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# CO<sub>2</sub> uptake by a bed of recycled concrete aggregates crossed by a flow of gas with a high concentration of CO<sub>2</sub>: influence of temperature and initial water content

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## Abstract

Accelerated carbonation of recycled concrete aggregates (RCA) is a promising approach for improving their performance and mineralising CO<sub>2</sub> from industrial gases. This study explores the combined effect of temperature (50°C – 80°C) and initial water content (4% - 10%) on carbonation of a bed of RCA (0/4 mm crushed mortar). To this end, carbonation tests were carried out in a reactor through which a model gas rich in CO<sub>2</sub> was passed, simulating industrial combustion gases. The evolution of carbonation was monitored both by analysing the gas passing through the reactor and by thermogravimetric characterisation. The results show that water content is a key factor: at low initial water content (4%), the temperature of 50°C limits drying, favouring the reaction, while at high water content (10%), a high temperature (80°C) accelerates drying and CO<sub>2</sub> diffusion.

## 1 Introduction

Construction and public works waste management is moving towards circular economy approaches, through the recovery of recycled concrete aggregates (RCA). However, their reuse in concrete mix design is limited by their lower performance to that of natural aggregates (NA), due to their often-higher porosity [1]. Accelerated carbonation of RCA, using CO<sub>2</sub>-rich industrial gases, appears to be a promising solution for improving their properties [2]. In addition, such process enables CO<sub>2</sub> to be mineralised in the form of calcium carbonate (CaCO<sub>3</sub>) and could be used as a carbon capture and utilisation (CCU) strategy [3].

Carbonation of RCA is based on a series of physico-chemical mechanisms. Firstly, CO<sub>2</sub> diffuses through the pores of the material and dissolves in the water present on the surface of the reactive phases, especially portlandite (Ca(OH)<sub>2</sub>) and hydrated calcium silicates (C-S-H). This dissolution leads to the formation of carbonate ions (CO<sub>3</sub><sup>2-</sup>), which then precipitate as CaCO<sub>3</sub> by reacting with the dissolved calcium from hydrates. This phenomenon modifies the microstructure of RCA by reducing their porosity and thus improving their compactness and mechanical properties [4,5].

Carbonate gases released by industrial sites such as cement factories are at temperatures ranging from 80°C to 200°C [6], and their use in carbonation reactors could lead to a rise in temperature. This raises important questions about the impact of this parameter on the carbonation process.

As part of the ANR CO<sub>2</sub>NCRETE project, this study aims to understand the influence of both temperature and water content of RCA to optimise a carbonation process using flue gases. To this end, a fixed-bed carbonation device through which a gas modelling a flue gas flows was developed. The present paper gives results and analyses on carbonation of a recycled sand at 50 and 80°C with initial water contents of 4 and 10%.

## 2 Materials and methods

### 2.1 Materials

A standardized mortar was produced to ensure a homogeneous composition of recycled concrete aggregates (RCA). It was formulated with a Water/Cement ratio of 0.5 and Sand/Cement ratio of 2.16, using CEM I 52.5 R cement and silica sand (0/2 mm). This mix design is commonly used in our

previous studies (With this ratio, the volume fraction of sand is 0.5). Silica sand was used to avoid any interference for the quantification of calcium carbonate from carbonation.

The mortar was poured in slab form (1.6 m<sup>2</sup>, 3.5 cm thick), broken after 24 h, then stored under water for 2 to 5 months before crushing into aggregate (0/4 mm). Some RCA were characterized before carbonation as a reference (RCA-NC).

## 2.2 Carbonation protocol

A “model” gas close to the gases emitted by industries was prepared by enriching air with CO<sub>2</sub> and water. As an example, the characteristic values for a gas emitted by a cement plant are as follows: water vapor pressure of the gas around 7 kPa and CO<sub>2</sub> concentration of 14-33% (v/v) [7]. The prepared gas was thus composed of 15% CO<sub>2</sub>, a water vapor pressure of 7 kPa at a temperature of 50°C. The total flow rate of the gas feeding the reactor was chosen at 20 NL/min.

Prior to carbonation, after preparation (crushing to target water content), both the tested RCA sample and the reactor were brought to test temperature for 4 h. The gas preparation line was started up 1 hour before the beginning of the test to allow the gas composition to stabilize.

