# French projects on the $CO_2$ uptake by carbonation of recycled concrete aggregates

**Philippe TURCRY[1*], Jean-Michel TORRENTI[2], Pierre-Yves MAHIEUX[1], and Abdelkarim AÏT-MOKHTAR[1]**

*[1]Laboratoire des Sciences de l'Ingénieur pour l'Environnement (LaSIE), UMR 7356 CNRS, La Rochelle Université, 17000 La Rochelle, France, philippe.turcry@univ-lr.fr, (corresponding author*), pierre-yves.mahieux@univ-lr.fr, karim.ait-mokhtar@univ-lr.fr*

*[2]Université Gustave Eiffel, MAST-EMGCU, 77454 Marne-la- Vallée, France, jean-michel.torrenti@univ-lr.fr*

## Abstract

Reducing the carbon footprint of concrete is becoming crucial as its global production is expected to increase in the coming years. One way to achieve this goal is to use its ability to mineralize $CO_2$. Several French projects have focused on the carbonation of recycled concrete aggregates (RCA). The present paper summarizes their main results. An initial project has shown that $CO_2$ uptake by the natural carbonation is not effective because, after crushing, RCA are stored in stockpiles for several months before use. The FastCarb national project has proved the feasibility of processes using industrial $CO_2$-rich gases and shown their limits through life cycle analysis. The ongoing ANR $CO_2$ncrete project aims at better understanding carbonation at both grain and bed scales with various conditions (temperature, pressure, gas flow) to propose solutions to maximize $CO_2$ uptake.

## 1 Introduction

Carbonation of concrete is a reactive transfer of gaseous $CO_2$ that has two main consequences: a drop in the pH of concrete and a mineralization of $CO_2$, the main greenhouse gas. For a long time, carbonation was studied with regard to the first consequence in the scope of reinforced concrete structures durability, e.g., [1]. Today, the second one is the subject of many works, since concrete can also be considered a carbon sink, e.g., [2]. Concrete carbonation is a promising process for Carbon Capture and Utilisation (CCU). It could be one way to achieve the reduction of concrete carbon footprint [3]. The latter is becoming crucial as the global production of cement and concrete leads to more than 7% of greenhouse gas emissions over the world.

Although concrete structures carbonate during their service life, the resulting $CO_2$ uptake is relatively low and depends on many factors [4]. At the end of life, concrete structures are demolished and crushed to produce recycled concrete aggregates (RCA) usually used for road construction and more and more re-used in concrete mixtures. RCA are *a priori* one of the best vectors for $CO_2$ uptake [3]. Indeed, the crushing of concrete waste creates large surface area increasing the exposure of cementitious hydrates to the reactive gas. In recent years, many RCA carbonation techniques have been proposed and mainly tested in laboratory [5]. These techniques involve more or less treatments of RCA before carbonation, from "direct carbonation" of RCA after crushing to "enforced wet carbonation" of recycled concrete powder (RCP) obtained by grinding RCA. The latter is obviously the most effective providing a high degree of carbonation. The carbonated RCP, having pozzolanic properties, could be then used as supplementary cementitious materials [6].

In recent years, several French projects have focused on the $CO_2$ uptake of RCA. It was chosen to consider the "direct carbonation" of RCA, i.e., a carbonation without pre-treatment such as grinding or process to separate cement paste from natural aggregates. This approach can be considered "low tech", limiting *a priori* the environmental impact of the process itself. The final product should be "carbon negative aggregates" for use as substitutes of natural aggregates in both road construction and concrete production.

$CO_2$ mineralisation by carbonation of RCA is envisaged as an accelerated process, using $CO_2$-rich gas, which raises several questions. First of all, we can question the real benefit of an accelerated process. In fact, RCA can carbonate naturally in contact with atmospheric $CO_2$. In life cycle analyses, it is often considered that the carbonation that takes place during the service life is strongly accelerated after crushing, thanks to the increase in the surface area [2]. Thus, this raises the question of the atmospheric carbonation of RCA on the recycling platform, where they are stored in stockpiles for several months. Secondly, accelerated processes have so far mainly been tested in the laboratory. The feasibility of the up-scaling should be addressed. Is it possible to develop “industrial carbonators” with low investment and quite few technologies? Both environmental and economic benefits are also determinant and must be verified before large-scale deployment [5]. Thirdly, the parameters of accelerated carbonation (such as relative humidity of the gas, temperature, pressure, and water content of the RCA) need to be understood to enable the process to be optimized.

