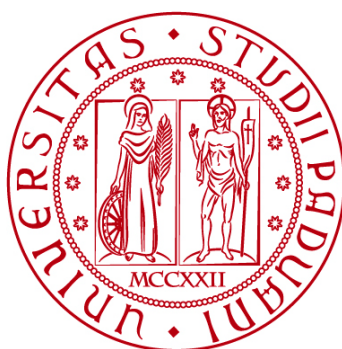


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Department Of Civil, Environmental and Architectural Engineering

Corso di Laurea Magistrale in Environmental Engineering



TESI DI LAUREA

**CARBON DIOXIDE CAPTURE BY ALKALINE WASTE
MATERIALS: AN EXPERIMENTAL ASSESSMENT**

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ANNO ACCADEMICO 2021-2022

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PREMISE

This written elaborate resumes the master thesis work carried out between February and November 2022. It consists of three parts. Part I plays an introductory role to give the key concepts for a full comprehension of the subject of accelerated carbonation. Part II consists of a scientific article that summarizes the theoretical considerations found in the literature with a systematic bibliographic research, and the experience of an experimental work performed in the laboratory. The second part is the real heart of the work, since it was written as a scientific article that will be submitted to a specialized peer reviewed journal in order to be published. Part III contains all the dirty work that does not appear in the previous two. It is made up of graphs, tables and calculations that have been utilized for the realization of the entire master thesis project.

PART I - INTRODUCTION

1.1 INTRODUCTION AND CONCEPTUAL MODEL

Since the aim of this study is to start from a bibliographic review to understand how the accelerated carbonation processes have been carried out in literature, a conceptual model was designed to further understand the mechanisms of the accelerated carbonation and to design the experimental campaign. In **Fig. 1** the schematic of the conceptual model is displayed. The systematic review has the purpose to identify the parameters for the evaluation of the process, to obtain a list of parameters with standards, equipment and references. Moreover, it has the objective to identify the potentiality of CO₂ capture of the different materials tested in previous accelerated carbonation study. At last the systematic review has as its goal to define the technology: classify the sorbent materials, the route of the process and the operative conditions. All these information are present in the paragraph related to the critical review. Since formulas for the theoretical prediction of the CO₂ uptake are widely used in the literature, a paragraph has been dedicated to mathematical modelling. Through the data collected by literature analyses, the design of experimental activities was created. The paragraph named “Part 2” related to the experimental campaign collects all the information regarding the execution of the experimental campaign. It has the purpose to investigate the possibility of applying the accelerated carbonation process on a mix of alkaline wastes with the aim of assessing their potential for CO₂ sequestration. Different conditions have been tested to provide the basis for a future scale up of the process. For the purpose of this work, several accelerated carbonation tests were performed at three different grain sizes (0/6mm, 6/16mm and 16/31mm) according to 3 different liquid to solid ratios (L/S=0.0 L/kg, L/S=0.2 L/kg and L/S=0.3 L/kg), for three different times (2, 4, 8h) to 4 different masses (200g, 100g, 50g, 25g) under fixed operating conditions (relative humidity, CO₂ concentration, pressure and temperature).

