



# Influence of the Composition of Original Concrete on the Carbonated Recycled Concrete Aggregates Properties

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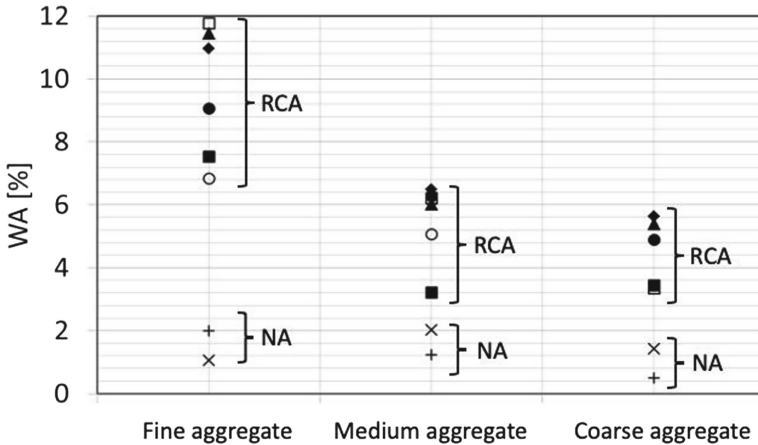
**Abstract.** Recycled concrete aggregates (RCA) heterogeneity, leads to different properties compared to the natural aggregates (NA), especially in terms of their water absorption (WA24). Moreover, the variability of these RCA properties is larger than the NA one. This is mainly due to the compositions of original concrete. These disparities in properties and their high variation range limit the reuse of RCA in concrete. In the construction industry, concrete production has a significant environmental impact. Indeed, the cement production induces high greenhouse gas emissions. Accelerated carbonation of RCA can combine the advantages of a capture of CO<sub>2</sub> issued from plant and a reduction of water absorption of aggregates. Indeed, the carbonation reaction clogs the capillary networks of aggregates and then reduces the accessible porosity that directly influences the water absorption. To reduce global carbon dioxide emissions and to enhance the recycling of RCA, the French national project FastCarb aims to optimize an accelerated carbonation process at an industrial scale. This work is to study the evolution of RCA properties, issued from several batches, following treatment in a carbonation chamber with defined parameters. This paper analyzes the influence of accelerated carbonation and its efficiency on the evolution of the RCA absorption and its variability. In order to identify the influence of the composition of original concrete, RCA with various original concrete compositions were crushed then tested. It was shown that accelerated carbonation decreases RCA absorption but not its variability. Carbonation (evaluated by mass gain) and absorption reduction efficiencies are not directly correlated.

**Keywords:** Recycled concrete aggregate · Accelerated carbonation · Absorption · Mass gain

## 1 Introduction

Recently, the use of recycled concrete aggregates (RCA) is a common practice in the construction industry. Obtained by crushing old concrete, RCA are composed of natural aggregates and cement paste. This composition leads to some differences in properties

with natural aggregates (NA) and a wide range of variation in these properties, particularly regarding their water absorption (see Fig. 1). These differences are due to the composition of the parent concrete (initial concrete before work demolition) and limit the use of GBR in new concrete at large scale.



**Fig. 1.** 24h water absorption (WA24) of RCA and NA and their variations [1]

The cement industry contributes 5–7% of the global greenhouse gas emissions.

In an attempt to reduce CO<sub>2</sub> emissions and to expand the volumes of RCA used in concrete production, the FastCarb project [2] intends to store the CO<sub>2</sub> released during cement production in RCA while improving their properties.

The carbonation of concrete has been widely studied in the past as a phenomenon that can lead to the corrosion of the rebars in concrete and consequently to the degradation of concrete structures [3]. On the contrary, carbonation is now studied for its positive environmental aspect, which is the mineralization of atmospheric CO<sub>2</sub> (in the case of natural carbonation) or of CO<sub>2</sub> from industrial processes (in the case of accelerated carbonation) [4, 5].

