Accelerated carbonation of recycled concrete aggregates: results and perspectives of the FastCarb project

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Synopsis: The carbon footprint of concrete can be reduced by using carbonated recycled concrete aggregates (CRCA). This is the main conclusion reached by the partners of the FastCarb project after four years of work that allowed them to optimize the parameters of accelerated carbonation of RCA in the laboratory, demonstrate the industrial feasibility of the process and identify the conditions for the development of the use of CRCA. In this paper, we present the main results and the guidelines of the project, including the possible financial valuation of the process.

Keywords: concrete, recycled aggregates, carbonation, CO2 impact

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INTRODUCTION

The FastCarb National Project is a collaborative research project supported by the French government as part of the "applied research in civil engineering" network. It follows on from the Recybéton project, one of whose conclusions was to show that the porosity of Recycled Concrete Aggregates (RCA) was a limiting parameter for their use in greater proportions in certain exposure classes [1]. At the same time, the natural carbonation of concrete, which leads to the formation of calcium carbonate, is well known. Applied at an accelerated rate, this phenomenon is likely to fill the pores of RCAs and thus improve their performance.

Accelerated carbonation of RCA is attracting growing international interest, as evidenced by the most recent publications (see for example [2-5]). FastCarb project aims to identify the mechanisms and conditions that will accelerate this natural phenomenon, to increase the amount of CO_2 stored in RCAs and thus their use in new concretes. The project was funded by the French Ministry of Ecological Transition and Territorial Cohesion, and 23 academic and industrial partners, for a total budget of €3 millions. IREX (https://irex.asso.fr) provided organizational support and financial control. Gustave Eiffel University provided leadership for the project, which started at the end of 2017 and runs until 2023.

The FastCarb project is set against the backdrop of the 2015 French Law on Energy Transition for Green Growth, which targets two important points for the construction sector: the promotion of the circular economy by integrating the recycling of construction materials, with a target for the building and public works sector to recover 70% of its wastes by 2030, and the reduction of greenhouse gas emissions by promoting low-carbon buildings over their entire lifecycle. Since then, this law has been supplemented by the Réglementation Environnementale 2020 (2020 Environmental Regulations), whose decrees impose binding thresholds on a building's CO_2 emissions over its entire lifecycle. These regulations encourage construction companies to reduce the carbon footprint of concrete, and the use of RCA as a carbon sink is one of the possible levers... Finally, the decree on the extended responsibility of producers of construction products and materials aims to organize and finance the collection and recycling of construction materials and will encourage the reuse of RCA in noble applications.

These legislative and regulatory developments demonstrate the relevance of the FastCarb project and the importance of understanding and optimizing the carbonation mechanisms of RCA, both in terms of capturing greenhouse gases and developing the use of RCA in new concretes and the development of the circular economy in the construction sector. The project work involved 3 main phases: a laboratory research and modelling phase, an RCA carbonation phase in industrial demonstrators, and a concrete formulation and manufacturing phase incorporating recarbonated RCAs for the production of concrete elements. Analyses of the environmental balance and economics of the carbonated RCA (CRCA) production chain were also carried out. This document presents a summary of the results obtained during these various phases and the resulting recommendations.

SUMMARY OF PROJECT RECOMMENDATIONS

Characterization of CRCA: Method for determining stored CO2

To enhance the value of the accelerated carbonation process, it is extremely important to be able to measure the quantity of CO_2 stored in the RCA. The aim is therefore to propose quantitative, representative and reproducible methods for accurately assessing the stored CO_2. During the project, five methods were evaluated.

Thermogravimetric analysis (TGA) is a reference measurement, most often carried out in laboratories, and is complex to implement. The accuracy of this method (repeatability and reproducibility data) is lower than that of the other methods evaluated, no doubt due to the small number of materials tested.

Loss-on-ignition measurement, adapted from cement standard EN 196-2:2013, requires an oven capable of temperatures above 950°C but is simple to implement. The repeatability and reproducibility of this method are satisfactory. With a test duration of 12 hours, this method can be used to determine the carbonate content, before and after carbonation, of well-sampled batches of RCA.

The Bernard calcimeter and Carbonate Bomb measure the amount of CO_2 released by the sample during its reaction with hydrochloric acid. The equipment needed to perform these measurements is simple to use, and the test duration is short. While the carbonate bomb test is simple to perform, the Bernard calcimeter test is a little more complex. The repeatability of both methods is average. Reproducibility is good for the Bernard calcimeter and even better for the carbonate bomb. These methods are well suited to production control, to correct any drift in carbonation process settings, for example.

The measurement of Total Carbon Content is based on the analysis of an infrared signal during combustion at 1400°C under oxygen scavenging. While the measurement is simple and very fast, it requires investment in very specific equipment. Its repeatability is good, but its reproducibility could not be validated during the project, as only two laboratories used it. Further evaluation of this method is therefore required.