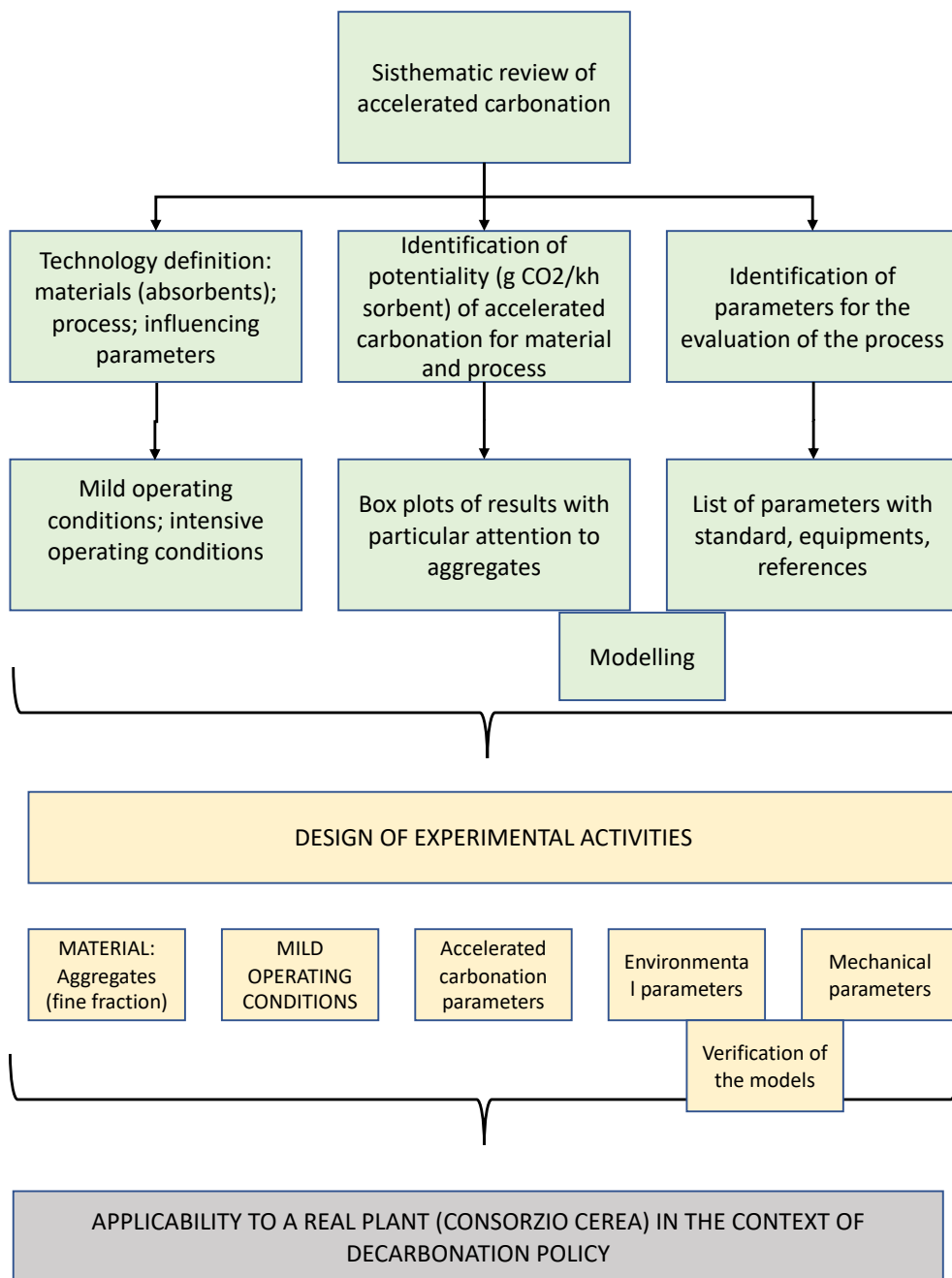


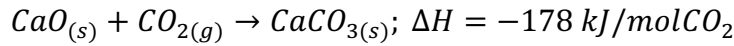
Figure 1. Schematic of the conceptual model.

1.2 CHEMISTRY

1.2.1 Introduction to carbonation

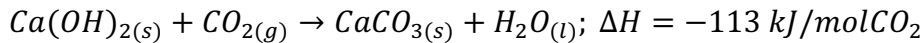
The process is based on acid-base reactions in which carbonate acid is neutralized by a base (alkaline mineral). After this neutralization reaction, carbon dioxide is fixed in solid state in the form of carbonate. Because of the wide range in composition of alkaline wastes, other oxides like (free CaO , $Ca(OH)_2$, Ca_2SiO_3 , MgO , K_2O , Na_2O) also react with carbon dioxide and contribute to the carbon sequestration. Not only $Ca(OH)_2$, but also other hydrate compounds can react with CO_2 . For instance, $Mg(OH)_2$ produce $MgCO_{3(s)}$ and the calcium-silicate-hydrates (C-S-H) produce $CaCO_3$ and a silica gel.