The objective of the FastCarb project is to propose an accelerated carbonation process on an industrial scale [6].

The efficiency of the different accelerated carbonation techniques is highly dependent on many parameters specific to each one: duration of the treatment, total pressure and relative humidity in the chamber, partial pressure of CO<sub>2</sub>, possible flow of gases through the chamber (if CO<sub>2</sub> flow), possible movement of the aggregates (by rotation of the chamber for example if dynamic method), etc. [7–10].

The properties of the RCA to be treated and their conditioning before carbonation also have a major influence on the efficiency of the carbonation techniques: previous carbonation state, water saturation degree, grain size, porosity, characteristics of the parent material (nature of the cement, nature of the aggregates), etc. [7, 11].

Some research studies recommend the implementation of pre-treatment methods for recycled aggregates before the application of the carbonation process (vacuum,

water equilibrium vs controlled relative humidity, preliminary immersion in a calcium hydroxide solution, etc.) [7, 12].

These methods could increase the storage of CO<sub>2</sub> as well as the beneficial effects on the physical and mechanical properties of the treated aggregates.

The composition of the RCA is a major factor of the carbonation reaction potential, especially its cement content. Indeed, the content and type of cement define the quantity of phases that can be carbonated (portlandite Ca(OH)<sub>2</sub>, hydrated calcium silicates C-S-H, ...) [13, 14].

In the case of carbonation of concrete, the progress of the carbonation front decreases with the cement content of the concrete due to a buffer effect. Also, the rate of carbonation can be correlated with the mechanical properties of the concrete, as high strength material induces a compact microstructure limiting the reactions with the CO<sub>2</sub> [15].

The mass gain during accelerated carbonation is an indicator that allows a qualitative evaluation of the process efficiency in some situations, e.g. when selecting the parameters of the accelerated carbonation process. Thus, Djerbi considers that the mass of CO<sub>2</sub> fixed during the carbonation is correlated to the dry mass gain. This approach underestimates the rate of CO<sub>2</sub> capture by 5 to 10%, but this error is assumed to be systematic for a given recycled aggregate [16].

The evaluation of the influence of mineralised CO<sub>2</sub> during carbonation on the properties of RCA, in particular their absorption, requires specific investigation. The objective of this study is to analyze the evolution of the absorption and its ranges of variation after an accelerated carbonation treatment of RCA. The analysis includes the influence of the composition of the parent concrete of the RCA. The parameters of the accelerated carbonation treatment were defined beforehand considering an analysis of literature and a consultation between the research teams of the FastCarb project. A qualitative approach to estimate the efficiency of the treatment, i.e. the quantity of mineralized CO<sub>2</sub>, is proposed by measuring the mass gain resulting from the carbonation. Furthermore, a pH test based on phenolphthalein pulverization was performed in order to check if the carbonation of the specimen was homogeneous after the treatment.

## 2 Materials and Methods

### 2.1 Materials

RCA were obtained by crushing in November 2020 concrete produced in the laboratory between February and March 2019. They were formulated as ordinary concrete. The composition parameters are given in Table 1.

Workability of concrete was controlled by slump test. Concrete were tested in compression 28 days of curing. The results are given in Table 2.

Concrete were crushed using a laboratory jaw crusher and then separated into 2 granular grades 0/4 mm and 4/10 mm. In this study, only the 4/10 mm RCA were studied and then carbonated. A control of the carbonation depth of the 16 × 32 cm specimen was performed before crushing to assure that a residual potential of carbonation existed.