In the following, we summarize some results of three French projects, namely “Carbone Gris”, “national project FastCarb” and “ANR $CO_2$ncrete”, giving some answers to the previous questions.

## 2 Can RCA be naturally carbonated in a stockpile?

### 2.1 Modelling a case study

The “Carbone Gris” project addressed the atmospheric carbonation of RCA stockpiles during the several-month period of storage after crushing. Our approach was the same as that proposed by Thiery et al. [7], i.e., modelling the reactive transfer of $CO_2$ inside a granular medium made of RCA. We considered ideal boundary conditions for carbonation, i.e., no rain during the storage period, assuming that wetting of the medium would stop any inter-granular gas diffusion and therefore carbonation. The main equation of the model is the mass balance of $CO_2$:

$$\frac{\partial n_c}{\partial t} = -\mathrm{div}(J_D + J_A) - r \qquad (1)$$

Where: $n_c$ is the $CO_2$ content in the granular volume ($kg.m_t^{-3}$), $J_D$ and $J_A$ are the diffusive and advective flux of $CO_2$ through the stockpile, respectively ($kg.m^{-2}.s^{-1}$) and r a sink term ($kg.m_t^{-3}.s^{-1}$). $m_t^3$ refers to the total volume of the granular medium.

The model was applied to predict the carbonation of a 6 m high industrial stockpile of Recycled aggregates (RA) in La Rochelle (fig. 1). From samples taken in the stockpile, input data of the model were determined using laboratory tests, namely:

- the porosity and water content of the stockpile,
- the gas diffusion coefficient for different water content (input for $J_D$),
- the intrinsic gas permeability (input for $J_A$),
- the $CO_2$ binding rate and binding capacity (input for r).

The latter were determined from an original test protocol detailed in [8]. Missing data were estimated from empirical models. For instance, the gas permeability was determined from a measured water permeability and an empirical model established for granular soils [9].

Fig. 1 Studied stockpile of RCA in La Rochelle (France) photographed by drone.

Thanks to oedometric tests, the porosity of the stockpile was found equal to 35%. For this packing density, the $CO_2$ diffusion coefficient of the granular medium at dry state was $1.2.10^{-6}$ $m^2.s^{-1}$ and the gas intrinsic permeability was $3.10^{-12}$ $m^2$. The stockpile is equivalent to a low-permeable soil. The $CO_2$ binding capacity was found equal to around 27 g of $CO_2$ per kg of RA (after 4 months in room conditions).

To simulate the effect of wind, a commercial code (Autocad CFD©) was used to assess the maximal pressure gradient due to gas flow on a pile with simplified geometry. For a wind of 20 $m.s^{-1}$ (high speed), we obtained a pressure gradient of 180 $Pa.m^{-1}$.

## 2.2 Simulation results

Fig. 2 gives the results in the most favourable case, i.e., considering that the granular medium is dry. This case ensures the highest $CO_2$ diffusion and, in a windy condition, the highest gas permeation. Two $CO_2$ binding rates (sink term) were used for the simulations, the highest and the lowest deduced from the measurements.

The main result from these simulations of a 4-month atmospheric carbonation is that the carbonation depth is very low (less than 25 cm for a slow binding) and even negligible compared to the stockpile dimensions. Only the “skin” of the stockpile is carbonated. The amount of bound $CO_2$ represents less than 1% of the total $CO_2$ binding capacity of the RCA in the stockpile. The effect of a high-speed wind (assumed to be continuous for 4 months) is almost negligible due to the low permeability of the medium. Note also that, if carbonation is assumed to be controlled only by gas diffusion (fast binding of $CO_2$ by RCA), the carbonation depth can be assessed with the square-root-of-time model from the binding capacity and the diffusion coefficient.