The general formula that summarizes the entire calcium carbonate formation process states that lime ($CaO_{(s)}$) reacts with carbon dioxide according to the following reaction:

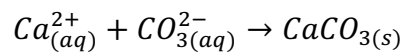
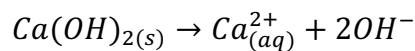
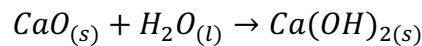
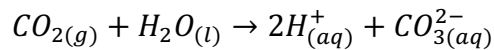


In calcium rich wastes, free CaO hydrates to $Ca(OH)_2$. The $Ca(OH)_2$ dissolves providing high concentrations of Ca^{2+} and OH^- . A high pH value conditions are es, the water acts as a CO_2 -trap where carbonic acid formed at equilibrium with the prevailing CO_2 partial pressure fully dissociates to CO_3^{2-} . The protons released are neutralized by OH^- whereas Ca^{2+} combines with the carbonate anion and precipitates as $CaCO_3$.

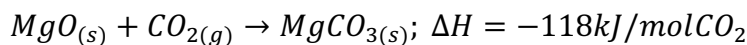
This process is represented by:



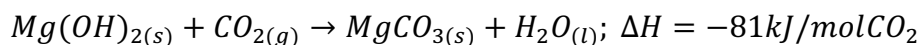
This process is better explained through the following equations:



As magnesium oxide is present in significant concentrations in alkaline wastes (in particular it is the most present after lime) magnesium oxides reactions with carbon dioxide are presented:



Even hydrated magnesium reacts with carbon dioxide to form magnesium carbonate as the following equation:



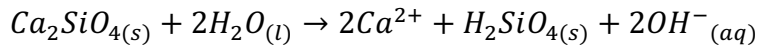
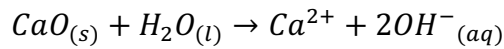
Since Enthalpy has a negative value, the reactions are exothermic, so it produces heat. According to Le Chatelier principle, if temperature increase, the product formation is inhibited.

1.2.2 Ion equilibrium solution for calcium carbonation

Carbonation involves 3 steps: the leaching of calcium in solid particles, dissolution of CO₂ into the solution, carbonate precipitation.

1.2.2.1 Leaching of calcium and metal oxides.

The first step of carbonation was believed to be the leaching of calcium in solid particles. Dissociation of calcium species present in alkaline solid waste is favored at low pH. The dissolution of lime and other metal oxides in water causes a strong increase in pH (10.88–11.88). Usually the alkaline-metal oxides are blocked into the silicate, aluminate or ferrite phase. One of the most present metal-oxide in alkaline waste is larnite $Ca_2SiO_{4(s)}$. While, for instance MgO is rarely present in pure form. The dissolution process of lime and larnite in water is presented through the following equations:



In order to improve this process, is necessary to disintegrate coarse, or larger granular feed solids to obtain a finer granulometry.

1.2.2.2 Dissolution of carbon dioxide into the solution

The second step of carbonation is assumed to be the dissolution of CO₂ into the solution. CO₂ has a critical point at 31.06°C and 73.8 bars and a critical density of 0.469 g/cm³. It could be dissolved from the atmosphere in available water, and carbonic acid can be created with a pH around 5.6. In addition, diffusion of CO₂ in water is approximately 10000 times lower than in air as Henry's constant will demonstrate. CO₂ can be physically absorbed in water (or a solvent) in accordance with Henry's law. Henry's law states that the relationship between the gas solubility in water and the partial pressure of the gas is strictly valid only for gases that can be infinitely diluted in solution. The binding of the solvent with CO₂ occurs at high pressure, and a reduction in pressure releases the gas.

