	28.9	4.1	2 305
	3	100	37.9	4.1	2 300
	3	100	39.2	4.2	2 323
	4	100	45.8	4.2	2 299
	7	100	79.5	4.0	2 285
8/10	0	0	0	5.4	2 231
	3	100	0	5.55	2 205
	3	100	45.5	4.2	2 271

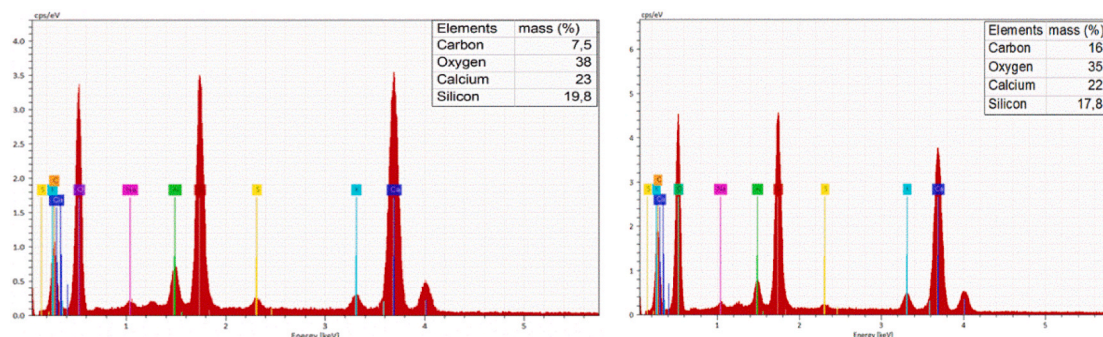


Fig. 6. EDX spectra of non-carbonated (left) and carbonated (right) samples; the values in Fig. 6 are the averages of two measures.

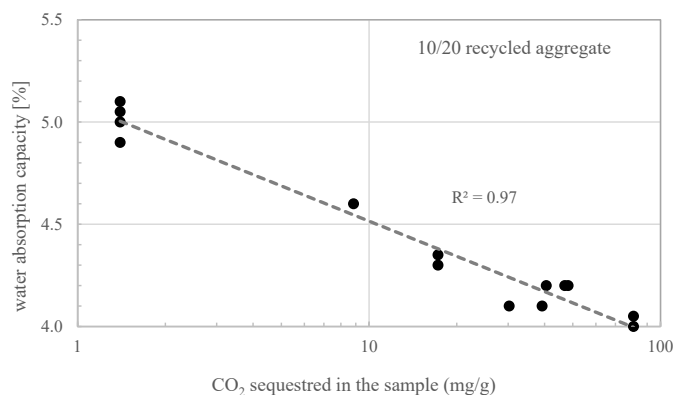


Fig. 7. Evolution of the water absorption capacity with the carbonation of the concrete aggregates (it was considered that the concrete aggregates were already slightly carbonated before the rolling carbonation, 1%).

can provide indirect but robust estimates of the material's physico-chemical evolution, providing hints on the microstructure.

The results of water absorption capacity and density of several recycled concrete aggregate samples before and after carbonation are collected in Table 6. As commonly known, given that both parameters are related to the paste composition and content, the absorption capacity and density are well correlated.

The results show that the water absorption values of carbonated samples were lower than those of non-carbonated concrete aggregates. Similar behavior was found in previous works (Johannesson and Utgenannt 2001), which showed that carbonated samples could have lower absorption capacity and higher density than un-carbonated samples. This result might be due to the carbonation products filling the pores as the carbonation of calcium hydroxide progresses (Thiery et al., 2011).

In another work, dos Reis et al. (2020) tested the rotating drum as a method to accelerate the carbonation process of recycled fine aggregates and found that this method increased the CO₂ uptake almost 5 times higher than the static carbonation method. The method reduced the water absorption capacity up to 25% and when compared to the non-carbonated samples and increased the aggregate densities. They also reported that the water absorption reduction was due to the carbonation products filling the pores as the carbonation of calcium hydroxide, which is one of the main reactions (Peng et al., 2020). These results follow the international literature (Lu et al., 2019). Xiao et al. (2012) reported that the water absorption of recycled aggregates could reflect the content of the attached mortar and porosity of recycled aggregates concrete; therefore, improving the characteristics of the aggregate by carbonation is a suitable technique to make recycled aggregates reusable.

