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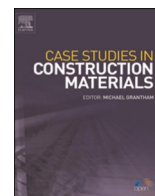
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The FastCarb project: Taking advantage of the accelerated carbonation of recycled concrete aggregates

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ABSTRACT

Recycled concrete aggregates (RCA) incorporate hydrates that can be carbonated. The FastCarb project aims to mineralize CO₂ within RCA, improving the quality of these aggregates by the clogging of the porosity and finally decreasing the CO₂ impact of concrete in structures. It has two main objectives: to optimize in laboratory conditions the accelerated carbonation process which can be transposed at an industrial scale at a suitable cost and to show that the process could be used in industrial conditions. This paper presents firstly the results obtained in laboratory conditions: it confirms that it is possible to store between 10 and 50 kg of CO₂/t of RCA depending on several factors (natural carbonation, water content, temperature, size of RCA...) and that the treatment allows an improvement of the properties like the water absorption. The feasibility of

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accelerated carbonation has been also demonstrated by setting up two industrial-scale demonstrators. The first results without optimization showed a CO₂ capture rate in the order of 3–4 %, i. e. 30 kg of CO₂ per ton of crushed concrete. 80 t of carbonated RCA were produced and used to produce C25/30 and C45/55 concretes. The measurement of the main properties (performance in the fresh state, mechanical performance, and durability) shows that the use of the carbonated RCA doesn't affect these properties. A variety of precast products (blocks, curbs, stairs) and parts of cast-in-situ structures (construction of walls) were fabricated showing the feasibility of using these aggregates in real situations. Finally, our project considers whether the accelerated carbonation process is environmentally acceptable and economically viable. The first results show that the distance of transport (by trucks) is a major factor and that transportation should be limited to maintain a positive impact of the accelerated carbonation (i.e. local sources of CO₂ should be used). The LCA and the economic study confirm that the sand fraction of RCA is the most interesting material for the uptake of CO₂. This is also interesting for a circular economy objective because recycled concrete sand is not easily used in concrete made with RCA.

1. Introduction

Involving more than 20 private and public partners, the FastCarb project (fastcarb.fr) is the result of the conjunction of two facts. On one hand, the production of cement is an important source of CO₂ in the world: the last IPCC report estimates that direct CO₂ emissions from carbonates in cement production are around 4 % of total fossil CO₂ emissions (1.6 % in France) [1]. On the other hand, a large quantity of recycled concrete coming from the deconstruction is available while natural resources have to be preserved. Even if recycled concrete aggregates (RCA) can already be used in concrete by adjusting mix designs (generally with an increase of the cement content [2,3]), their larger porosity, mainly for recycled sands, harms the performances of the concretes made with these aggregates [4–6].

However, recycled aggregates incorporate portlandite and calcium silicate hydrates that can be carbonated in a faster way. This has been demonstrated in laboratory conditions (see [7,8] for instance). The FastCarb project aims to mineralize CO₂ within RCA, improving the quality of these carbonated recycled concrete aggregates (CRCA) by clogging the porosity and finally decreasing the CO₂ impact of concrete in structures. The project has two main objectives: to optimize in laboratory conditions the accelerated carbonation process which can be transposed at an industrial scale at a suitable cost and to show that the process could be used in industrial conditions.

This paper presents firstly the results obtained in laboratory conditions, then in the industrial scale demonstrators. The study of concretes made with these CRCA has started and is presented. Finally, the first results concerning the life cycle analysis and the economic costs of the process are discussed.

2. Carbonation in laboratory conditions

2.1. Materials

In the project, several RCAs were used. Some of the aggregates were produced in laboratory conditions from known concretes and some were coming from industrial production (and, in this case, the parent concrete was not known). Table 1 summarizes the production of the different recycled aggregates used in the project.

2.2. Factors affecting accelerated carbonation of RCA

The carbonation rate of RCA depends on the water saturation state of the aggregates [7]. Indeed, when the water content is too low, the quantity of water present in the pores does not allow sufficient dissolution of the CO₂. On the opposite, too much water content in the pores slows the diffusion of CO₂, because the diffusion of a gas is much faster in the air than in water. The water content parameter

Table 1

Origin of the recycled aggregates used in the project FastCarb.

| RCA | Fraction (mm) | Production |
|-----|---------------|--|
| OC | 12–20 | obtained by crushing in the laboratory 20 years old ordinary concrete samples (W/C = 0.49, limestone aggregates, 353 kg/m ³ of Portland cement) |
| F1 | 0–4 | produced from laboratory samples of concrete, conserved in water for 5 months, then crushed. Made with 350 kg/m ³ of CEM I cement. |
| F2 | 0–4 | produced from laboratory samples of concrete, conserved in water for 5 months, then crushed. Made with 350 kg/m ³ of CEM II cement. |
| F3 | 0–4 | produced from laboratory samples of concrete, conserved in water for 5 months, then crushed. Made with 350 kg/m ³ of CEM III cement. |
| RS | 0–4 | obtained from returned ready mix concretes in a recycling center. The aggregates were obtained by crushing the hardened concretes. |
| RS | 1–4 | coming from the RS aggregates (previous line). The fraction 0–1 was suppressed for laboratory reasons. |
| RG | 4–16 | obtained from returned ready mix concretes in a recycling center. |

is therefore decisive in promoting accelerated carbonation and the uptake of CO₂ [7]. In addition, this parameter is easy to measure on an industrial installation and can be corrected if it is too low quite easily.

The temperature is another main parameter controlling the carbonation rate of concrete [9] and RCA [10]. The increase in temperature causes an acceleration of the diffusion of CO₂ in the cement matrix of the RCA, which subsequently leads to an increase in accelerated carbonation and therefore in the uptake of CO₂. However, there is an optimum temperature (less than 100 °C) to obtain maximum CO₂ uptake [9,10]. The CO₂ partial pressure enhances the CO₂ solubility in the water absorbed by the aggregate and facilitates carbon mineralization [11].

The use of gas issued from industrial processes, such as cement manufacturing (where it comes from combustion and decomposition of limestone), is naturally a way to have an environmental gas, with elevated temperature and higher CO₂ concentration. The use of this type of gas can bring both an acceleration of the carbonation process and a reduction of the CO₂ emitted by these operations. However, the gas from cement plants is impure and complex. Chen et al. [12] have shown that there are 29 different components in the gas emitted by a cement plant. Among them, CO₂ represents around 15 % while NO₂ and SO₂ are also present in small but significant quantities. Furthermore, according to Ahmad and Gersen [13], the solubility of CO₂ gas in water drops considerably in the presence of other gases such as NO₂ and SO₂. It should be added, that recycled concrete waste has an interesting potential to treat gases and capture NO_x and SO₂. Consequently, the presence of NO₂ and/or SO₂ impact the carbonation process and change the emission reduction of industrial gases.