The amount of CO₂ dissolution in water can be expressed by Henry's law as shown in previous equations:

$$C_{CO_2} = H_{CO_2} \cdot P_{CO_2}$$

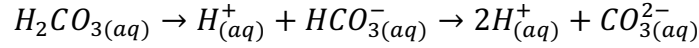
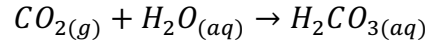
Where C is the concentration of CO₂ dissolved in aqueous solution (M); H_{CO₂} is Henry's constant for CO₂ (= 10^{-1.46} M/atm at 25°C); and P_{CO₂} is the partial pressure of CO₂ in the gas phase (atm). Therefore, the higher partial pressure of CO₂ will result in a large amount of dissolved CO₂.

In addition, Henry's constant is a function of temperature that can be modified by the following equation: (Morel and Hering, 1993)

$$K_{H,T} = K_{H,298K} \cdot \exp [C \cdot (1/T - 1/298)]$$

Where C is the constant for all gases (2400 K for CO₂) and T is the temperature (K).

Dissolution of CO_2 into water is pH-dependent because of the dissociation of carbonic acid (H_2CO_3) into carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) ions. The dissociation of the carbonic ions is shown in the following equations:



The corresponding equilibrium constants could be expressed as function of K_a , K_b :

$$K_a = \frac{[H^+][HCO_3^-]}{[H_2CO_3]}; \quad K_b = \frac{[H^+][CO_3^{2-}]}{[HCO_3^-(aq)]};$$

where $K_a = 10^{-6.3}$; $K_b = 10^{-10.3}$ at $25^\circ C$.

Fig.2 shows the mole balance and equilibrium conditions for carbonation of alkaline solid wastes.

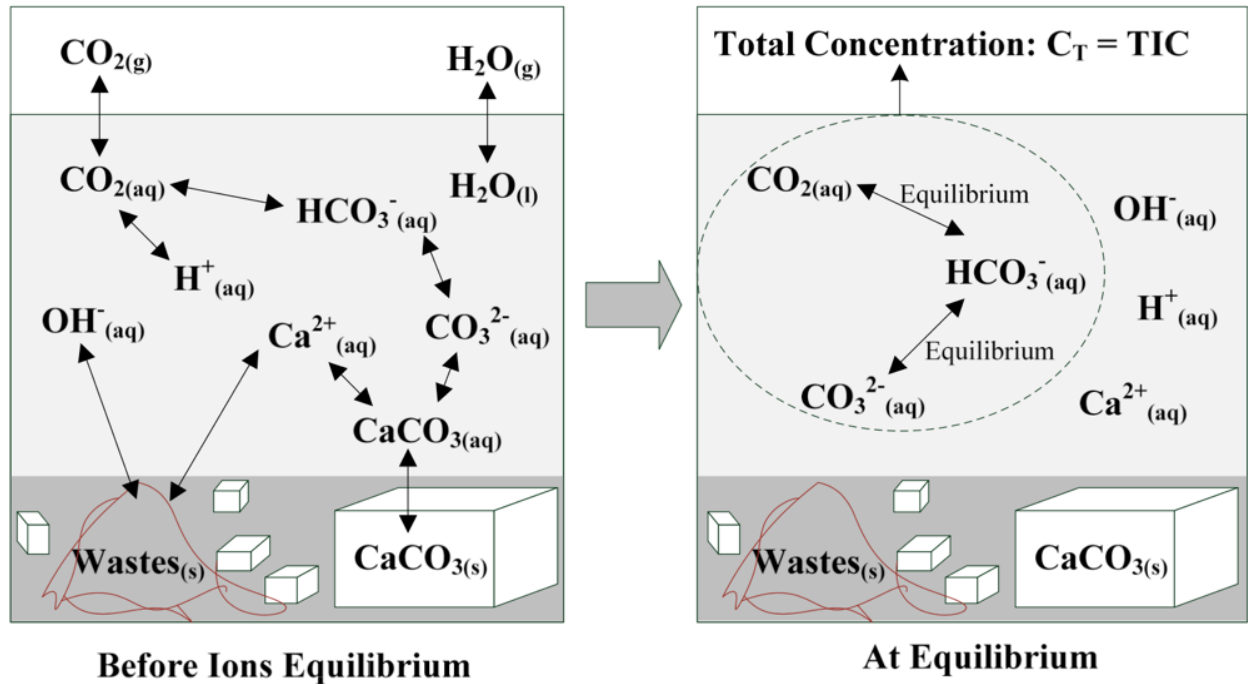


Figure 1 Mole balance and equilibrium conditions for carbonation of alkaline solid wastes.