By the results in Table 6, it is also possible to infer that the degree of carbonation (in terms of CO₂ uptake values) can be linked with the water absorption values. The sample with the highest CO₂ uptake value also obtained the lowest water absorption value. The carbonation level seemed to explain the evolution of the concrete aggregates' water absorption capacity (Fig. 7).

As will be discussed in Section 3.2.4, the amount of mortar in the coarse recycled concrete aggregate may change after attrition. Therefore, in principle, this can influence (even without the carbonation) the properties of the material, increasing density and decreasing the RCA's water absorption capacity. However, as the comparison made for the 8/10 aggregate in Table 6 suggests, the variation of such properties due to attrition only is within the experimental error. Conversely, the same attrition level associated with carbonation significantly decreased the water absorption of the concrete aggregate (Table 6) to a similar level as for the coarser aggregate.

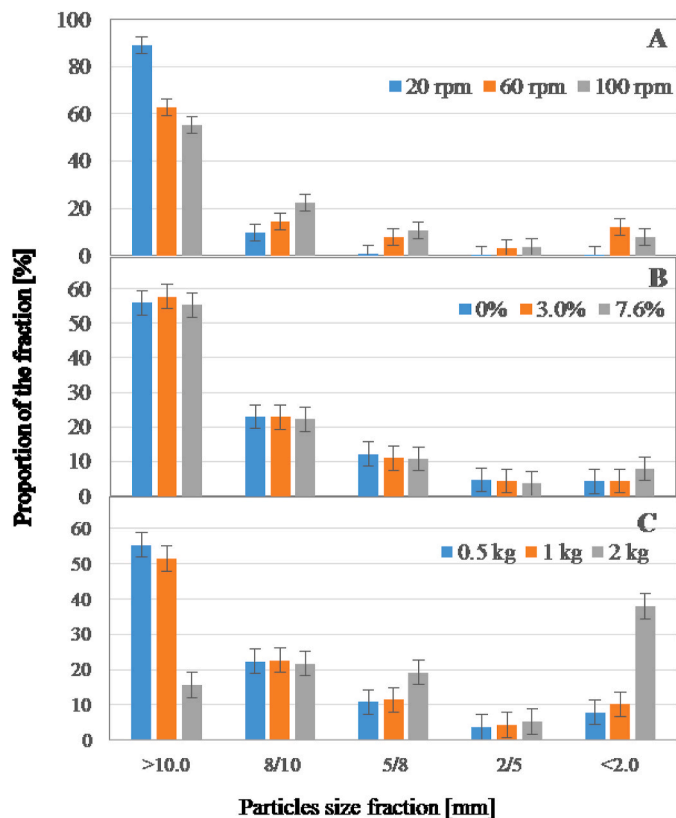


Fig. 8. Effect in the production of the fines of (A) the rolling speed; (B) the initial moisture; (C) the quantity of the aggregates in the drum; the standard deviation was obtained from 3 repetitions of one test configuration.

3.8. Particle size distribution

Granular materials subject to low energy collisions and bulk shear typically display surface chipping or wear, and this is true also for RCA, mainly due to their heterogeneous nature characterized by a less resistant mortar phase (Grande et al., 2005). Therefore it is expected that bulk shear induced by the rotating drum motion produces attrition. Given the nature of the grains, two-particle breakage mechanisms may play a role: cleavage fracture due to the propagation of cracks in the particle or surface chipping or abrasion. The first mechanism may produce a wide distribution of fragments, but also few coarse fragments in the case of heterogeneous materials such as concrete. The second mechanism nearly does not affect the parent particle's size but generates a bimodal particle size distribution by populating the fine fractions.

As it was already discussed, particle breakage may be a possible factor for intensifying the process. Thus, it is interesting to characterize the evolution of the particle size distribution during the process. In this work, the attrition progress was analyzed independently from the carbonation process (so at atmospheric pressure exclusively constituted by air) in a series of dedicated tests. Fig. 8 shows the samples' grading after mixing at different rotating speeds, different initial moistures of the aggregates, and different filling ratios of the drum. All tests were performed with the same duration (3 days) and with the same initial size of the aggregate (10/20 mm).