**Table 1.** Concrete compositions.

Concrete ref	Natural Aggregate origin	Cement type	W/C ratio	Cement content (kg/m <sup>3</sup> )
BT19-1	Rolled, silico-calcareous	CEM I-52.5	0.66	280
BT19-2	Rolled, silico-calcareous	CEM II-42.5	0.66	280
BT19-3	Rolled, silico-calcareous	CEM III-42.5	0.66	280
BT19-4	Rolled, silico-calcareous	CEM I-52.5	0.58	350
BT19-5	Rolled, silico-calcareous	CEM II-42.5	0.58	350
BT19-6	Rolled, silico-calcareous	CEM III-42.5	0.58	350

**Table 2.** Compressive strength results, 28 days of age, (MPa).

Concrete ref	Comp. Strength	Slump class
BT19-1	34	S4
BT19-2	25	S5
BT19-3	26	S4
BT19-4	37	S3
BT19-5	34	S3
BT19-6	30	S4

## 2.2 Methods

In order to limit damage to the cementitious matrix, the RCA were dried at 60 °C until the mass was completely stabilized, before any characterisation on RCA or RCA carbonated (RCAC), and also before carbonation tests on RCA. A pH test based on phenolphthalein pulverization was performed in order to check if the carbonation of the specimen was homogeneous after the treatment.

**RCA Characterization.** Absorption tests (W24) were performed on non-carbonated RCA and carbonated RCA according to NF EN 1097-6 standard. For each composition and carbonation state, the absorption test was performed for three samples (150g each). Mass increase in terms of gain between non-carbonated RCA and carbonated RCA were measured.

**Accelerated Carbonation Test.** A carbonation test was performed on the RCA with the following carbonation parameters:

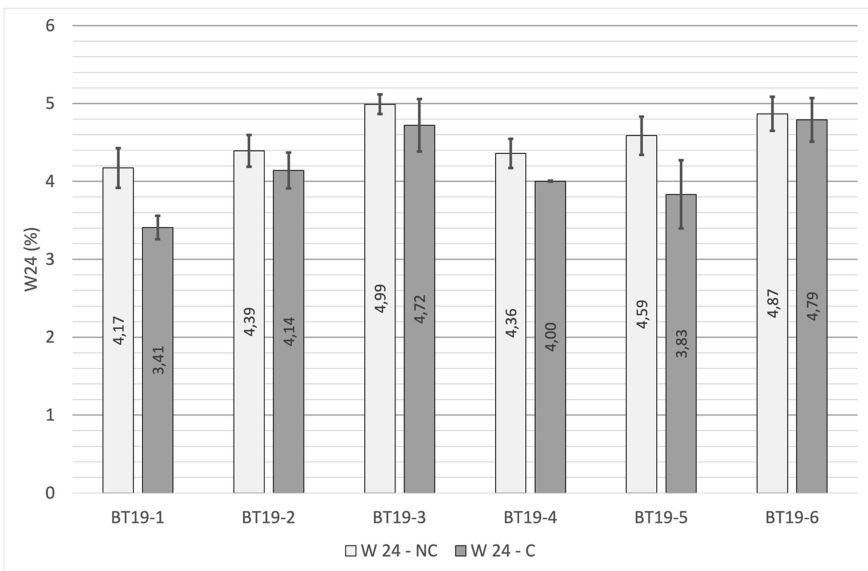
- Pre-humidification, applied water content: 75% of W24 (measured on an initial batch of non-carbonated RCA)
- CO<sub>2</sub> content in the chamber: 15%.
- Relative humidity set point in the chamber: 60%.
- Pressure in chamber: atm
- Temperature set point in the chamber: 20 °C

– Carbonation time: 24 h

The 6 RCA samples (BT19-1 to 6) were processed during a single carbonation test. For each composition, 1200g sample was submitted to carbonation test. Water content was controlled after the carbonation process. Evolution of the relative humidity set point in the chamber was controlled.

### 3 Results and Discussion

Considering all the specimens the mean absorption is  $4.56\% \pm 0.37$  before carbonation (RCA) and  $4.20\% \pm 0.60$  after it (RCAC). The decrease of absorption is about 0.37 points (Absolute  $\Delta$  W24) that represents a relative value of 8% based on the initial value before carbonation (Relative  $\Delta$  W24%). The increase of the standard deviation (variability of absorption) is 0.22 points (Absolute  $\Delta$   $\sigma$  W24) that represents a relative value of 60% based on the initial value before carbonation (Relative  $\sigma$  W24%). Detailed values by composition are given Fig. 2 and Table3.