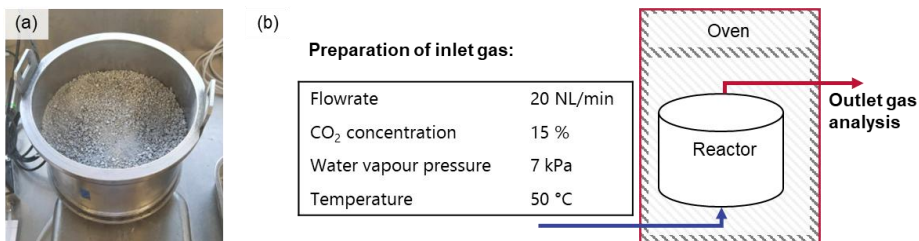


Fig. 1 (a) Photo of the fixed-bed reactor filled with a RCA sample and (b) diagram of the device.

The RCA batch of 2.5 kg was inserted into the reactor (Fig. 1a), which was closed with a lid. The reactor is cylindrical in shape, with a diameter of 20 cm and a height of 15.5 cm. Gas inlets and outlets are provided by holes drilled in the bottom and top of the reactor respectively. The RCA bed, about 7 cm high, is placed on a grid at the bottom of the reactor. This grid, cut from an 80 μm sieve, is adjusted to the shape of the bottom of the reactor. It allows the gas to pass through, while retaining the RCA to prevent them from obstructing the gas supply.

Testing began when the reactor was connected to the gas line between the gas preparation and analysis systems. The gas was fed in at the bottom of the reactor and discharged at the top (Fig. 1b). The carbonation tests were stopped after 2 hours. At the end of the test, the reactor was disconnected from the gas line, and the gas preparation line was connected to the analysis system to check that the composition of the prepared gas remains unchanged between the start and end of the test. The RCA were weighed before being placed in an oven at 60°C. Once their mass has stabilized after several days (< 0.05%/24h), we measured the mass of the dry sample to obtain the water content at the end of the test. The water content was determined by taking the mass of RCA at 60°C before carbonation as a reference. The sample was then quartered, and 500 g of material was analysed by “Macro-TGA”. This protocol is described below.

## 2.3 Monitoring of carbonation

Two innovative methods were used to monitor carbonation. The first is an indirect method based on measuring the drop in CO<sub>2</sub> molar flow between the reactor inlet and outlet. The system for measuring the gas characteristics consisted especially of an infra-red sensor for measuring CO<sub>2</sub> concentration, two barometers and a temperature and humidity probe.

The inlet gas, considered stable, was monitored before and after the test, and the outlet gas was analysed continuously during the test. By calculating the flow balance of the various components (Fig. 2), it was possible to obtain the evolution of the CO<sub>2</sub> uptake of RCA and water exchanges between the RCA sample and the gas. The mass of CO<sub>2</sub> uptake by the RCA ( $Q_{CO_2}^{uptake}$  [kg]) and the mass of water removed from the RCA ( $Q_{H_2O}^{exchange}$  [kg]) are determined by integrating the difference between the outflow and inflow.

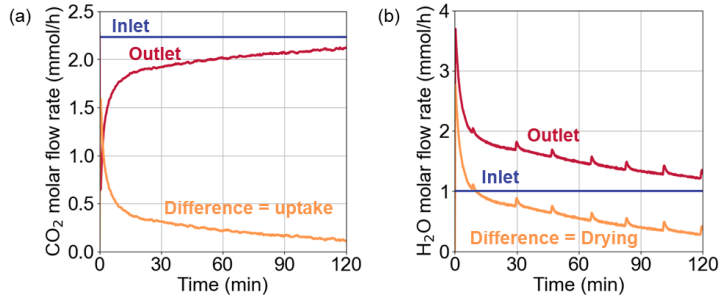


Fig. 2 Example of the evolution of flow rates of (a) CO<sub>2</sub> and (b) H<sub>2</sub>O during the carbonation test at 80°C and initial water content of 4%.