Three tests were repeated for the nominal rotational speed (100 rpm) and 0.5 kg of initially moisten aggregates at 7.6%. The standard deviation of the size fraction content was determined to about 3.5% (as a mean variation of all measured size fractions).

The tests performed at the lowest rotation speed, 20 rpm, produced nearly negligible attrition (see Fig. 8A). After the test, only about 1% of the material had a size inferior to 8 mm. The aggregate size decreased mostly by the breakage of some of the coarsest particles in large pieces.

Table 7

Solid fraction dissolved by the hydrochloride acid (CPC) the normalized standard deviation of the measures was 1.8%.

Carbonation duration (days)	Rotation Speed (rpm)	Size (mm)	CPC (%)
Virgin aggregates	–	10/20	42.2
3	20	10/20	42.1
3	100	10/20	38.5
3	100	0/0.5	93.3
7	100	10/20	38.0

Higher rotational speeds produced both breakages of coarse particles and abrasion of particle surfaces into fines. At high velocities, the abrasion is significant (about 10% of material in the fraction 0/2.5 mm). Both mechanisms seem to be significantly increased by the rotational speed. At 100 rpm, the general particle aggregates size decrease. From initial particle size >20 mm, after speed rotation of 100 rpm, it presented the lowest fraction of aggregates with a particle size above 10 mm and therefore larger fraction of particles smaller than 10 mm looking at the raging from 2 to 10 mm, at 100 rpm presented the largest fractions. At higher speeds, the attrition among the particles is maximized, provoking its breakage. However, for the particle sizes below 2 mm, the rotation speed at 60 rpm presented a higher fraction than 100 rpm. This could be explained by the fact that at 100 rpm, the aggregates could create more efficient compaction and, therefore, high resistance to shear (Dubé et al., 2013). Also, the aggregates can form a pack that moves inside in the same regime, a plug-flow, reducing the contact between particles but instead.

Fig. 8B seems to point out that it is negligible for both attrition and coarse breakage regarding the RCA's initial moisture influence.

Finally, Fig. 8C shows that the quantity of concrete aggregates inside the rotating drum did influence the production of the fines. The higher the filling ratio, the higher was the attrition. As much aggregates are inside the reactor, more fines are produced (in percentage); this result can be linked by the fact that with higher amounts of aggregates inside the reactor, (i) wall slip can be reduced and (ii) stresses inside the granular material are higher, therefore increasing the potential for

attrition.

Solid fraction dissolved by the hydrochloride acid.

The evolution of the carbonation process obviously depends on the amount of mortar present in the recycled aggregate (and on its composition). In particular, the amount and quality of the paste determine the potential for CO₂ capture. Also, the grading of the aggregate continuously evolves during the present process due to breakage, as well as the composition of the coarse grains (Artoni et al., 2017). Analyzing the evolution of the amount of mortar in the grains allows understanding the underlying mechanisms of fracture, contributing to the overall process kinetics' intensification. For this reason, we determined, with the technique described in Section 2.3.2, the solid fraction dissolved by the hydrochloride acid for different size fractions for chosen material samples (both virgin aggregates and RCA treated in the rotating drum) detailed in Table 7.

In this work, we determined the solid fraction dissolved by the hydrochloride acid, which overestimated the cement paste content in the samples. For a process with a rotation speed of 20 rpm, the solid fraction dissolved by the hydrochloride acid has not changed after three days of rolling carbonation compared with the non-carbonated aggregate. This observation agrees with the attrition results for the same rotational speed and duration, which showed (Fig. 8A) that almost all material is still in the fraction larger than 8 mm. The attrition tests discussed in the previous section showed that a significant part of the material is in fine fractions for higher rotational speed. For these experimental conditions, the solid fraction dissolved by the hydrochloride acid decreased in the coarse fraction of RCA, while the fine fraction was composed of more than 90% of mortar. These data confirm that RCA attrition is very selective for the mortar phase because of the mortar phase's lower strength.