**Fig. 2.** Absorption (W24) of RCA (NC) and RCAC (C).

Considering a same cement paste volume (or a same water-to-cement ratio), respectively 28% for samples BT19-1 to BT19-3 and 32% for 4 to 6 samples BT19-4 to BT19-6; in the case of RCA samples, absorption increases with clinker content and mechanical strength. Clinker contents are respectively 98% for CEM I, 86% for CEM II and 37% for CEM III. For RCA samples 4 to 6, increase of absorption could also be attributed to decrease of mechanical strength. In the case of RCAC samples, except for the BT19-5, the same conclusion can be announced. (n.b. BT19-5 C presents a high variability).

Considering comparison between samples BT19-1 with BT19-4, BT19-2 with BT19-5 and BT19-3 with BT19-6, a higher cement content with a lower water-to-cement ratio (leading to a higher cement paste volume) increases slightly absorption in the case of CEM I and CEM II. As a consequence, the increase of mechanical strength due to higher cement and paste cement content doesn't lead to a decrease of absorption.

For all the RCA, carbonation treatment decreases absorption. On contrary variability (3 repetition tests) is deteriorated for RCAC compared to RCA (Table 3). This contradiction could be explained by a modification of the pore size distribution, despite a global reduction of porosity.

**Table 3.** Absorption and variability evolutions between RCA and RCAC

Concrete ref	Absolute $\Delta$ W24	Relative $\Delta$ W24 (%)	Absolute $\Delta$ $\sigma$ W24	Relative $\sigma$ W24 (%)
BT19-1	-0.76	-18.33	-0.13	-40.81
BT19-2	-0.25	-5.74	0.03	12.45
BT19-3	-0.27	-5.39	0.26	165.95
BT19-4	-0.36	-8.22	0.28	120.24
BT19-5	-0.75	-16.42	0.24	78.80
BT19-6	-0.08	-1.58	0.07	27.52

As a global result, carbonation leads to a decrease of absorption but to an increase of absorption variability in absolute and relative values.

Considering all the specimens the mean mass gain due to carbonation is  $5.92\% \pm 2.63$ , that represents  $5.95 \text{ g/kg}$  of RCA (Fig. 3).

Gain mass seems to be independent from composition, mechanical strength and initial absorption of RCA.

Regarding the evolutions of water content of the samples and of the relative humidity point in the chamber during the treatment (up to 90%), it could be considered that a non-conservation of the thermohydric equilibrium contributes to dispersing the results, especially mass gain. Indeed, the mean water content variation is  $0.50 \pm 0.10$  points that represents a relative value of 15.12% based on the initial value before carbonation (Fig. 4). A slight correlation between mass gain and water content evolution is observed: the more the sample loss water, the less gain mass is observed. Furthermore, simultaneous carbonation of many samples issued from different compositions and initial absorptions could lead to a global hydric equilibrium in the chamber.

Monitoring the efficiency of the treatment by measuring the mass gain does not allow to dissociate the evolutions of mass due to different origins: release of bound water, reduction of the amount of portlandite, formation of calcite, ... Moreover, when gain mass is used to qualify the carbonation of RCA, only the carbonation of portlandite is concerned, for the other hydrates (CSH, ettringite) the water loss balance is difficult to estimate [17].