To determine changes in water content, the mass balance of the water in the sample is analysed:

$$m_{H_2O}^f = m_{H_2O}^i - Q_{H_2O}^{exchange,f} + Q_{H_2O}^{carbo,f} \quad (1)$$

With:  $m_{H_2O}^i$  [kg] and  $m_{H_2O}^f$  [kg] the final and initial masses of water in the material determined with the water content of the samples before and after carbonation respectively.  $Q_{H_2O}^{exchange,f}$  [kg] is the mass of water added to the gas throughout the experiment by water exchange with the RCA

$Q_{H_2O}^{carbo,f}$  [kg] is the total mass of water produced during the 2 hours carbonation reaction by the dehydroxylation of the portlandite and the dehydration of the hydrates, which can therefore be deduced from the previous data:

$$Q_{H_2O}^{carbo,f} = m_{H_2O}^f - m_{H_2O}^i + Q_{H_2O}^{exchange,f} \quad (2)$$

The carbonation of certain hydrates, such as C-S-H, does not necessarily produce water molecules [8]. As a result, the evolution of water production during the reaction  $Q_{H_2O}^{carbo}$  is difficult to assess. To estimate the average water content of the sample,  $w$  [%], and its uncertainty over time, three different hypotheses were considered regarding the production of water by carbonation.

1. Early water production: At the beginning of the reaction, each mole of CO<sub>2</sub> trapped leads to the formation of one mole of water ( $Q_{H_2O}^{carbo} = Q_{CO_2}^{uptake} M_{H_2O}/M_{CO_2}$ ), until the total produced water reaches a final value  $Q_{H_2O}^{carbo} = Q_{H_2O}^{carbo,f}$ . After this point, water production is assumed to stop. This hypothesis tends to overestimate the water content.

2. Delayed water production: No water is produced at the beginning of the reaction. Water formation starts only when:  $Q_{CO_2}^{uptake} = Q_{CO_2}^{uptake,f} - Q_{H_2O}^{carbo} M_{CO_2}/M_{H_2O}$ . From this point on, each mole of CO<sub>2</sub> bound results in one mole of water produced ( $Q_{H_2O}^{carbo} = (Q_{CO_2}^{uptake} - Q_{CO_2}^{uptake,f}) M_{H_2O}/M_{CO_2} + Q_{H_2O}^{carbo,f}$ ). This hypothesis underestimates  $w$ .

3. Proportional water production: Water production is assumed to be proportional to the progress of the carbonation reaction:  $Q_{H_2O}^{carbo} = Q_{CO_2}^{uptake} \times Q_{H_2O}^{carbo,f} / Q_{CO_2}^{uptake,f}$ .

For all hypothesis, evolution of water content is then calculated as:

$$w = \frac{m_{H_2O}^i - Q_{H_2O}^{exchange} + Q_{H_2O}^{carbo}}{m_{60^\circ C}^i} \times 100 \quad (3)$$

The second method is a thermogravimetric characterisation using macroscopic quantities of material (Macro-TGA). After carbonation and quartering of 500 g, the aggregates were sieved to 2 mm, 1 mm, 0.5 mm and 0.2 mm. Each mass fraction obtained was placed in an oven to measure its mass at 3 temperatures (415°C, 475°C and 950°C). For each mass measurement at a given temperature, the samples were kept at the desired temperature for at least 3 h, then the temperature was lowered to 105°C so that the samples can be weighed outside the oven.

Following a preliminary study, we have shown that under these heating conditions, between 415°C and 475°C, portlandite (Ca(OH)<sub>2</sub>) decomposes into CaO and releases one molecule of water, and between 475°C and 950°C, calcium carbonate (CaCO<sub>3</sub>) releases one molecule of CO<sub>2</sub> to form CaO. From the masses measured, the contents of portlandite, calcium carbonate and CO<sub>2</sub> were determined. In this study, the carbonation of C-S-H was not specifically quantified, as the Macro-TGA cannot properly detect it.

## 2.4 Experimental program

The initial water content of the RCA was chosen at 4% and 10%, while the temperature of the oven in which the reactor was placed was set at 50°C and 80°C. Thus, carbonation was carried out for the 4 possible combinations of these parameters.