To better understand the mechanisms of accelerating the carbonation of RCA in industrial conditions, experiments were conducted at a laboratory scale in three main directions: material characteristics and environmental parameters maximizing the carbon absorption, estimation of the CO₂ uptake capacity, and effect of the fast carbonation on the RCA properties. Experimental campaigns were developed in static, dynamic gas (CO₂ flow conditions) and dynamic granular packing (rotating drum) conditions, with pure CO₂, 15 % CO₂, or multi-component gas (CO₂, SO₂, and NO₂).

2.3. Fast carbonation in static conditions

The experiments on several RCA and the results of these tests were already published in [14]. The main conclusions of the tests are the following:

- The water content is indeed the most important parameter and there is a correlation between the optimum water content and the water absorption coefficient. To obtain the maximum CO₂ uptake, it is necessary to have an optimum water content of about 80% of the RCA's water absorption coefficient.
- Carbonation decreases the water absorption coefficient by filling pores due to the formation of calcium carbonates. The capillary porosity is decreased due to clogging of the pores. In addition, carbonation increases the microporosity of RCA, especially the porosity of the C-S-H gel, as a result of decalcification [15].

2.4. Effect of the temperature

The influence of temperature was tested by introducing an aggregate bed in a closed environment (a Panasonic MCO-5AC-PE CO₂ incubator) with a controlled temperature (40 °C) and CO₂ concentration (15 %).

The static tests at the 2 temperatures (20 °C in the desiccator and 40 °C in the incubator) were performed with a 1–4 mm aggregate obtained by direct recycling of unused newly produced concrete in a concrete ready-mix plant (RS 1–4) and a new recycled concrete aggregate produced by crushing laboratory samples of ordinary concrete, more than 20 years old, to obtain a 12–20 mm fraction (OC 12–20). The CEM I cement proportion in this concrete was known, i.e. 353 kg/m³.

For all these tests the duration was 24 h on aggregate with initial moisture prepared at a level of 2.8 %, which corresponded to the

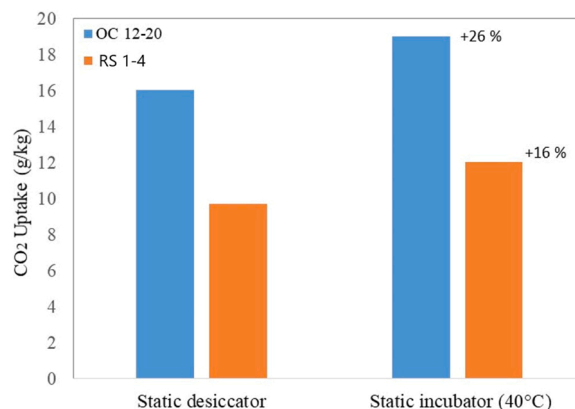


Fig. 1. Level of mass increase for static (desiccator and incubator) protocols at 24 h, for the OC 12–20 and RS 1–4, with initial moisture of 2.8 %.

optimum water content for the static test at 20 °C. The results in Fig. 1 show the comparison between the CO₂ uptake rate of OC 12–20 and RS 1–4 at 20 °C and 40 °C. Due to thermo-activation, there is an increase in the CO₂ uptake.

2.5. Fast carbonation in an agitated granular system

The results of experiments using a rolling carbonation process as a solution to accelerate the carbonation of coarse RCA have been already published [16,17]. Only the main results are summarized here.

The rotation of the drum had an attrition effect on the aggregates, which facilitated their carbonation. The rotational speed influenced the carbonation kinetics. The rotation allowed for better continuous mixing of the solid particles with the gas which allows for better performance in terms of CO₂ sequestration. Although attrition is not the main phenomenon for increasing the carbonation process but rather the mixing process, the rolling carbonation promoted abrasion between the aggregate particles by agitation and this is the major aspect that improves the efficiency of the whole carbonation process.

Note that the rolling drum that is used as one of the industrial demonstrators (Section 5) is based on this principle but with a low rotational speed. In this case, attrition is reduced and does not contribute to the carbonation process.

2.6. Fast carbonation in a multi-component gas

Multi-constituent gas containing CO₂, SO₂, and NO_x, like industrial flue gases, can adversely influence CO₂ mineralization. The experimental method developed consisted in introducing multi-component gas into an oven at controlled temperature, gas rate, and relative humidity. The gas supply was provided by a CO₂/air mixture bottle (CO₂ content of 15 %) and a CO₂/SO₂/NO₂/air mixture bottle (15 % of CO₂, 390 ppm of NO₂, and 35 ppm of SO₂) whose flow rate was regulated by a manometer. The oven was equipped with a ventilation system to ensure the mixing and homogenization of the atmosphere. Before proceeding with carbonation, the relative humidity in the incubator was regulated with ammonium nitrate salt solution (NH₄NO₃). This solution allowed maintaining the relative humidity at about 63 % before the tests and during the carbonation. The tests were performed at 21 °C. The tests were continued until complete carbonation, verified by phenolphthalein in a section of some particles.

In this experimental program, we intended to follow with phenolphthalein the evolution of carbonation front in the aggregate. As a consequence, RCA with a size of about 40 mm was chosen. 4 different sources were tested. The first is an industrial recycled aggregate containing about 60 % of non-cementitious impurities. The other 3 RCA were produced from laboratory samples of concrete, conserved in water for 5 months, then crushed for the carbonation tests. These concretes were produced with CEM I, CEM II, and CEM III cement respectively, in a proportion of 350 kg/m³ (F1, F2, and F3). Siliceous sand and coarse aggregate were used in these mix-designs.

The chemical composition of the samples was tested before and after carbonation by TGA, at several duration of carbonation (between 0 and 1, 2, ..., 8 days). The liberated CO₂ was determined on the TGA measures, by the difference of mass between 550 and 850 °C. Attention was also made to distinguish between the precipitation of Aragonite, Vaterite, and Calcite, by measuring the mass loss at intermediate temperatures. So, the absorbed gas was determined by direct chemical analysis and not by weighing the samples.

The absorbed CO₂ during total carbonation in the different aggregates is given in Table 2. It should be noted that laboratory-produced aggregates were fixed with siliceous original aggregates. They were only slightly naturally carbonated and were composed of 100 % of homogeneous mix-designed concrete. The CO₂ uptake was in line with the expected trends. The F1 and F2 aggregate CO₂ uptake were rather similar, while the aggregate F3, produced with a CEM III cement, absorbed about half of the quantity. At this stage, one can observe that the multi-component gas reduced the carbon mineralization capacity by about one-quarter. This was observed for all three aggregates produced in the laboratory. Concerning the crystal structure of the CaCO₃, the Vaterite, Aragonite, and Calcite were produced in significantly similar proportions, excepting the F3 aggregate, in which the Calcite was poorly precipitated due to the large quantity of slag in the cement. These tendencies were only slightly changed by the multi-component gas.