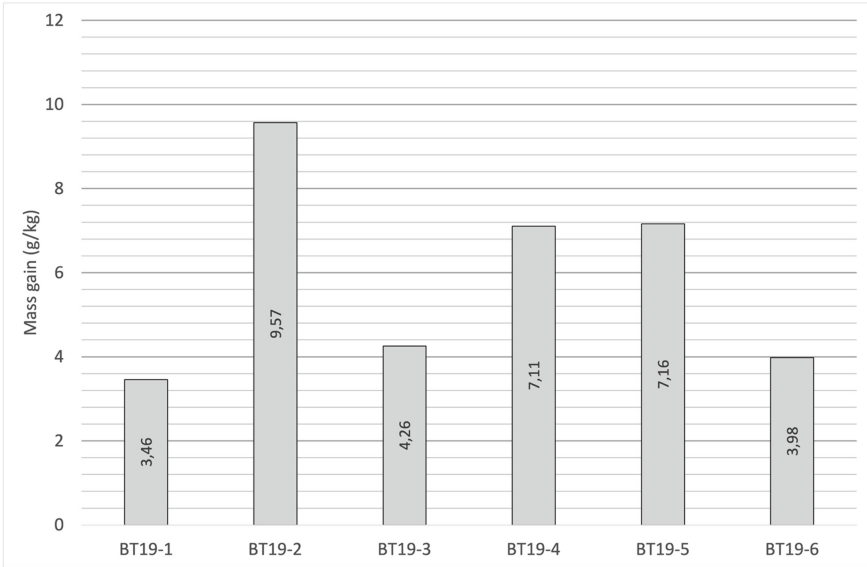


Fig. 3. Mass gain between RCA (NC) and RCAC (C).

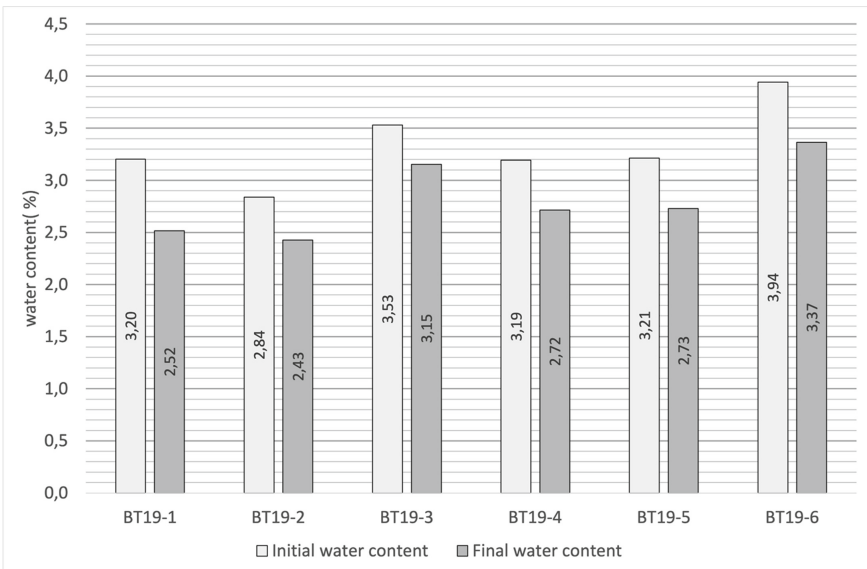
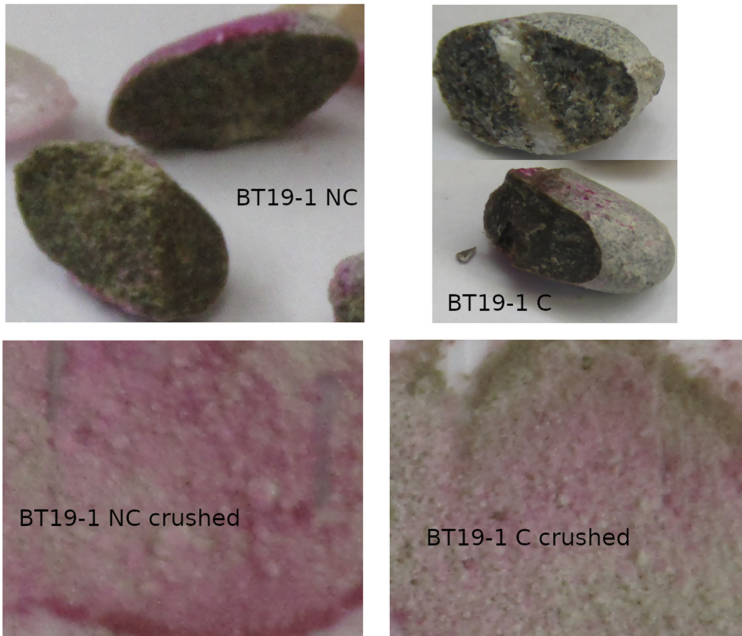


Fig. 4. Pretreatment water content of RCA (NC) and water content of RCAC (C) at the end of the carbonation treatment.