The desired water content was obtained after curing and RCA grinding. The water content is measured at 10% and they are then considered to be fully saturated. Some tests were carried out on these water-saturated aggregates, which were kept for a maximum of 12 days in a sealed bag before use. The RCA prepared with a water content of 4% were obtained in two stages. Drying was first carried out in an oven at 60°C until the mass stabilised. The aggregates were then moistened by filling a bag with the dry aggregates and the mass of water corresponding to the target water content. The mixture was homogenised by shaking the bag, which then remained closed for several days. Note that the water content was not measured after preparation.

## 3 Results and discussion

### 3.1 Characterisation of non-carbonated mortar

The non-carbonated RCA (RCA-NC) was characterised using Macro-ATG (Fig. 3). The smallest grains have a higher CO<sub>2</sub> content because they contain more cement paste. Cement paste breaks more easily than sand grains during crushing [9]. Note that the quantity of CO<sub>2</sub> bounded by the material is expressed in g per kg of material heated to 950°C. The latter mass is a reference value because it does not change during carbonation and water exchange with the environment.

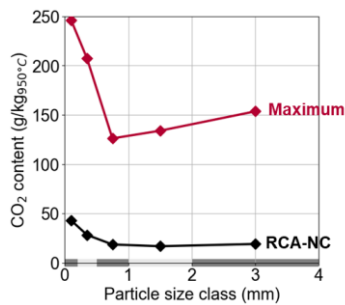


Fig. 3 CO<sub>2</sub> content of the RCA-NC and theoretical maximum versus the particle size class.

The theoretical maximum CO<sub>2</sub> content that can be obtained per fraction was calculated according to the chemical compositions of the cement and the mortar mix proportion. It was then distributed among the particle size class according to the paste fraction deduced from portlandite and CaCO<sub>3</sub> content.

From the measured CO<sub>2</sub> content of the carbonated material ( $T_{CO_2}$  [mol/kg<sub>950°C</sub>]), a degree of carbonation (DC [%]) is calculated as follows:

$$DC(\%) = \frac{T_{CO_2} - T_{CO_2}^{NC}}{T_{CO_2}^{max} - T_{CO_2}^{NC}} \times 100 \quad (4)$$

### 3.2 Influence of water content and temperature on carbonation

Continuous measurement of the concentration at the outlet of the carbonation reactor was used to determine changes in the quantity of CO<sub>2</sub> bound by the RCA bed (Fig. 4).

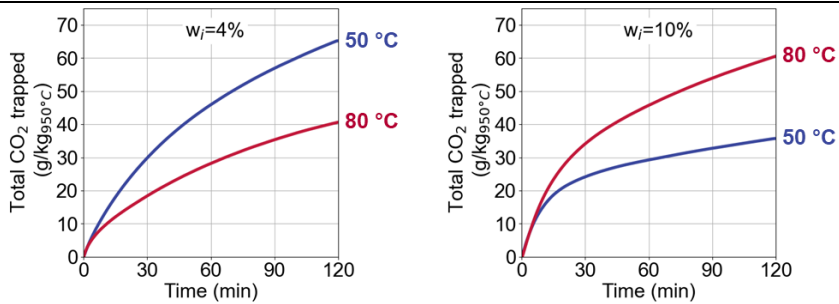


Fig. 4 Evolution of the total CO<sub>2</sub> trapped determined with the indirect method with initial water content of 4% (left) and of 10% (right).

All the curves have a similar shape, with a decrease of the carbonation rate with time. Over 120 min, the most effective carbonation occurred at 50°C for an initial water content of 4%. Under these conditions, the RCA sample bound 65 g/kg<sub>950°C</sub> of CO<sub>2</sub>.

The effect of temperature is not the same depending on the initial water content. When the initial water content is 4%, carbonation is less effective the higher the temperature. Conversely, when the initial water content is 10%, carbonation is more effective at 80°C.

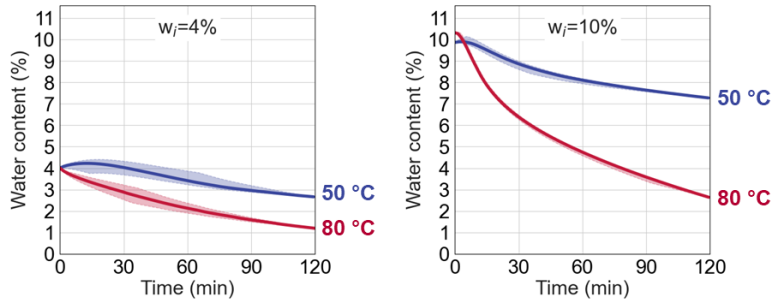


Fig. 5 Evolution of the water content (left) with initial water of 4% and (right) with initial water content of 10%. The solid line corresponds to the third hypothesis described in Section 2.3, while the shaded area represents the uncertainty range estimated from the first two hypotheses.