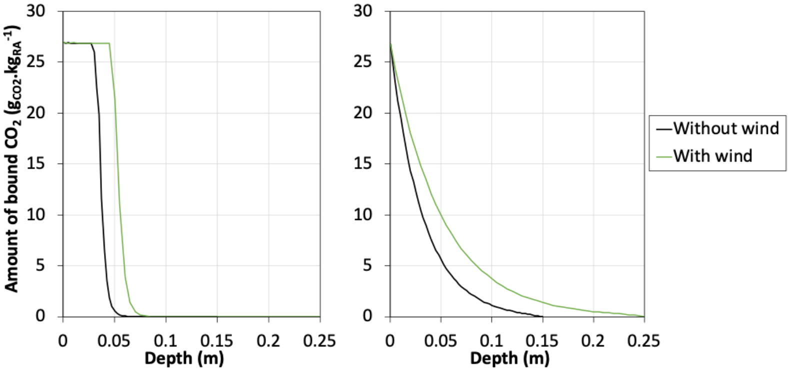

Fig. 2 Profiles of the amount of $CO_2$ bound by the stockpile (considered dry) after 4 months. Simulations were carried out with and without wind and considering two $CO_2$ binding rates, fast (left) and slow (right).

These simulations allow us to conclude that it would be preferable to opt for an accelerated carbonation to benefit from the $CO_2$ sequestration capacity of RCA. During the most common use of RCA, i.e. as pavement layer, the $CO_2$ uptake will be very slow, according to Hou et al [10]. This also suggests that RCA should be fast-carbonated before use.

# 3 Can RCA be fast carbonated on a large scale using industrial gases?

## 3.1 Demonstrators

During the national project FastCarb, large-scale demonstrators were developed from existing facilities and turned into “carbonators” [11]. These were installed in cement works and connected to exhaust gases with a $CO_2$ concentration of around 15%. The relative humidity was over 70% and the temperature was between 70 and 90°C.

Two types of dynamic systems were studied (Fig. 3):

- A rotating drum, whose movement allows a good contact between RCA and the feeding gas (temperature of around 50°C and relative humidity of 90%).
- A fluidized bed dryer tested in semi-continuous mode.

From these reactors, which were not optimized during the project, 3 to 10 tons of RCA can be treated in 1 hour. During the project, the amount of bound $CO_2$ was between 31 and 39 $kg_{CO2}/t_{RCA}$ for fine RCA (sand 0/4mm) and between 5 and 12 $kg_{CO2}/t_{RCA}$ for coarse RCA (4/16 mm) for 1 hour of treatment. The treated RCA were obtained from crushing of ready-mixed concrete not used on site.

In laboratory, such carbonation levels were obtained after 24 hours of carbonation at 20°C in static conditions for an equivalent $CO_2$ concentration [12]. The difference in $CO_2$ binding rate between laboratory and industrial reactor is due to multiple factors, such as temperature, gas moisture, and RCA movement.

Fig. 3 Two demonstrators developed during the project FastCarb. Left: rolling drum reactor at the Créchy plant (©Vicat). Right: fluidized bed reactor at the Val d'Azergues plant (©Holcim).

The experiments carried out with the demonstrators have proved that it is possible to use industrial processes for accelerated carbonation of RCA at sites that emit $CO_2$. The installation of these carbonators did not require any major investment (180 000 euros maximum) or changes to the production process. Of course, many optimizations that better control $CO_2$ concentration, humidity, and temperature of the gas used could be proposed to maximize the $CO_2$ uptake. Note also that secondary gases in the flue gas could lead to precipitation of specific compounds. However, laboratory studies did not show adverse effects on the $CO_2$ binding due to $SO_2$ and Nox in low concentrations (these gases are present in cement plant exhaust gas for instance).

### 3.2 Assessment of $CO_2$ balance

An assessment of the $CO_2$ emissions of prototype processes, such as those presented previously, was carried out using the LCA principle. From data collected by the industrial support, i.e., the Vicat factory in Créchy (rolling drum reactor), the total emissions of the carbonation process were estimated at 1.2 $kg_{CO2}$ per ton of carbonated RCA [13]. The crushing process was not taken into account in this calculation. This value can be compared to the amount of sequestered $CO_2$, i.e., -5 $kg_{CO2}/t_{RCA}$ for the coarse RCA (4/16 mm) and -31 $kg_{CO2}/t_{RCA}$ for the finer fraction (0/4 mm). This comparison reveals that the low binding capacity of the coarsest RCA in the tested process conditions makes them of little interest for $CO_2$ mineralisation.