For future tests, gain mass measurement to qualify the efficiency of a carbonation process should only be used for a comparison between RCA whose parent concrete compositions (especially the cement phase) are close [18].

Phenolphthalein test observations confirm that RCA samples were carbonated by the treatment. As illustrated Fig. 5. For BT19-1, crushed RCAC are higher carbonated than RCA.



**Fig. 5.** Phenolphthalein test of RCA (NC) and RCAC (C) - Aggregate and crushed aggregate

Part of results observed in this study are confirmed by literature. In particular, it was shown in this study that accelerated carbonation decreases RCA absorption for all compositions (parent concrete) and phenolphthalein tests confirm the carbonation of RCA after treatment. This conclusion is commonly accepted and mentioned by Torrenti and al [19].

No notable correlation between decrease of the absorption or gain mass (due to carbonation) and initial composition or properties of the samples were identified. But it is known in literature that content and type of cement define the quantity of phases that can be carbonated [13, 14]. One could expect that compositions with higher cement/clinker content should present a higher decreasing of absorption and a higher mass gain. This no correlation could be explained from one part by initial mechanical strength of RCA which were linked to clinker content and cement paste content. Indeed high strength material induces a compact microstructure limiting the reactions with the  $\text{CO}_2$  [15]. It is also important to mention that for the same cement paste content and whatever the sample state (carbonated or not), it was shown on this study that absorption is correlated to clinker content but for the same cement type none of cement paste content and compressive strength have any significant influence on absorption. Thus, it could be concluded that efficiency of accelerated carbonation cannot be directly correlated to



initial composition or to initial properties (mechanical strength and absorption) of RCA because influences of these parameters are contradictory. Furthermore, it was concluded that mass gain measurement is insufficient to quantify efficiency of carbonation process mainly because it is based on the carbonation of the only portlandite [17].

## 4 Conclusion and Perspectives

The influence of accelerated carbonation and its efficiency on the evolution of the RCA absorption and its variability were analyzed and deepened. The main conclusions and perspectives of this study are:

- It was confirmed that accelerated carbonation decreases RCA absorption and increases carbonation depth but after accelerated carbonation, absorption variability increases for almost all the samples although absorption decreases.
- Initial compositions and properties of RCA could lead to opposite influences on the efficiency of accelerated carbonation process (carbonation potential and accessible porosity).
- No correlation between mass gain and initial absorption were identified. The change of water content over the carbonation treatment influences the mass gain. This change is linked to the thermo-hydric equilibrium. The release of water during the carbonation of cement hydrates and the drying in the carbonation chamber (HR 60%) disrupt this equilibrium.
- As a recommendation for further tests, samples of different compositions and/or different absorptions should be treated separately to avoid mutual interactions. Especially as water content is managed by initial absorption.
- For the evaluation of the carbonation efficiency, measurement of  $\text{CaCO}_3$  content should be more accurate than the one of mass gain, owing to various complex origins of this gain mass.
- In order to enhance the comprehension of correlation between absorption and  $\text{CO}_2$  fixation, more investigations on  $\text{CaCO}_3$  content and crystallography (calcite, aragonite, vaterite polymorphous) should be performed. Indeed, their formations are influenced by the physico-chemistry of the environment (pH, temperature, etc.). Different shapes of the three  $\text{CaCO}_3$  polymorphous crystals should lead to different “clogging” of the porous network and as a consequence to different influences on the absorption evolution. Moreover, as mentioned by Thiery [20], the clogging of porous network prevents the ions transfer and as consequence, progression of carbonation.
- Current research conducted within the scope of the French national project Fastcarb should contribute to clarify several of these questions.