Depending on the context in which it is necessary to determine stored CO_2, the use of certain methods is to be preferred. For example, while ATG can be used when changing deposits, loss on ignition is well suited to qualifying average batches before and after carbonation. As for calcimetry (Bernard or carbonate bomb), it can be used for quality control between 2 loss-on-ignition measurements.

Parameters favouring accelerated carbonation of CRCAs

Optimization of CO_2 mineralization in recycled concrete aggregates depends on numerous parameters listed below and taken from converging results of laboratory studies (part of which is published in [6]) and modelling:

- the *type of cement* in the original concrete influences CO_2 storage potential. Recycled aggregates containing CEM III can store up to 3 times less than those containing CEM I.
- the *initial natural carbonation* state of the aggregates affects CO_2 storage potential. Spraying with a coloured indicator such as phenolphthalein provides a qualitative check on whether carbonation is still possible.
- *water content* is the predominant factor. The optimum water content in recycled aggregates for maximum carbonation lies within a range of 65% to 85% of the 24-hour water absorption value of RCA.
- *aggregate size* has a significant impact on CO_2 storage rates. The storage rate of the sand fraction can be double that of 12-20 mm gravel.
- *higher temperatures* (40°C and above) have a favourable effect on carbonation. The gain from partial pressure of over 15% CO_2 in the gas is less significant. For these two reasons, cement kiln exhaust gases (with a CO_2 content of around 15% and a temperature between 70 and 90°C) are suitable for accelerated carbonation.

- the presence of *other combustion products* in the gas (SO_2, NO_2) can, however, significantly disrupt CO_2 mineralization kinetics.
- *granular agitation* is more efficient than carbonation in a fixed granular bed. Nevertheless, granular agitation also produces attrition, with desirable or undesirable effects on the aggregate (creation of carbonate fines, favourable for CO_2 storage, reduction in aggregate angularity).
- the effect of *forced gas flow* in the aggregate bed improves carbonation kinetics. Gas pressure also plays a significant role in increasing the storage rate. However, if the pressure gradient is too high (> 1.5 bar), a condensation phase is observed, limiting CO_2 penetration.
- a *longer treatment* logically increases the amount of CO_2 stored. However, this effect is limited by the filling of porosity and increased saturation of the medium, which slow down carbonation kinetics.

Industrial accelerated carbonation processes

Several industrial demonstrators (rotary dryer and fluidized bed in cement plants, container) and a pilot plant (gravel filter in a laboratory) have been developed. These experiments have confirmed that it is entirely feasible to implement industrial processes for accelerated carbonation of recycled concrete aggregates at CO_2-emitting sites [7, 8].
The main lessons learned from these industrial trials are as follows:

- The use of *cement plant gases* with a CO_2 content of around 15% (by vol.) is fully satisfactory for accelerating artificial carbonation, with an optimum relative humidity of between 50 and 70%, and a temperature of between 70 and 90°C.
- *Fine fractions* of recycled concrete aggregates should be processed within a relatively short time (within 3 months if possible) of production and availability, to limit natural atmospheric carbonation, which limits the potential for accelerated carbonation.
- For industrially carbonated recycled concrete aggregates, *capture rates* of between 31 and 39 kg CO_2/t for the recycled sand and between 5 and 12 kg CO_2/t for the recycled gravel have been measured on average for typically 1 hour of treatment (contact with gas) in cement plants.
- In demonstrators, which are based on existing plants, mastery of the triptych "*CO_2 concentration, water content and gas temperature*" is crucial to maximizing CO_2 capture rates.
- In the case of gravel filters, CO_2 capture rates depend on the same parameters as for industrial plants, namely the above-mentioned triptych. The effects of the combination of the main gas (N_2) and the secondary gases (SO_2 and NOx) do not lead to the precipitation of specific compounds, but can nevertheless adversely affect the effective carbonation rate.

With a view to industrializing processes, it will be necessary to optimize control of key parameters such as relative humidity, temperature and treatment time for equipment, as well as the water content of recycled aggregates. The site will also need a reliable method for determining CO_2 capture rates, to be able to consider these values in aggregate EPD calculations, as well as to economically valorize the CO_2 not emitted.

Production of concrete containing CRCA - Specifications

The fresh and hardened properties of concrete made with carbonated recycled aggregates from the demonstrators of the FastCarb national project are comparable to those of concrete made with non-carbonated recycled aggregates [9]. The impact of their use is therefore unlikely to alter the observed trends in concrete properties (fresh state, mechanical strength, durability).

These results need to be nuanced because they are linked to the intrinsic properties of RCAs (grain size, water absorption) and the amount of CO2 stored in their cementitious matrix. The possibilities for optimizing industrial processes could thus lead to a return to the certain effect of carbonated aggregates on these properties, as described in the literature.