Indeed, one must consider the transportation of RCA since, in the present approach, RCA are carbonated at the site emitting $CO_2$. Two transport distances must be considered: the distance between the recycling platform where concrete wastes are crushed and the carbonation site, and the distance between the latter and the construction site (road application) or the concrete plant. Fig. 4 shows the $CO_2$ emissions related to 1 ton of carbonated RCA versus the total distance. The $CO_2$ emissions are the sum of the process emissions, the amount of $CO_2$ by carbonation (negative) and the transport emissions. The latter were calculated assuming an emission factor of 0.16 $kg_{CO2}.t^{-1}.km^{-1}$ [13]. Fig. 4 confirms that the coarse RCA are little interesting since the distance that hinders the $CO_2$ uptake (carbon neutral) is lower than 25 km. This value is low because of the order of magnitude of the distance over which RCA are usually transported to the worksite to our knowledge. On the contrary, the $CO_2$ balance remains negative in the case of the carbonated sand for long distances.

From these assessments, the locations of both recycling platforms and carbonation sites appear as key factors that should be optimized. They also show that the benefits of $CO_2$ mineralization lie in the high efficiency of the carbonation process.

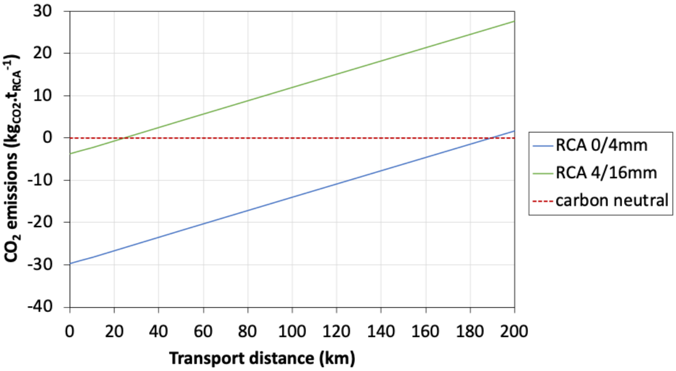

Fig. 4 $CO_2$ emissions per ton of carbonated recycled aggregates (fine and coarse) considering the $CO_2$ emissions of the reactor, the amount of $CO_2$ bound by carbonation and the emissions of the total transport distance.

## 4 Towards a high-efficiency carbonation process

As recalled in the introduction, we chose the $CO_2$ mineralisation technology for producing "carbon negative RCA" without treatment of the aggregates before carbonation. However, this approach does not exclude the treatment of the gas used for carbonation. LCA has shown that the transport of RCA to the carbonation site is a key factor for the $CO_2$ balance. It might be preferable to carry the $CO_2$ to the recycling platform rather than transporting the RCA to the treatment site, as was studied as part of the FastCarb project. Therefore, high-pressure carbonation was addressed during the project ANR "$CO_2$ncrete".

A laboratory study was carried out on mortar specimens using the set-up shown in Fig. 5. In the reactor, the specimen is exposed to pure $CO_2$ with controlled temperature and pressure. The tested mortar was made with a Portland cement (CEMI 52.5 N), a water-to-cement mass ratio of 0.5 and a siliceous 0/4 mm sand volume fraction of 50%. The tested specimens were cylinders of 27 mm in diameter. Various parameters were investigated, including the initial water content of the mortar, the gas temperature and the gas pressure. All details of the experimental protocols can be found in [14].

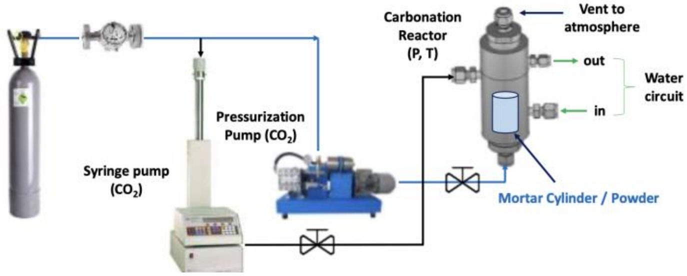

Fig. 5 Experimental set-up of high-pressure carbonation [14].