The identification of gases adsorbed during carbonation was also performed by TGA coupled with mass spectroscopy for F1 and F3. The summary results of the analysis of the amount of adsorbed CO₂, NO₂, and SO₂ gases are presented in Table 3.

According to the results of our analyzes, NO₂ and SO₂ gas were well adsorbed and/or precipitated on amorphous CaCO₃. Compared to the NO₂ concentration in the multi-component gas, a significant amount of NO₂ was trapped during the precipitation of calcium carbonates. This could explain a lower amount of CO₂ trapped in the multi-constituent case than in the CO₂/air case. This phenomenon was similar in the case of F1 and F3. These results could be explained by the fact that, on the one hand, the dissolution of CO₂ decreases in the presence of NO₂ and SO₂, and, on the other hand, the competitiveness between these three gases is important, which favor the NO₂ and SO₂ adsorption despite their low concentrations.

Table 2
Estimation of the absorbed CO₂ during carbonation test (after 8 days).

| | F1 | F2 | F3 |
|-----------------------------|------|------|------|
| CO ₂ /air (g/kg) | 56.1 | 62.3 | 34.1 |
| multi-component gas (g/kg) | 36.7 | 47.2 | 26.7 |

Table 3Results of the quantity of gases CO₂, NO₂ et SO₂ for F1 et F3.

| | F1 | | F3 | |
|------------------|---------------------|--------------------------|---------------------|--------------------------|
| | Multi-component (%) | CO ₂ /air (%) | Multi-component (%) | CO ₂ /air (%) |
| H ₂ O | 27.6 | 20.2 | 19.5 | 21.0 |
| CO ₂ | 66.0 | 79.5 | 66.4 | 67.6 |
| NO ₂ | 6.1 | 0.1 | 2.7 | 0.1 |
| SO ₂ | 0.4 | 0.2 | 0.3 | 0.4 |

2.7. Perspectives

In addition to the obtained results, several ideas remain unexplored:

- Due to the problem of the representativeness of the samples for the TGA tests, which leads to high dispersion in the results, cross-trials are underway to define the operating modes of some methods able to evaluate the CaCO₃ formed and the rate of CO₂ absorbed. This is particularly important for the valorization of the stored CO₂.
- The effect of the temperature and multi-component gas is more deeply studied. A "semi-industrial" pilot was recently developed which allows the gas to be sent inside a reactor to carbonate a significant quantity of RCAs. The idea of the reactor is initially based on the principle of an "Aggregate Filter", which is already well known in the industry. The aim is also to use multi-component gas to be fed into the reactor.

3. Modeling of carbonation

The modeling of carbonation provides a degree of understanding of the phenomena involved in the CO₂ uptake of RCA. Such investigation is complex because it uses parameters that are sometimes difficult to measure experimentally because of the small size of the particles, such as the spatial distribution of the carbonates formed. Two phenomena are investigated: the atmospheric carbonation of concrete structures during their lifetime and the accelerated carbonation of RCA.

3.1. Modeling of natural carbonation

Reinforced concrete structures carbonate in contact with the ambient air throughout their service life and it has to be taken into account in the emissions related to the cement industry [18,19]. To assess the CO₂ uptake resulting from this carbonation, two types of models were used [20]. The first is physicochemical models based on mass conservation equations of the main carbonation reactants [21–26]. Solving this equation system is done numerically. The second type of model corresponds to analytical models based on a square root of time evolution of the carbonation depth. The formulation of this kind of model is based on the hypothesis that carbonation is controlled by the gaseous diffusion of gaseous CO₂ while the chemical reactions are assumed to be instantaneous. The main model used in the project is that of the European Standard EN 16757 [27].

A comparison of the two types of models was carried out in a test case, namely the 1D carbonation of a 20 cm CEMI cement concrete wall at 20 °C and 80 % RH. The results of the standard model and the simulations of the numerical model (SDReaM-crete model of CERIB [20,28]) were compared for a 100-year lifetime. Even if differences are observed in the carbonation depths, the two models provide similar results as to the amount of CO₂ fixed (Fig. 2). About 2 kg CO₂ per m² of wall are predicted by the two models, which corresponds to about 8 % of the CO₂ emitted by the calcination of raw materials during the manufacture of the cement used in the wall. This satisfactory comparison makes it possible to support the use of an empirical model such as that of the standard, at least for concrete made with CEMI cement.

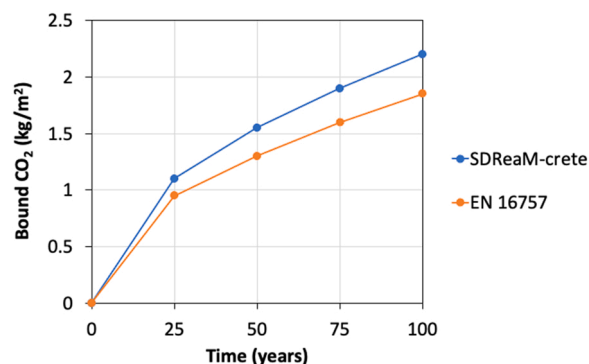


Fig. 2. Amount of bound CO₂ per square meter of concrete exposed to atmospheric carbonation in the case of a 20-cm concrete wall [20].

The standard model [26] was also used to assess the CO₂ sequestration by an existing building, i.e., the Aziyadé student residence located in La Rochelle. This building has the particularity of being completely raw stripping with a high concrete surface in contact with the ambient air. By considering all the environments to which the surfaces are subjected, the calculation results in a total of 12 % of the CO₂ emitted from calcination being reabsorbed by the concrete after 50 years. This case study is currently being continued with the use of numerical models.

Our work on atmospheric carbonation shows that concrete structures constitute efficient carbon sinks, which confirms most of the recent literature, e.g. Xi et al. [29]. However, during their theoretical service life, i.e., 50 years, a high fraction of the concrete remains non-carbonated. The uptake of CO₂ by carbonation of RCA, therefore, makes perfect sense. In addition, these studies provide input for the databases required for the LCA of concrete structures.

3.2. modeling of accelerated carbonation

The uptake of CO₂ by RCA was studied using the different physicochemical models [21–25] with the main purpose of studying the influence of parameters controlling carbonation [30]. Beforehand, the models were compared during a benchmark relating to the 1-year accelerated carbonation of a cement paste. The experimental results came from Bertin [31]. The numerical simulations were rather satisfactory since the profiles obtained by the different models were close to the experimental data. However, the differences in the design of the models are significant, as in the case of the CO₂ diffusion coefficient for example. Rather than selecting a particular model, we have chosen to continue the work with the different models. We consider that this diversity of models could be representative of the diversity of behavior of RCA (which are usually mixtures of crushed concretes from different origins).