The results obtained by the FastCarb project on the properties of concretes manufactured with CRCA enable us to comply with the thresholds adopted in France for recycled aggregates. The requirements of standard NF EN 206+A2/CN are therefore applicable to concretes made with carbonated recycled aggregates.

Conditions for developing the use of GBRCs

RCAs can become carbon sinks. Therefore, future RCA carbonation plants will only be able to develop if it is environmentally and economically attractive for operators to create an activity that brings RCA closer to CO_2 sources and delivers the CRCA produced to construction sites with the smallest possible carbon footprint [10].

Results obtained from industrial demonstrators show that the carbonation of one tonne of fine RCA fraction captures 31 to 39 kg of CO_2, while that of the coarser fraction captures 5 to 12 kg of CO_2. On the other hand, the project data indicate that the CO_2 emissions for one tonne of CRCA transported 30km from the recycling site to the carbonation site and then 20km from this site to the ready-mix concrete production unit are 11kg. The balance of CO_2 emissions under these conditions is - 1 kg of CO_2 per tonne of gravel and - 28 kg of CO_2 per tonne of sand.

The following recommendations for CRCA production can be made:

- Optimize the *location of deconstruction concrete recycling platforms* to reduce the share of transport in the environmental balance sheet, by encouraging modal transfer (river, rail) where possible; this action is consistent with one of the objectives of the extended responsibility of producers of construction products and materials, which is to increase the number of platforms for deconstruction waste.
- Continue operational work on the *types of crushers* to be used in concrete recycling, to concentrate the cementitious fraction, which recarbonates, compared with the granular fraction, whose core is aggregate with no recarbonation potential.

In view of the initial results obtained, it would appear more profitable for the accelerated carbonation unit to be fed solely with the sand fraction of the RCAs since it captures more CO_2. Applying the results obtained with the CRCAs studied to the case of an exterior wall concrete (XC4/XF1), with 500 kg of carbonated recycled gravel and 180 kg of carbonated recycled sand, the saving is 7.3 Kg of CO_2 per m3 of concrete compared with the use of natural aggregate. Using the performance-based approach to validate a formula containing, for example, 100% of recycled gravel and 50% of recycled sand, 17.4 Kg of CO_2 per m3 of concrete could be saved, i.e. around 10% of the climate change impact of concrete.

Concerning RCA resources, a study by the French Union Nationale des Producteurs de Granulats (UNPG) concludes that the available tonnage of recycled concrete aggregates to be carbonated is around 20 Mt in France, with a concentration of deposits in major cities. Improved waste sorting at all stages of deconstruction (building sites, platforms, etc.) should make easier the identification, location and marking of concretes so that they can be directed to recycling channels, increasing tonnages of RCA and eventually CRCA.

In terms of point sources of CO_2 emissions, the study carried out as part of the project counted over 2,300 in France, including those subject to the ETS Directive and those controlled under the ICPE, as well as methanization facilities injecting methane into gas networks.

For companies subject to the ETS directive (that establishes a system for greenhouse gas emission allowance trading within the Union), one tonne of CO_2 captured in an RCA saves an allowance valued between €80 and €90/t (in August 2023). For others, this emissions reduction can earn Carbon Offset Credits which, like unused CO_2 quotas, will contribute to the profitability of future CO_2 capture projects.

Whenever possible, the use of compressed CO_2 to carbonate RCAs on the dismantling site or a recycling platform will limit transport-related CO_2 emissions.

CONCLUSIONS

The FastCarb project has already yielded lessons concerning the accelerated carbonation of recycled concrete aggregates. Following laboratory research, which highlighted the key factors in the phenomenon and led to recommendations on tests to quantify the CO_2 captured, two demonstrators were installed in cement plants, where it was possible to use kiln gases directly on an industrial scale. The quantity of CO_2 stored reached 40 kg/t of RCA for the sand fraction.

The LCA on these demonstrators confirms that the sand fraction of RCA is the most interesting material in terms of CO_2 absorption and impact on climate change. Indeed, the CO_2 eq./t indicator for carbonated recycled sand is negative. The use of carbonated RCA can significantly reduce the carbon footprint of a m3 of concrete. This is also interesting for circular economy purposes, as recycled sand is more difficult to use in concretes including RCA.

The FastCarb project shows that accelerated carbonation is feasible on an industrial scale with a reduced optimization phase, that results are consistent with laboratory experiments and that environmental impacts are positive when RCA transport is limited. The amount of CO_2 that could be mineralized by these processes alone will not solve the problem of CO_2 emissions from the concrete industry, but it is a possible and interesting contribution that completes the interest of the use of RCA in concretes with regard to the circular economy.

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