A quasi-full carbonation degree of the mortar cylinder (94%) was obtained in 30 min in the case of carbonation at 100 bar and 80°C for an average water content of 1.6% (i.e., a water saturation degree of around 20%). The carbonation degree was defined as the ratio of the amount of bound $CO_2$ and the binding capacity was assessed from the CaO content of the cement. The $CO_2$ uptake was 138 $kg_{CO2}/t_{cylinder}$. Fig. 6 gives tendencies highlighted thanks to a parametric study (for a 30-min carbonation). The initial water content is a key parameter. Surprisingly, the lower the water content, the higher the carbonation degree. There is no "optimum" of moisture conditions as usually observed for carbonation at atmospheric pressure (note that no experiment was carried out for a fully dried cylinder). The carbonation rate is increased when increasing both gas pressure and gas temperature. The effect of pressure is explained by an increase in $CO_2$ solubility with pressure. Temperature has an influence more difficult to explain due to competing effects (decrease of solubility, increase of gas and liquid diffusion). The lack of difference in carbonation degree between 60°C and 80°C may be a proof of this competition.

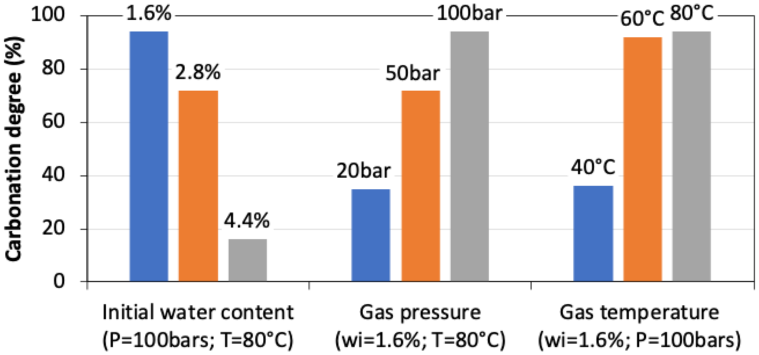

Fig. 6 Effect of test parameters on the carbonation degree of the mortar cylinder after 30 min: initial water content of the cylinder, gas pressure and gas temperature. Values of the changed parameter are given above the bars. Values of the unchanged parameters are in the x-axis title. This graph was obtained from data provided by [14].

The process studied is highly effective in taking full advantage of the material $CO_2$ binding capacity. However, it requires a certain amount of energy to heat and pressurize the gas. This raises questions about the total carbon footprint of the process. Knowing the energy consumption of the system studied here, it was calculated that the $CO_2$ emissions of the process represent about 10% of the $CO_2$ bound in the most favourable situation (100 bars, 80°C). For this calculation, a high-carbon energy mix was used (1100 $g_{CO2}$/kWh). This result is, of course, promising, but remains to be confirmed first for RCA and then for large-scale facilities.

## 5 Conclusion

Several French projects have recently focused on the $CO_2$ uptake by RCA. The first one, "Carbone Gris", addressed the atmospheric carbonation of RCA stockpiles during the several-month period of storage at recycling platforms. Based on input data from a case study, numerical simulations have shown that a natural carbonation of 4 months is limited to the skin of the stockpile even for the most favourable tested conditions. Thus, it is preferable to use an accelerated process to carbonate RCA to benefit from their full $CO_2$ binding capacity.

During the national project "FastCarb", large-scale reactors demonstrated the industrial feasibility of an accelerated process for RCA exposed less than two hours to a cement plant fume with a high $CO_2$ concentration. However, environmental studies have also shown that the efficiency of the process, quantified by the amount of bound $CO_2$ per ton of RCA, must be high, otherwise, the environmental benefit is cancelled out by the $CO_2$ emissions during the RCA transport.

The ongoing project ANR "$CO_2$ncrete" is now focusing on gaining a better understanding of carbonation phenomena at both scales of grains and granular medium. The aim is to better master the parameters of $CO_2$ binding processes, such as temperature, water content, and gas pressure. A high-pressure $CO_2$ process resulting in a carbonation degree higher than 90% is promising. It should be kept in mind that such technologies will first have to be assessed from both an economic and an environmental point of view before they can be transferred on a large scale [11].

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