## References

1. Déodonne, K.: Études des caractéristiques physico-chimiques des bétons de granulats recyclés et de leur impact environnemental, Strasbourg, Ph.D. thesis, Strasbourg University (2015)
2. Fastcarb National Project. <https://fastcarb.fr/>. Accessed 29 Nov 2022

3. Baroghel-Bouny, V., Carpa, B.: La durabilité des armatures et du béton d'enrobage, in La durabilité des bétons. In: Ollivier, J.P., Vichot, A. (eds.) Presse de l'Ecole Nationale des Ponts et Chaussées, Paris, pp. 303–385 (2008)
4. Fernández-Bertos, M.: A review of accelerated carbonation technology in the treatment of cement-based materials and sequestration of CO<sub>2</sub>. *J. Hazard. Mater.* **112**(3), 193–205 (2004)
5. Jang, J.G.: Review on recent advances in CO<sub>2</sub> utilization and sequestration technologies in cement-based materials. *Constr. Build. Mater.* **127**, 762–773 (2016)
6. Sereng, M.: Improvement of recycled aggregates properties by means of CO<sub>2</sub> uptake. *Appl. Sci.* **11**(14), 1–22 (2021)
7. Zhan, B.: Experimental study of CO<sub>2</sub> curing for enhancement of recycled aggregate properties. *Constr. Build. Mater.* **67**(A), 3–7 (2014)
8. Brück, F.: Accelerated carbonation of waste incinerator bottom ash in a rotating drum batch reactor. *J. Environ. Chem. Eng.* **6**(4), 5259–5268 (2018)
9. Chang, E.-E.: Accelerated carbonation of steelmaking slags in a high-gravity rotating packed bed. *J. Hazard. Mater.* **227–228**, 97–106 (2012)
10. Simoes dos Reis, G.: Effect of the accelerated carbonation treatment on the recycled sand physico-chemical characteristics through the rolling carbonation process. *J. CO<sub>2</sub> Utilization* **39**, 1–12 (2020)
11. Kurdowski, W.: *Cement and Concrete Chemistry*, 1st edn. Springer, Heidelberg (2014)
12. Pan, G.: Effect of CO<sub>2</sub> curing on demolition recycled fine aggregate enhance by calcium hydroxide pre-soaking. *Constr. Build. Mater.* **154**, 810–818 (2017)
13. Mistri, A.: A review on different treatment methods for enhancing the properties of recycled aggregates for sustainable construction materials. *Constr. Build. Mater.* **233**, 1–12 (2020)
14. Liang, C.: Utilization of CO<sub>2</sub> curing to enhance the properties of recycled aggregate and prepared concrete: a review. *Cement Concr. Compos.* **105**, 1–14 (2020)
15. Shah, V.: Determination of carbonation resistance of concrete through a combination of cement content and tortuosity. *J. Build. Eng.* **2022**(60), 105176 (2022)
16. Djerbi, A.: Stockage du CO<sub>2</sub> dans les granulats recyclés: développement des procédés de carbonatation accélérée. *Acad. J. Civil Eng.* **40**(3), 1–19 (2022)
17. Cazagliu, B., Djerbi, A.: Méthode de détermination du CO<sub>2</sub> piégé. In : *Fast Carb Conférence: Le béton recyclé, un puits de carbone!*, Paris, France, 27 Sept 2022
18. Braymand, S.: Carbonatation accélérée de granulats de béton recyclé – Évolution des propriétés selon leur classe granulaire. *Acad. J. Civil Eng.* **40**(1), 76–79 (2022)
19. Torrenti, J.M.: The FastCarb project: taking advantage of the accelerated carbonation of recycled concrete aggregates. *Case Stud. Constr. Mater.* **17**, e01349 (2022)
20. Thiery, M.: Carbonation kinetics of a bed of recycled concrete aggregates: a laboratory study on models materials. *Cem. Concr. Res.* **46**, 50–65 (2013)