4. Discussion

4.1. Interaction between attrition and carbonation

From the results outlined in the previous sections, it is evident that

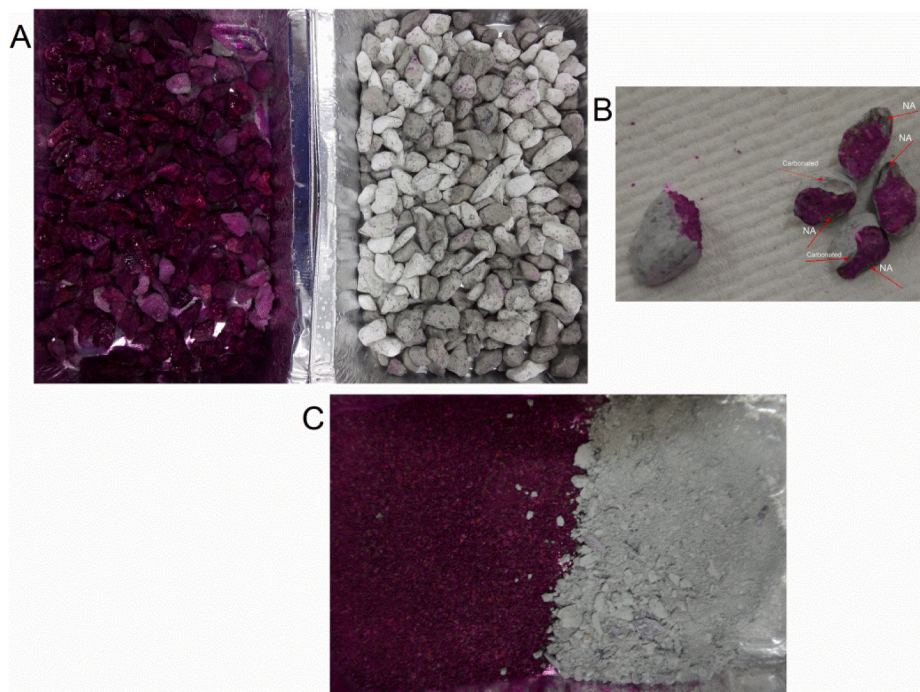


Fig. 9. (A) Phenolphthalein test before (left) and after (right) rolling carbonation (72 h); (B) view inside some grains which were broken for visualizing the interior the grains; (C) Phenolphthalein test of particles less than 2 mm, obtained by shearing 72 h in the drum without carbonation (left) and with carbonation (right); the particles were ground before the phenolphthalein test.

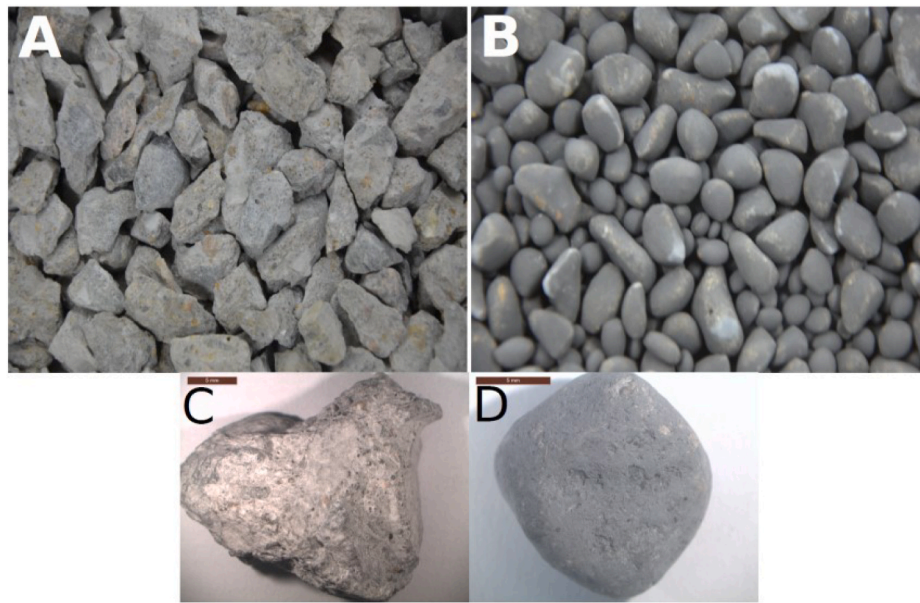


Fig. 10. Appearance of the aggregates before (A, C) and after (B, D) rolling carbonation.