In order to study the effect of temperature, we need to consider the hydrous state of the cementitious materials, which is variable and has a strong impact on carbonation. We were able to estimate the average water content of the RCA bed by monitoring the flow rates of H<sub>2</sub>O in the gas as it exited. Figure 5 shows that the average water content of the beds fell over the course of all the tests. Drying during the tests is therefore faster than the production of water due to the carbonation reaction. The uncertainty linked to the evaluation of the water produced by the reaction is not considered to be important.

The water content of the samples decreases more rapidly the higher the temperature. The water vapour in the gas entering the reactor is between 6 kPa and 8 kPa, which corresponds to a relative humidity (RH) of 60% at 50°C and 15% at 80°C. Increasing temperature therefore has a dual effect on water saturation: it increases the deviation from water balance and accelerates water exchange rates. The final water content of the sample initially saturated and carbonated at 80°C is even lower (2.6%) than that of the sample initially at 4% and carbonated at 50°C (2.7%). These two conditions also have close to the same amount of bound CO<sub>2</sub>.

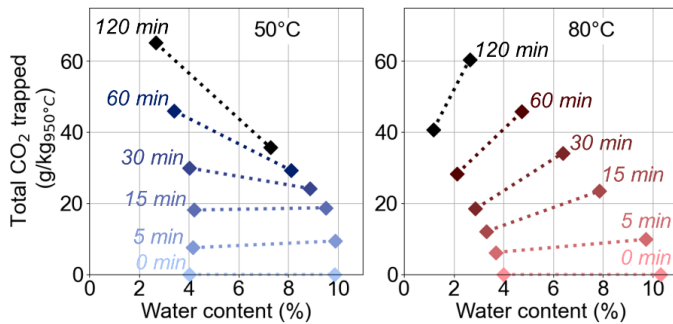


Fig. 6 Changes in cumulative CO<sub>2</sub> fixed determined with indirect method as a function of the average water content of RCA at 0, 5, 15, 30, 60 and 120 min at 50°C and 80°C. The segments are plotted to aid reading but do not represent data.

The total amount of CO<sub>2</sub> trapped as a function of water content is shown at different time intervals in Fig. 6. At 50 °C (left), two distinct phases can be observed. During the first 15 minutes, carbonation is slightly more efficient in the fully saturated sample. After that, samples with lower initial water content tend to trap more CO<sub>2</sub>. This suggests that, in the early stages of the reaction, carbonation primarily occurs at the surface of the grains. Indeed, the diffusion of CO<sub>2</sub> through the pore network is a limiting factor from the very beginning of the process (Morandea et al., 2014), and the enhancement of carbonation by increasing the exposed surface area (Kikuchi and Kuroda, 2011) supports the idea that the reaction mainly takes place at the surface. In this initial stage, higher water content may facilitate the dissolution of reactive species at the grain surface, without requiring CO<sub>2</sub> to diffuse deeper into the material.

As carbonation progresses, CO<sub>2</sub> must penetrate further into the porous structure. At this point, diffusion becomes a more critical step, and high-water saturation slows down CO<sub>2</sub> transport within the material, thereby reducing the overall carbonation rate in saturated samples.

At 80 °C (Fig 6. right), the initially saturated sample systematically shows higher levels of carbonation. This may be due to a more rapid decrease in water content at higher temperatures, which accelerates gas diffusion. Furthermore, with rapid drying, the sample with an initial water content of only 4% may have a water deficit, limiting the carbonation reaction due to insufficient dissolution of reactive species.