The parametric study was therefore carried out with four numerical models to simulate the CO₂ uptake of a RCA of spherical geometry and with homogeneous properties. The carbonation conditions for the simulations were the following: CO₂ concentration of 15 % (as used for the experiments of the FastCarb project – see Section 2), temperature of 20 °C, and relative humidity equal to the equilibrium humidity with the initial water content of the RCA. The parameters that were studied are the porosity, the content of carbonatable elements, the degree of water saturation, and the grain diameter. In total, around a hundred simulations were carried out for a carbonation period of 3 days, which made it possible to map the influence of the parameters, as shown for instance in Fig. 3.

Analysis of these numerical experiments shows that, as expected, the CO₂ binding capacity of RCA increases with the content of the carbonatable element and, to a lesser extent, with porosity. The water saturation degree and the diameter are two parameters with coupled effects, which results from the competition between the diffusion of gaseous CO₂ and the chemical reactions. For low water saturation degrees (less than 0.6), the uptake of CO₂ is controlled by chemical reactions: the influence of grain size is negligible. For higher saturation degrees, conversely, the rate of uptake becomes controlled by gas diffusion and the diameter has a major effect on the uptake. Fig. 4 highlights the competition between diffusion and reaction with the profiles of the content of CO₂ bound in the grain. At a low degree of saturation (0.3), carbonation is homogeneous, while at a high degree (0.9) a carbonation front is observed (Fig. 4).

3.3. Perspectives

In the case of accelerated carbonation of RCA, theoretical studies have started to understand the influence of the gas temperature used for the treatment of RCA. The gas captured from a cement plant is indeed very hot (60–80 °C) according to the field experience of the FastCarb project scale 1 demonstrators (see Section 5). The effect of temperature on carbonation rate is not very well described in the literature and should be considered.

Alongside the experiments conducted in the project on concrete made with the carbonated RCA, a modeling study is planned to estimate the durability of reinforced structures containing various proportions of carbonated RA, concerning two phenomena that attack reinforcement: carbonation (XC) and chloride transfer (XS). This study is planned for the first part of 2022.

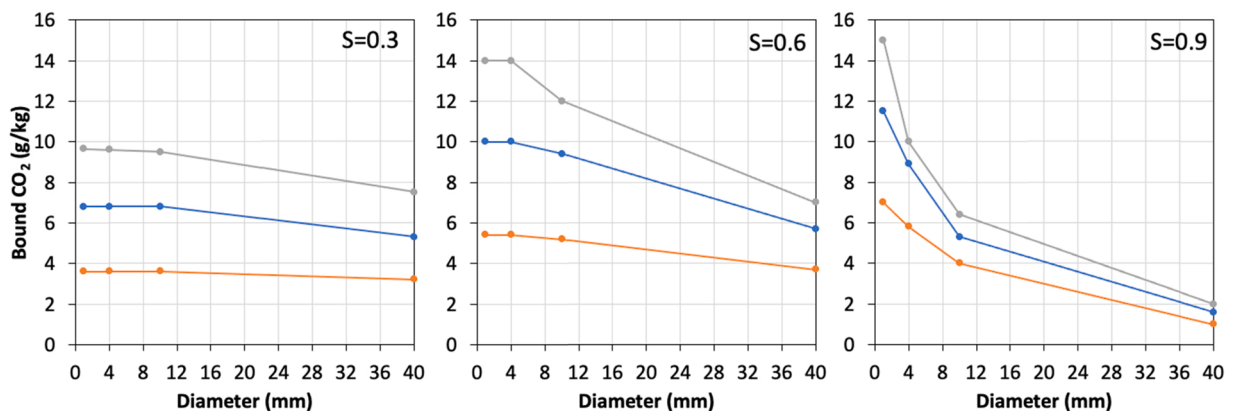


Fig. 3. Mapping from the parametrical study (for a porosity of 20 %): bound CO₂ in g per kg of RCA versus grain diameter, for 3 water saturation degrees (S) and 3 portlandite contents (orange: 500, blue: 1000, grey: 1500 mol/m³).

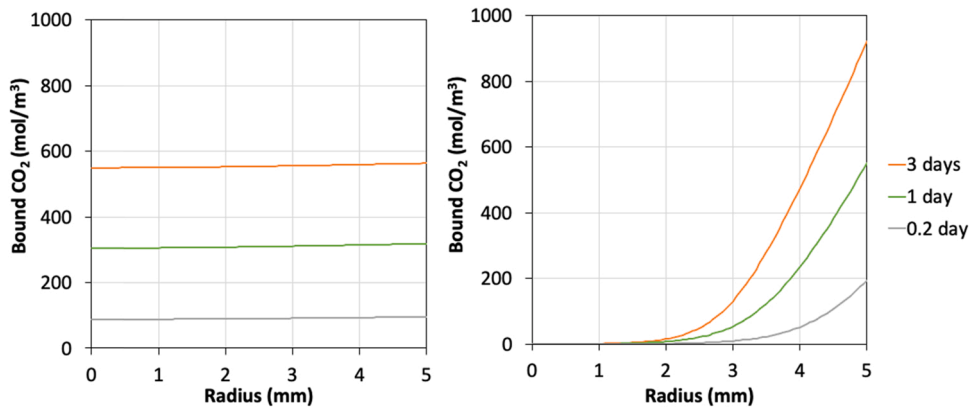


Fig. 4. Calculated profiles of bound CO₂ after 3 times of accelerated carbonation of a 10 mm diameter RCA with an initial water saturation degree of 0.3 (left) and 0.9 (right).

4. Bio-carbonation

Bioprecipitation of calcium carbonate (CaCO₃) offers an innovative and ecological way to treat cementitious materials [32], lengthening their lifespan by sealing fissures [33], protecting them from chloride ions penetration, and enhancing their appearance in the case of concrete structures [34]. Bacteria are one of the most studied microorganisms capable of performing bioprecipitation through many mechanisms [35]. In the FastCarb project, bio-carbonation is an alternative method for mineralizing atmospheric CO₂ on RCA. The objective is to form a gangue of CaCO₃ around the aggregates in an accelerated way by using bacteria to decrease the porosity and improve the mechanical properties of RCA.

In a first study [36] CaCO₃ was formed on the surface of RCA but not homogeneously. The challenges are therefore to identify a microbial strain capable of:

1. Growing at alkaline pH and forming a biofilm.
2. Producing CaCO₃.
3. Form a biofilm and produce CaCO₃ on the surface of cementitious materials.

To respond to these challenges, micro-organisms were isolated from RCA that had been stored outdoors for several years at the Nantes site of the Eiffel University. About twenty strains were cultivated. These strains were then tested to ensure their ability to grow at pH 11 (pH close to the surface of the aggregates); then to form a biofilm (an important characteristic so that the bacteria colonize the surface of the aggregates homogeneously and the product of their activity adheres to the aggregates).