The mole balances of the carbonic acid system can be expressed as:

$$C_T = [H_2CO_{3(aq)}] + [HCO_3^-(aq)] + [CO_3^{2-}(aq)]$$

where C_T is the total inorganic carbon (TIC) concentration (M).

By substitution of K_a , K_b into Eq.15, an explicit formula for $[H_2CO_3^*(aq)]$, $[HCO_3^-(aq)]$ and $[CO_3^{2-}(aq)]$ is obtained as:

$$[H_2CO_{3(aq)}] = \alpha_0 \cdot C_T; \alpha_0 = \frac{[H^+]^2}{[H^+]^2 + K_a[H^+] + K_aK_b};$$

$$[HCO_3^-] = \alpha_1 \cdot C_T; \quad \alpha_1 = \frac{K_a[H^+]}{[H^+]^2 + K_a[H^+] + K_a K_b};$$

$$[CO_3^{2-}] = \alpha_2 \cdot C_T; \quad \alpha_2 = \frac{K_a K_b}{[H^+]^2 + K_a[H^+] + K_a K_b};$$

Where the fraction (α_i) of each carbon species present is dependent on the solution pH.

At a low pH (~ 4), the production of H_2CO_3 dominates, at a mid pH (~ 8) HCO_3^- dominates, and at a high pH (~ 12) CO_3^{2-} dominates. Therefore, accelerated carbonation is favored at a basic pH due to the availability of carbonate ions. Alkaline wastes, due to the high concentration of metal oxides, are characterized by a basic pH. Thus, they establish good conditions for carbonation therefore they are suitable materials for carbon capture and storage studies.

Dissolution of Carbon Dioxide into Solution - Kinetics

In Fig.3 the kinetics of dissolution of CO_2 and dehydration of H_2CO_3 are presented. It was noted that these two reactions should occur simultaneously as shown in the figure below.

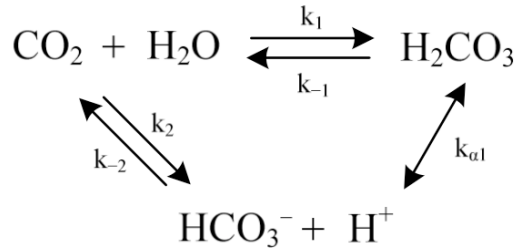


Figure 3. Dissolution of Carbon Dioxide into Solution

Therefore, the rate expression of the dissolution-dehydration reaction is:

$$-\frac{d[CO_2]}{dt} = (k_1 + k_2)[CO_2] - k_{-1}[H_2CO_3] - k_{-2}[HCO_3^-][H^+]$$

By substituting the relations of K_a, K_b into the rate expression of the dissolution-dehydration reaction, we obtain:

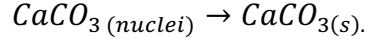
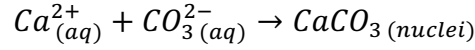
$$-\frac{d[CO_2]}{dt} = (k_1 + k_2)[CO_2] - (k_{-1} + k_{-2}K_a)[H_2CO_3] = k_{CO_2}[CO_2] - k_{H_2CO_3}[H_2CO_3]$$

where the overall rate constants in Eq. were simplified to be k_{CO_2} and $k_{H_2CO_3}$. The values of k_{CO_2} and $k_{H_2CO_3}$ at $25^\circ C$ were 0.032 s^{-1} and 26.6 s^{-1} , respectively. However, at higher pH ($pH > 9$), an alternative reaction pathway would be expressed as:

$CO_2 + OH^- \leftrightarrow HCO_3^-$; with kinetic rate constant k_4 . Where k_4 (i.e., 8500 l/M-s at $25^\circ C$) and k_{-4} (i.e., 0.0002 s^{-1} at $25^\circ C$) are the rate constants.