many parameters influence the CO₂ sequestration by the RCA in the rotating drum process. Simple physicochemical reasons explained the effect of the initial size and the moisture of the aggregates. On the other hand, the effect of the rotating speed and the test duration allows concluding that the primary phenomenon that enhances the CO₂ sequestration in the rotating drum process is the grains' attrition. Grading analyses and dissolution by acid proved that attrition is characterized by the predominance of surface abrasion mechanisms, which were also shown to remove the RCA's mortar phase selectively. Now, as we mentioned above, the overall rate of the process is faster for smaller particles. So there are two possibilities, depending on the relative rate of attrition and of carbonation: (1) abrasion produces fine grains which are subsequently quickly carbonated, or (2) coarse RCA surface layers are first carbonated. Then abrasion phenomena remove part of these already carbonated layers (and the carbonated layers' reduction speeds up the reaction by reducing the internal diffusion resistance). It is essential to understand this point because a carbonated surface layer may give the RCA better mechanical properties. As the water absorption, density, and chemical analyses suggest, at least the coarse grains' surface is carbonated, so the second mechanism discussed above seems to prevail. A systematic inspection of the carbonated samples using the phenolphthalein test was made to confirm this. A typical result is presented in Fig. 9. As can be seen in Fig. 9A, it appears that all the surface of the coarse concrete aggregate was carbonated (the carbonated zone was colorless, and the non-carbonated zone had a pink coloration). However, Fig. 9B shows that the carbonation was mostly limited to the concrete aggregates' surfaces.

Fig. 9C shows particles under 2 mm obtained by the shearing of coarse aggregate in the rolling drum for three days, without CO₂ (left) and with CO₂ (right). Before analyzed by the phenolphthalein test, these particles were ground. No pink-colored zones were observed on the particles obtained by rolling carbonation. This result suggests that the carbonation was mostly completed for this size of concrete aggregates. This observation agrees with the high cement paste content of the fine fractions and their very high specific surface compared with the coarse concrete aggregate.

These results suggest that the coarse concrete aggregate's carbonation was slightly faster in our rotating drum process than their attrition. One can deduce that the coarse aggregate's carbonation depth increased during the process, contributing to progressively decreasing carbonation rate in the processing time.

Quality of the coarse RCA after the rolling carbonation.

From the previous discussion, it is evident that the attrition of the grains provided by the solid's motion in the drum acts as an enhancer for CO₂ sequestration. On the other hand, as we already mentioned above, the process can be potentially attractive because it can improve the quality of the coarse RCA by removing part of the mortar phase, which is responsible for low particle strength and high water absorption, but also by hardening the surface of the grains by carbonation.

On the other hand, attrition can reduce the particles' angularity, and it is, therefore, essential to quantify the change in the particles' angularity due to the rotating drum process. For this reason, the angularity evolution was evaluated for the base case configuration (3 days; particle size 10/20 mm; moisture of 7.6%; 100 rpm). Fig. 10 presents pictures of the aggregates before and after the carbonation process. Before the rotating drum process, the aggregates showed irregular shapes and rough surfaces (see Fig. 10A and C). However, after the process, the same aggregates presented more spherical shapes with smooth surface textures (see Fig. 10B and D). These qualitative results are reproduced by the analysis of the same samples made with the VDG video grader. The corresponding angularity values are 0.226 and 0.186 (dimensionless) for non-carbonated and carbonated samples, respectively. This considerable variation of the angularity index corresponds to the typical difference between crushed and rounded aggregates (Descantes et al., 2006b).

Therefore, the coarse aggregates produced during the process had better surface properties, mainly significantly lower water absorption capacity, but were more rounded than the original ones. The following property may restrict the possible final use of the aggregates in producing new concrete. Indeed, if rounder coarse aggregate did not necessarily increase the segregation sensitivity of fresh concrete (Esmailkhanian et al., 2014), it could be to the origin of the new concrete's lower mechanical strength (Rocco and Elices 2009) as angular aggregate shape inhibit debonding (Saouma et al., 1991) increasing the fracture energy (Guinea et al., 2002). Conversely, aggregates having more angularity demand more water to produce workable concrete than the rounded aggregates with smooth surface texture (Murdock 1960) as they induce better compactibility of the aggregate (Shergold 1953) and better workability (Kaplan, 1958). It can be concluded that the use of rolling carbonated coarse recycled aggregate in new concrete requires more careful mix-design, but could facilitate the manufacturing of the new concrete, mainly given their lower water absorption capacity.