At the end of these tests, a strain was selected as a candidate for precipitating calcium carbonates and for analyzing its development on the surface of cementitious materials. Then, the ability of the strain to produce CaCO₃ on agar was tested. For this purpose, 275 μL bacteria culture aged 72 h was diluted to the thousandth and plated with 12.5 mL of calcium acetate (350 g L⁻¹) on nutrient agar at pH



Fig. 5. Surface of a 13 cm Petri dish containing nutrient agar at pH 11 inoculated with B.H. and calcium acetate for 15 days.

11. After 2 weeks of incubation, CaCO_3 formations are visible on the agar (Fig. 5).

Different parameters (incubation time, microorganism concentration, and calcium acetate concentration) were tested. The production of CaCO_3 could be confirmed by performing TGA (Thermogravimetric Analysis), XRD (X-ray diffraction), and micro-Raman analyses. This showed that B.H. formed CaCO_3 in the form of vaterite and calcite. To obtain an optimal yield, it is necessary to have the entire surface of the petri dish homogeneously covered by a microbial mat and to have a calcium acetate concentration of 13 mg cm^{-2} .

The next step was to observe whether B.H. was able to form a homogeneous biofilm on the surface of mortar disks. Thus, mortar disks were immersed in nutrient broth for 4 h and then 3 mL of B.H. suspension was added to the surface. The surface of the samples was observed after 2 weeks. The mortar samples inoculated with B.H. were able to retain moisture on their entire surface, which revealed the presence of a homogeneous biofilm (Fig. 6).

After biofilm formation, calcium acetate is added to form calcium carbonates. This phase is currently being optimized and will be followed by a run on RCA.

5. Industrial demonstrators

The main objective of FastCarb is to demonstrate the feasibility of fast carbonation of RCA by contact between the recycled material and emission gases from a cement plant. These gases have a CO_2 content of approximately 15 % in volume and a high temperature (around 70°C), these parameters will be used to accelerate the carbonation process.

Industrial or semi-industrial demonstrators are developed to produce several metric tons of recarbonated aggregates of each granulometry (sand and gravel) with the perspective to characterize them as recarbonated materials (gap of porosity, rate of fixed calcium carbonate -sequestration) as well as aggregates in concrete and measure the benefit of recarbonation process on concrete properties (fresh and hardened).

The first principle was to avoid making specific development of dedicated equipment for carbonation but to divert existing facilities from their original use and turn them into carbonators. The second principle is the simplicity of facilities to be incorporated into the cement production process.

This is what has been realized considering 2 dynamic systems, a rolling drum dryer, and a fluidized bed dryer. Two static systems (a simple container and a gravel bed filter) were also tested but are not presented here. The two dynamic systems were connected to exhaust gases from full-scale cement works circulating in counter flow.

The rotating drum constituting the Fastcarb pilot located in the Vicat plant in Créchy is 11 m long and has a radius of 2 m (Fig. 7). The rotary movement associated with the lifters welded to the tube linings allows the surface of the aggregates to be brought into contact with the gas. The gas feeding the drum has a temperature between 40 and 60°C and a CO_2 content varying from 11 % to 16 %. The relative humidity in the drum is 90 %. To increase the residence times in the tube, it was decided to rotate the tube intermittently at 4 rpm for 2 min every 15 min. The optimal residence time was identified as 60 min for 3 t of GBR. Considering the filling and emptying time, this system carbonates three tons of aggregate in two hours. The amount of the investment is estimated to 150,000€.

The fluidized bed dryer is installed on the site of the Val d'Azergues cement plant (Holcim). The first phase of tests allowed to demonstrate the industrial feasibility of accelerated carbonation of recycled aggregates by using directly the gases of the cement plant (CO_2 content of about 20 %). During this first phase, it was not possible to work in a continuous process because the flow of materials on the bed was insufficient. Several factors can explain this difficulty: high humidity in the carbonator chamber and insufficient inclination of the bed. The first tests were therefore conducted in semi-continuous mode. If this defect is eliminated, a treatment of 10 tons per hour is expected. The amount of the investment is estimated to 180,000€.

Two batches of 40 t each of each size fraction were produced by the dynamic systems. These carbonated RCA have been used to produce concretes (see Section 6). Carbon uptake as neoformed calcium carbonate is in the range of 20 kg/t for coarse RCA to 40 kg/t for the sand fraction of RCA depending also on initial carbonation due to storage of variable duration. Fig. 7 shows the two industrial dynamic systems.

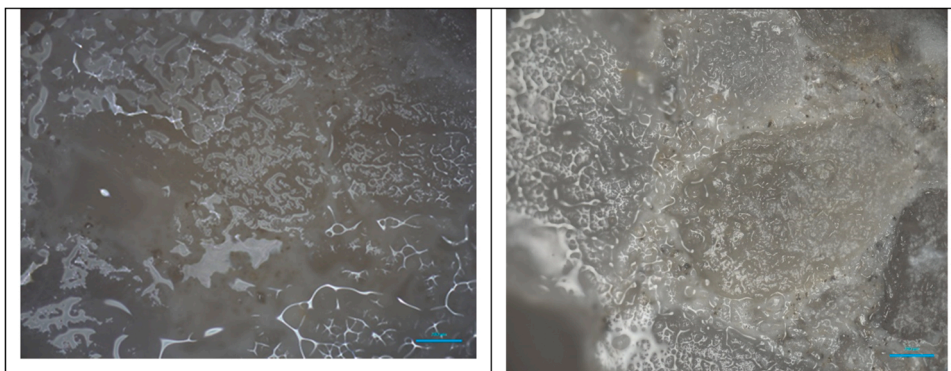


Fig. 6. Optical microscope photograph after 2 weeks of incubation of the surface of the samples.



Fig. 7. The two dynamic systems. On the left, the rolling drum carbonator (Créchy cement works) and, on the right, the fluidized bed carbonator (Val d'Azergues cement works).

6. Properties of concretes made with carbonated RCA

This part deals with the properties of concrete containing carbonated RCA and applications to precast elements and structures that are cast in situ. Concretes are manufactured with RCA that has been carbonated with the two dynamic systems (see Section 5). The experimental survey allows measuring the impact of the CO₂ treatment on the density of the aggregates and their contribution to the mechanical performance of the concrete in which they are recycled to identify any impact on the optimization of mix design. The contribution of the CO₂ treatment to the durability properties is also studied, in particular towards the risk of corrosion in reinforced concrete.