1.2.2.3 Carbonate precipitation

The carbonation formulation can be simply described by:



Contact between Ca^{2+} -ions and CO_2 leads to $CaCO_3$ precipitation, which is almost insoluble in water at pH levels above 9 (the solubility of $CaCO_3$ is 0.15 mmol/L at 25°C).

The solubility-product constant (K_{sp}) of calcium carbonate as shown in Eq. 124 ranges from 3.7×10^{-9} to 8.7×10^{-9} at 25°C, and 4.47×10^{-9} .

$K_{sp} = [Ca^{2+}][CO_3^{2-}]$. Where $[Ca^{2+}]$ and $[CO_3^{2-}]$ are the concentrations of calcium and carbonate ions (M) in the solution, respectively.

Rate of Calcium Carbonate Precipitation

The carbonation reaction is regulated by solution equilibrium, and the reaction of calcium ions combining with carbonate ions is very fast. It was widely assumed that the carbonation reaction as shown in previous equation was of the first order with respect to the concentrations of Ca^{2+} and CO_3^{2-} .

Therefore, the rate of carbonation could be expressed by the following differential Equation:

$$Q_{CO_2} = \frac{d(CaCO_3)}{dt} = \frac{d(CO_3^{2-})}{dt} = k[Ca^{2+}][CO_3^{2-}]$$

where $CCaCO_3$ is the concentration of calcium carbonate (mole/L), and k is the reaction rate coefficient (1/mol-sec).

The precipitation rate is related to the CO_3^{2-} concentration in the liquid phase, but not to the concentration of other species containing carbonate. In addition, the reaction rate coefficient (k) is dependent on the reaction temperature according to Arrhenius's law.

As the calcium ions are converted to metal carbonates and precipitated out, more calcium hydroxide dissolves to equalize the concentration of metal ions. Although the Ca^{2+} ion dissolution kinetics improved with increasing temperature, carbonation precipitation was retarded at higher temperatures due to reduced CO_2 solubility. That's why it is convenient to operate at mild conditions (T=room temperature).

Dissolution or crystallization of precipitations does not occur instantaneously. Some characteristic time, often longer than the time constant for the overall process, is needed to achieve a new equilibrium.

In nature, calcium carbonate crystallizes most commonly in hexagonal form (as calcite) but also occurs in orthorhombic form (aragonite). Analyzing the results of the carbonation process using steelmaking slag, the carbonates crystallize mainly in calcite ($CaCO_3$). The crystal volume of calcium carbonate is approximately 11.7 % more than that of calcium hydroxide. This means that in general, the pore structures of solid wastes are thought to become finer with carbonation, leading to a solid of lower porosity, and lower pore area with calcite infilling the pore space after carbonation. Therefore, when carbonation occurs it can be expected that after a rapid increase of carbonate mass, there will

be a smaller increase. This is also demonstrated by experimental results, where the curve representing CO₂ uptake during time is steep for the first hours and then become asymptotic.

Alkaline wastes contain lots of different metal oxides, and as reported before, the dissolution of them in water causes an increase in pH. Calcium is not the most soluble, it is preceded by magnesium, and followed by other metals with less relevance for carbonation. The descending order for solubility of metal oxides is as follows: $Mg^{2+} > Ca^{2+} > Zn^{2+} > Cd^{2+} > Pb^{2+}$.