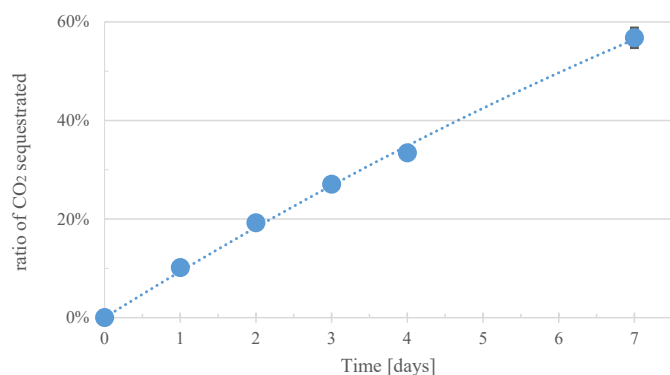


Fig. 11. Kinetic of CO₂ sequestration as a ratio of the potential of CO₂ capture; the process parameters were 100 rpm, moisture ranging from 7.4 to 7.6%, the drum of 5 L filled with 500 g of 10/20 mm concrete aggregates.

4.2. Potential for CO₂ uptake

The concrete aggregate's paste content gives the possibility to estimate the potential of CO₂ capture during a complete carbonation process. By considering that for this kind of cement, the production of 1000 kg liberates about 470 kg of CO₂ due to the decarbonation, the potential to carbonate 1 kg of the tested recycled concrete aggregate is about 140 g (so 140 mg/g of recycled concrete aggregate). This value is over-estimated. Indeed, the acid attack's solid fraction dissolved is, as explained in section 2.3.2, higher than the cement paste content in the recycled aggregate.

So, in the present optimal process conditions (rotation speed of 100 rpm, around 8% of initial moisture), one can assume that at least 8 to 10% of the available paste was carbonated in each day of rolling carbonation. After seven days, at least 60% of the potential of CO₂ capture was effective (Fig. 11).

5. Conclusions

This paper presents a rolling carbonation process as a solution to accelerate the carbonation of coarse recycled concrete aggregate. Performing carbonation in a rotating drum promotes selective attrition of concrete aggregates. The process accelerates the carbonation by removing the carbonated zones on the surface of coarse aggregates and then facilitating the access of CO₂ to un-carbonated zones. In this laboratory study, the optimal initial carbonation condition was found to be at 100 rpm, aggregate moisture around 8%, and 500 g of aggregates in a 5 L reactor. The tests were performed at standard atmospheric conditions, with a partial pressure of CO₂ kept at about 0.75 bar. The main tests were conducted on 10/20 mm aggregate, but it was shown that the carbonation process was 20% faster for smaller size aggregate (5/8 mm).

The dynamic process highly increased (about 6 times) the carbonation kinetics compared with static carbonation of the same duration. The number of revolutions is a significant process parameter to explain the amount of increase in the carbonation kinetics induced by the drum rotation. The carbonation duration acts in a complementary way as in a static process. Of course, this conclusion is limited to the range of the process parameters tested in the present study.

In one day process with optimal initial moisture and rotation speed of 100 rpm, about 10% of the potential of carbonation of the 10/20 concrete aggregate was achieved. The carbonation kinetics slightly decreased during the processing time, so about 60% of carbonation was obtained after seven days. Indeed, the coarse concrete aggregate's carbonation was faster than their attrition for the given process parameters. One can deduce that the coarse concrete aggregate's carbonation depth increased during the process, contributing to progressively decreasing carbonation rate in the processing time. The fine particles were mostly completely carbonated, probably, before they were created.

The size, shape, composition, water absorption capacity, and enveloped density of the recycled concrete aggregate was evolved during the process. The shearing significantly rounded the recycled concrete aggregate. Conversely, the shape of the finer parts, created by attrition, seemed not to be changed when using carbonation or without carbonation (aggregates sheared in the drum without CO₂). The fines were mainly composed of cement paste (more than 90%). Consequently, the paste content of the recycled concrete aggregated diminished after shearing. Furthermore, the water absorption capacity exponentially diminished with the amount of CO₂ sequestrated. It seemed that the decrease in paste content of the concrete aggregate did not significantly participate in the decrease of the water absorption.

Declaration of competing interest

The authors declare that they have no known competing financial interests relationships that could have appeared to influence the work reported in this paper.

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