6.1. Mix design

Two families of concrete (C25 and C45) are studied with 7 different compositions of aggregates (Table 4). To simplify the terminology, natural aggregates are named NS (Natural Sand) and NG (Natural Gravel), RCAs are named RS (Recycled Sand) and RG (Recycled Gravel), and carbonated RCAs are named CRS1 and CRS2 (Carbonated Recycled Sand), CRG1 and CRG2 (Carbonated Recycled Gravel), respectively for cement plant (1 – Val d'Azergues, fluidized bed carbonator) and (2 – Créchy, rolling drum carbonator).

The concretes met the requirements of exposure classes XC4/XF1 (table NA.F.1) of NF EN 206/CN standard [37]. Except for aggregates, the 14 concretes are similar in terms of other constituents: for each family (C25 or C45), the same cement type CEM II/A, the same amount of cement (320 kg for the C25, 450 kg for the C45), water to cement ratio (0.55 for the C25, 0.37 for the C45), admixture, and mixing water (177 l for the C25 and 165 l for the C45). The consistencies targeted are the same for all the concretes: S4. Note that the aggregates were saturated before mixing.

6.2. Properties

6.2.1. Rheology

Within the framework of these tests, it was observed difficult management of formulation due to a certain heterogeneity within the same batch of aggregates, but concretes with carbonated RCA showed an easier behavior to manage in terms of rheology than non-carbonated RCA (Fig. 8).

Table 4

Substitution rates (in mass) of the 14 concretes of the study.

| | | Sand | | | | Gravel | | | |
|--------|-----------------------|--------|--------|----------|----------|--------|--------|----------|----------|
| | | NS (%) | RS (%) | CRS1 (%) | CRS2 (%) | NG (%) | RG (%) | CRG1 (%) | CRG2 (%) |
| C25/30 | B1_C25_100NS-100NG | 100 | | | | 100 | | | |
| | B2_C25_20RS-50RG | 80 | 20 | | | 50 | 50 | | |
| | B3_C25_20CRS1-50CRG1 | 80 | | 20 | | 50 | | 50 | |
| | B4_C25_20CRS2-50CRG2 | 80 | | | 20 | 50 | | | 50 |
| | B5_C25_40RS-100RG | 60 | 40 | | | | 100 | | |
| | B6_C25_40CRS1-100CRG2 | 60 | | 40 | | | | 100 | |
| | B7_C25_40CRS2-100CRG2 | 60 | | | 40 | | | | 100 |
| C45/50 | B8_C45_100NS-100NG | 100 | | | | 100 | | | |
| | B9_C45_100NS-50RG | 100 | | | | 50 | 50 | | |
| | B10_C45_100NS-50CRG1 | 100 | | | | 50 | | 50 | |
| | B11_C45_100NS-50CRG2 | 100 | | | | 50 | | | 50 |
| | B12_C45_100NS-100RG | 100 | | | | | 100 | | |
| | B13_C45_100NS-100CRG1 | 100 | | | | | | 100 | |
| | B14_C45_100NS-100CRG2 | 100 | | | | | | | 100 |

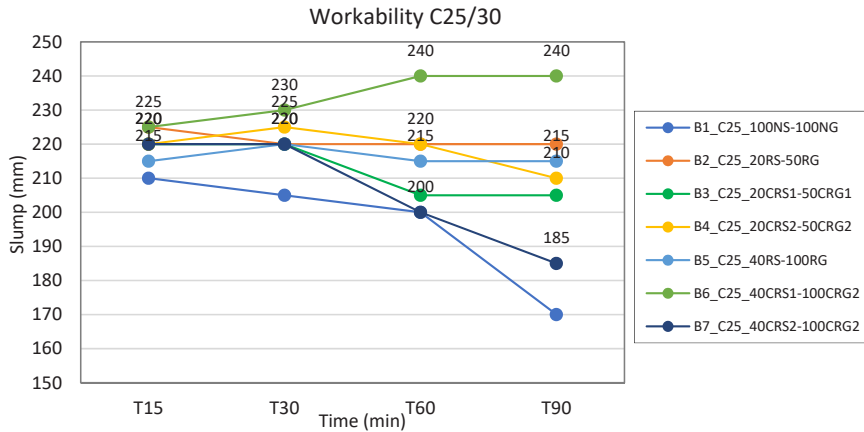


Fig. 8. Rheological behavior of the C25/30 concretes.

Greater loss of workability was observed for C45/50 class concretes, which are more sensitive to water variations and thus to the variability of water absorption of recycled aggregates, carbonated or not.

6.2.2. Mechanical characteristics

The results in Fig. 9 show, for C25 concretes, that, for a given 28-day compressive strength class, concrete with carbonated recycled aggregates differs little from concrete with recycled aggregates on the majority of compressive results. In line with the future EC2 [4], for low substitution rates (up to 25 %), the effects of introducing carbonated or non-carbonated recycled aggregates on concrete properties are minor. The results are similar for the C45 family.

Before the tests, an improvement in the mechanical properties was expected. Investigations are conducted to understand why this is not the case. A possible explanation is an effect of mixing during the production of concrete. Natural carbonation of recycled aggregates is another possibility.

6.2.3. Durability

The key durability indicators of water-accessible porosity, chloride ion migration, gas permeability, and accelerated carbonation rate show no significant negative or positive impacts associated with the use of carbonated RCA compared to non-carbonated RCA. The

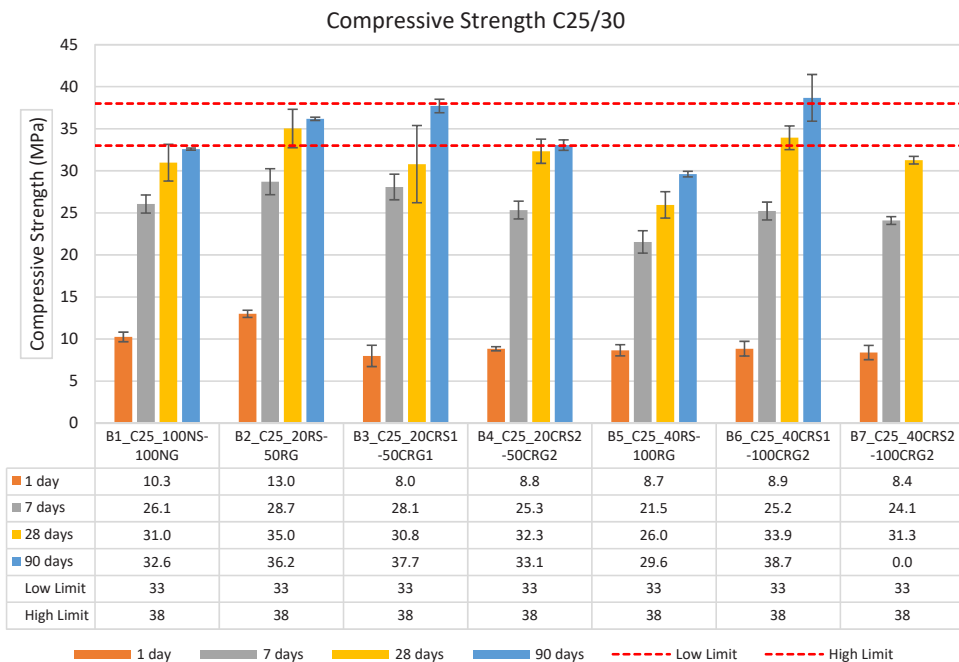


Fig. 9. Compressive strength of the C25/30 concretes.

variations from natural aggregate concrete are consistent with the findings in the literature.