These observations highlight the significant role of temperature in influencing the drying behavior of the samples during the carbonation process. Higher temperatures accelerate water evaporation, which alters the internal moisture distribution and therefore affects local carbonation conditions. While this drying can enhance carbonation in initially saturated samples by bringing them closer to optimal water content, it may conversely reduce carbonation efficiency in samples with low initial moisture, as the material becomes too dry for the dissolution and diffusion steps required by the reaction.

### 3.3 Effect of the size of RCA

The above results relate to the entire bed, including the entire 0/4 mm sand. To have more details on the state of material carbonation, we carried out Macro-TGA on the different particle size classes after two hours of carbonation.

The influence of the input parameters on the degree of carbonation is shown separately for each size class in Fig. 7 Overall, the highest degrees of carbonation follow the diagonal from ‘coldest and driest’ to ‘hottest and saturated’. An optimum is obtained in conditions where the two parameters, initial water content and temperature, provide water content during the experiment that ensures good dissolution of the reagents while not restricting diffusion too much.

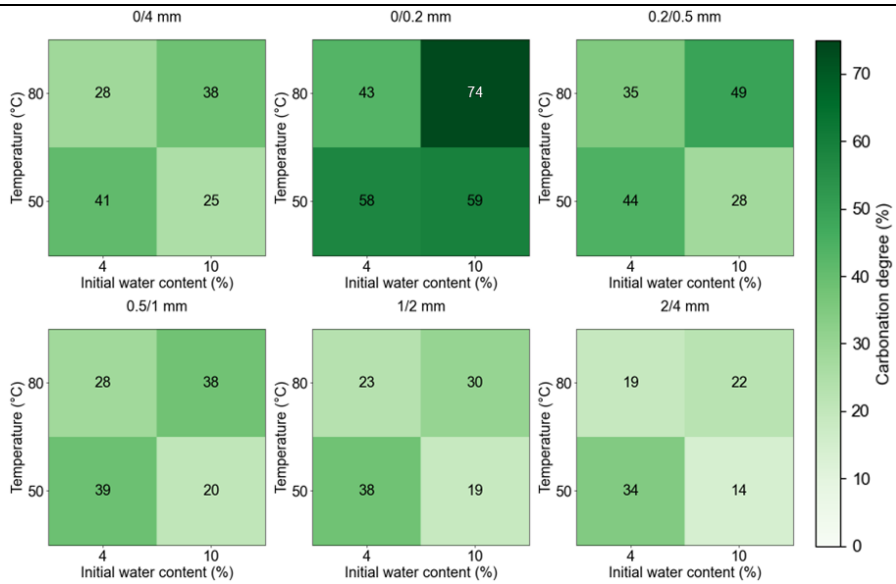


Fig. 7 Carbonation degree measured with Macro-TGA depending on the temperature and initial water content of the RCA for each particle size class.

The smaller the RCA, the greater the degree of carbonation, as the surface area in contact with the gas is increased. The effect is greatest for an initial water content of 10%. While gaseous diffusion is limiting under these conditions, the reduction in size means that diffusion has less effect on the rate of carbonation.

#### 4 Conclusions

The combined effect of temperature (50°C – 80°C) and the initial water content of RCA (4% - 10%) on the carbonation of a RCA bed through which CO<sub>2</sub>-rich gas flows was studied. The results show that the water content plays a decisive role in ensuring the efficient diffusion of CO<sub>2</sub> and the availability of the hydrates required for the reactions. For initially slightly water-saturated RCA (4%), the temperature of 50°C favours carbonation by limiting excessive drying. On the other hand, for initially fully saturated RCA (10%), a high temperature of 80°C accelerates drying, thus improving CO<sub>2</sub> diffusion and carbonation. At the optimum temperature, a degree of carbonation of over 40% was achieved in only 2 hours. The maximum amount of bound CO<sub>2</sub>, i.e., 65 g/kg<sub>950°C</sub>, was achieved for an initial water content of 4% at 50°C. The smaller RCA, the faster carbonation and the more carbonation can take place at a high initial water content.

A continuation of this work would be to characterise the homogeneity of carbonation and the water content in the bed. The water content was assessed for the entire bed. It is not known whether the water is in the intragranular or intergranular porosity. These results can be used to optimize an industrial system. The temperature in the reactor could be adjusted according to the initial water content of the GBR by adjusting the supply of hot gases emitted by industry.

#### Acknowledgements

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