6.2.4. Production of elements

Real structures or precast elements are or will be also realized:

- 3 walls made with 3 of the concretes of Table 4 (B2_C25_20RS-50RG, B3_C25_20CRS1-50CRG1, and B4_C25_20CRS2-50CRG2) have been cast. The three walls have been cored for sampling test bodies falling within the durability experiment plan. The first results show that, due to the placing of concrete, the porosity of the cores is larger than the one of the laboratory samples but without a notable effect of the use of recycled aggregates, carbonated or not.
- Three different precast products (blocks, stairs, borders) are studied for three different formulas: a "classic" concrete with natural aggregates usually used by the plant to elaborate the product and two concretes with carbonated recycled aggregates. Each of the products will be tested according to its standard. The stairs will be tested for durability as well, linked to the risk of reinforcement corrosion.

7. Environmental and economic analysis

7.1. Life cycle analysis

The FastCarb research project assesses the possible benefits of mineral carbonation, in terms of both concrete aggregates' recovery and CO₂ uptake. The present research aims to evaluate the environmental impacts of accelerated carbonation processes, based on a life cycle approach [38]. The different mineral carbonation processes are currently under study. These processes are prototype ones. The objective is to be able to find the best industrial process from the environmental point of view.

The data specific to the mineral carbonation process has been collected with the industrial partners involved in the project. The functional unit chosen is 1 ton of carbonated recycled concrete aggregates supplied to the construction site. The boundaries of the considered system are presented in Fig. 10.

The conclusions of this preliminary study are the fact that:

- The carbonation of the 4–16 mm fraction is of little interest since the quantity of CO₂ bounded is low, and of the same order of magnitude as the CO₂ emitted during the carbonation process.
- The carbonation of the 0–4 mm fraction is more interesting in terms of CO₂ uptake.
- Some methodological questions arise as perspectives to perform:
- How to account for the CO₂ captured from industrial gas?
- To whom can a CO₂ reduction be attributed: the emitter (industrial site - cement plant) or the user (material sector)?
- How should the prototype effect be considered and what are the differences with an industrial system?

7.2. Economic viability of the accelerated carbonation process

A cost/benefit analysis has been performed to evaluate the economic viability of the accelerated carbonation processes. The

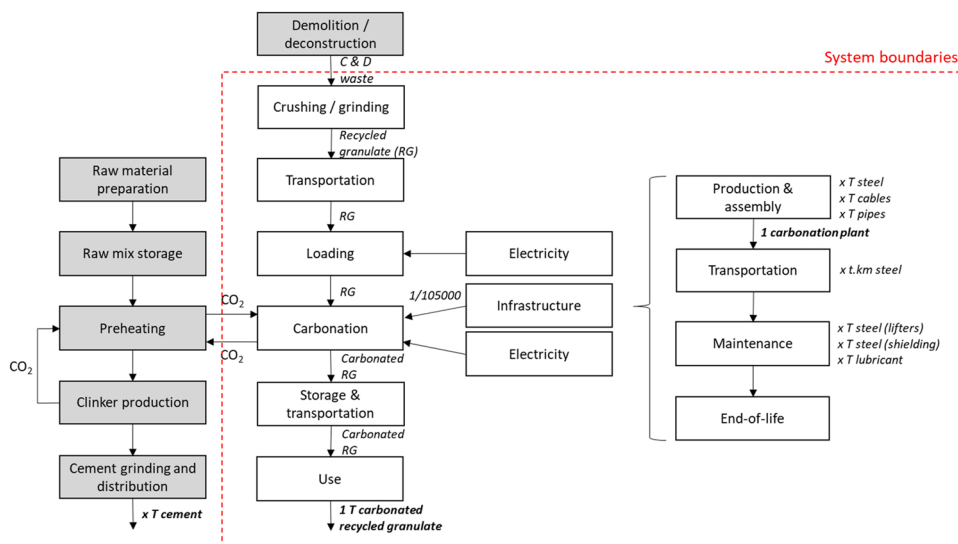


Fig. 10. Boundaries of the considered system for LCA.

industrial demonstrators proposed by Vicat and LafargeHolcim were tested. Both companies did not develop specific equipment but converted existing industrial equipment into a carbonator. These processes would be considered sustainable only if they were environmentally acceptable and economically viable.

The analysis is required to compare the production costs and the benefits associated with the accelerated carbonation process to the costs of more traditional solutions. Four scenarios were developed and examined:

- Scenario A: use of natural aggregates.
- Scenario B: use of recycled aggregates for roads.
- Scenario C: use of recycled aggregates for building use.
- Scenario D: use of recycled aggregates after accelerated carbonation.

Fig. 11 illustrates the different scenarios.

The analysis of these scenarios involves formulating hypotheses about the distances between quarries/recycling platforms and concrete manufacturing sites. A maximum distance of 30 km was retained for natural aggregate while the minimum and maximum distances between the recycling platform and the concrete manufacturing site were 5 and 25 km respectively. However, there was a need to consider upstream and inbound flows. In half of the cases, the trucks are empty when they reach the platform. Consequently, minimum, and maximum distances covered by trucks were 7.5 and 37.5 km.

Scenario D requires integrating a hypothesis concerning the distance between the carbonation site (the cement plant in the case of the demonstrators) and the construction site. Since the number of cement plants located next to large urban areas remains limited, we assumed that the carbonation process could also take place in any plant generating CO₂ emissions and being able to capture CO₂. We assumed that the distance covered a spectrum going from 5 to 50 km.

Construction, maintenance, and operation costs of the demonstrator were compiled and the cost of one ton of carbonated recycled aggregates was calculated by playing on two variables:

1. The distance traveled by the trucks that load and unload the aggregates during their transformation (crushing, recycling, carbonation).
2. The cost of transport would be in the range of 1–2 euros for a ton for 10 km (i.e. between 2.50 € and 5 € for a truck of 25 tons traveling 1 km).

The cost of natural aggregates upon leaving the quarry was 13 euros. The costs of recycled aggregates for road and building were respectively 11 and 13 euros. The application of these hypotheses to the demonstrator developed by Vicat indicated that carbonated recycled aggregates suffer from a competitive disadvantage compared to traditional solutions (Table 5). This is due to transport costs and the additional industrial process. When transport distances are maximal, the gap is huge. It shows that this industrial process would be more competitive around large urban areas where the potential for recovering aggregates from deconstruction sites is high and construction/renovation projects are abundant.

Then the economic viability of the industrial carbonation process was examined by considering the environmental assessment based on the Life Cycle Assessment method that quantifies environmental impacts (cf. Section 7.1). Consequently, CO₂ emitted and bounded during the process and the transport of aggregates were counted. Each ton of CO₂ was priced according to the evolution of the carbon market.

For the coarse fraction, calculations (Fig. 12) indicated that the carbon emitted during the industrial process was higher than carbon gained from carbonation. Consequently, the competitive disadvantage was even worse. For the sand fraction, the gains related to carbon capture were higher than the carbon emitted during the process. These gains and the evolution of the price per tonne of carbon would reduce the price of carbonated recycled aggregates (sand fraction).

This analysis showed that transport is the predominant variable in the economic viability of carbonated recycled aggregates. This is in line with the conclusions of Dias et al. [39] in the case of recycled aggregates in Portugal and Braymand et al. [40] who showed that the distance between cities, natural aggregate sites, and recycling platforms strongly impacted the results of the environmental analysis related to the production of recycled aggregates. The results also indicate that the implementation of a carbonation process on a deconstruction site or a recycling platform would probably increase the competitiveness of this approach. In addition, optimization of transport costs would be possible. Finally, carbon prices are likely to rise as they did in 2021. In January the price for one ton was 33 euros. It reached 64 euros at the beginning of October. In the case of their R&D projects, some industrial companies already rely internally on €100/tCO₂. This should reinforce the competitiveness of any industrial process allowing carbon capture.

8. Conclusions

The project FastCarb which will end in 2022 has already obtained some conclusions concerning the accelerated carbonation of recycled concrete aggregates. The laboratory experiment and modeling have confirmed the influence of several parameters like the water content of the RCA, the temperature, and the size of the RCA. A relation between the optimum water content and water absorption has also been obtained. And the accelerated carbonation improves the water absorption of the RCA, especially for the sand fraction.

Concerning the modeling, the influence of the main parameters such as the water content or the size of aggregates is confirmed. The order of magnitude of natural carbonation of concrete buildings is also in line with results from the literature. The influence of

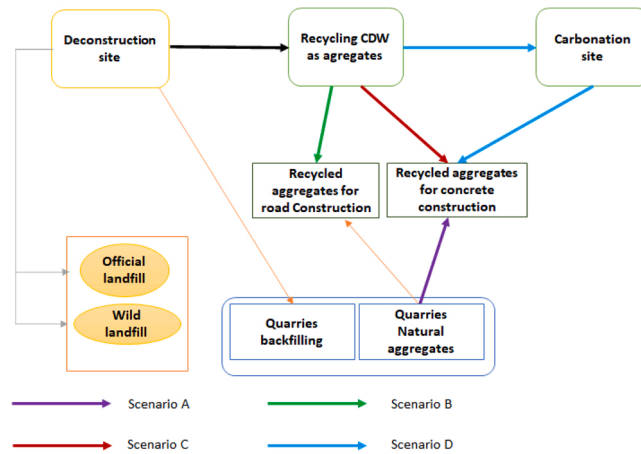


Fig. 11. Representation of the different scenarios.

Table 5
Cost analysis resulting from the four scenarios.

| | Total distance (km) | | Cost (1€/t/10 km) | | Cost (2€/t/10 km) | |
|--|---------------------|-------|-------------------|------|-------------------|------|
| | Mini | Maxi | Mini | Maxi | Mini | Maxi |
| Scenario A - Natural Aggregate | 5 | 30 | 13.5 | 16 | 14 | 19 |
| Scenario B – Recycled aggregates for road underlayment | 7.5 | 37.5 | 11.8 | 14,8 | 12.5 | 18.5 |
| Scenario C – Recycled aggregates for building | 7.5 | 37.5 | 13.8 | 16.8 | 14.5 | 20.5 |
| Scenario D – Carbonated recycled aggregates | 22.5 | 212.5 | 28.6 | 47.6 | 30.9 | 68.9 |

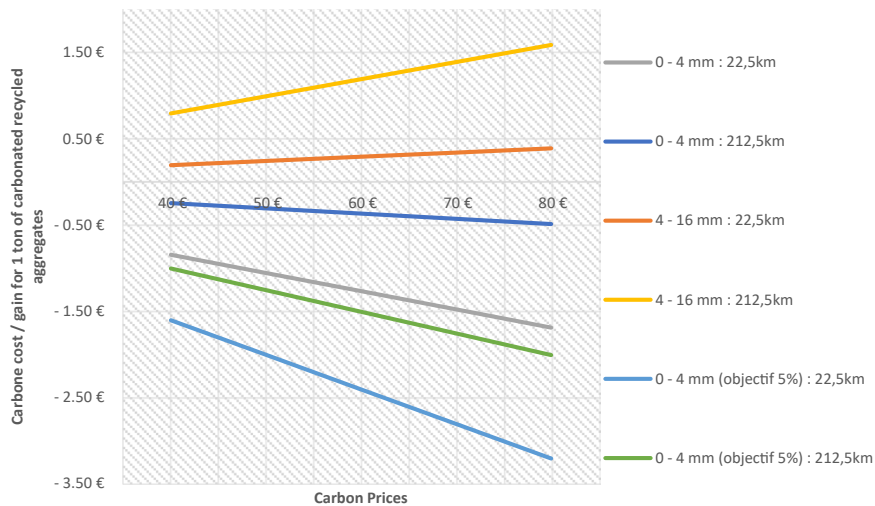


Fig. 12. Sensitivity analysis according to the evolution of carbon prices.

temperature on accelerated carbonation has still to be modeled.

On the industrial scale, demonstrators were developed. Two of these demonstrators were installed in two cement plants where it was possible to use directly exhaust gases. The quantity of stored CO₂ was 20 kg/t for the coarse RCA and 40 kg/t for the sand fraction. An important quantity of carbonated RCA has been prepared that will allow preparing twelve concretes incorporating these CRCA, real structures, or precast elements.

The LCA and the economic study on these demonstrators confirm that the sand fraction of RCA is the most interesting material for the uptake of CO₂. This is also interesting for a circular economy objective because recycled concrete sand is not easily used today in concrete made with RCA.

Finally, the Fastcarb project shows that accelerated carbonation is feasible on an industrial scale with results in line with laboratory experiments and with positive impacts when the transportation of the RCA is limited. The amount of CO₂ that could be mineralized will

not solve alone the problem of CO₂ emissions of the concrete industry but is a possible contribution that is also interesting to favoring a circular economy.

Declaration of Competing Interest

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

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