It is interesting to note that for magnesium oxide carbonation, which is present mainly in steel slags, mild operating conditions would not be effective. Indeed, limited $MgCO_3$ formation for carbonation of steel slag under ambient conditions was expected due to the relatively low magnesium oxide content in the slag (with respect to calcium and silicious oxides), low pressure of CO₂ and short reaction times (we remember mild operating conditions was defined as P= 1bar (atmospheric pressure), T=25°C (room temperature)). Typical process conditions for the formation of magnesium carbonation via aqueous carbonation are $pCO_2 > 100$ bar and a reaction time of hours. However, $MgCO_3$ formation could be observed when natural ores (serpentine, olivine) were selected as the feedstock for carbonation, but this is out of concern of this study.

1.2.3 Final considerations

In order to obtain carbonation reactions there are challenges to face. The first one is that the dissolution of calcium species in alkaline solid waste is favored at low pH, however it is not favored for the precipitation of calcium carbonate. Moreover, the Ca^{2+} ion dissolution kinetics improved with increasing temperature as carbonation precipitation (since it follows the Arrhenius law) but this one was retarded at higher temperatures due to reduced CO₂ solubility. After all, carbonation reactions need water to happen, that's why a wet route experiments are preferred. Therefore, this suggests that a well-designed reactor to enhance the mass transfer between the gas, liquid, and solid phases is needed to facilitate the carbonation reaction and increase the carbonation conversion. In the end, finding a balanced operating condition between these two mechanisms is essential for optimizing the overall carbonation process.

1.3 MATERIALS FOR CARBONATION

Both alkali metals (i.e., Na, K, etc.) and alkaline earth metals (i.e., Ca, Mg, etc.) can be carbonated from a chemical elements perspective and a thermodynamic view point. A number of other metals such as Mn, Fe, Co, Ni, Cu, and Zn could potentially be carbonated, but most of these metals are impractical due to their unique and precious features. To provide significant storage of CO₂, large amounts of raw materials are required as feedstock for carbonation, which must be abundant and cheap. One kind of feedstock for accelerated carbonation is natural silicate minerals, such as wollastonite (CaSiO₃), serpentine (Mg₃Si₂O₅(OH)₄), olivine (Mg₂SiO₄), talcum (Mg₃Si₄O₁₀(OH)₂), pyroxene, and amphibole, which are rich in calcium or magnesium content, or industrial residues, the former being abundant but generally difficult to access and the latter relatively scarcer but easily accessed. Although the CO₂ storage capacity of these natural Ca-Mg-silicate minerals is sufficient to fix the CO₂ emitted from the combustion of fossil fuels, the technological carbonation of these minerals is slow and energy demanding. One way to avoid some of these drawbacks is to utilize alkaline waste residues.

The goal of accelerated carbonation is to reduce the time that is necessary for the natural weathering processes, in which CO₂ reacts with metal oxide bearing materials to form stable and insoluble carbonates. Calcium or magnesium oxides are the most favorable metal oxide in reacting with CO₂. Besides that, carbonation is an exothermal reaction, so energy consumption and costs may be reduced by its inherent properties. In all cases, carbonation must provide base ions, such as monovalent sodium and potassium, or divalent calcium and magnesium ions to neutralize the carbonic acid. Other carbonate-forming elements such as iron carbonates are not practical due to their unique and precious features. Industrial residues such as steelmaking slags, combustion residues, waste concrete, fly ashes, etc. are alkaline and also appear to be potential raw materials for CO₂ sequestration by accelerated carbonation due to the fact that these materials are generally rich in metal oxides including calcium, magnesium, aluminum, iron, and manganese oxide. Tab.1 shows the mineralogical composition of these alkaline wastes.

Waste group	Major compounds in Carbonation
Steelmaking Slag	Larnite (Ca ₂ SiO ₄); Brownmillerite (Ca ₂ FeAlO ₅); Lime (CaO); Ettringite (Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ •26H ₂ O); Portlandite (Ca(OH) ₂)
MSWI fly ash	Lime (CaO); Portlandite (Ca(OH) ₂); Ca(OH)Cl; Gehlenite (Ca ₂ Al ₂ SiO ₇)
MSWI bottom ash	Gehlenite (Ca ₂ Al(AlSiO ₇)); Portlandite (Ca(OH) ₂); Ettringite (Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ •26H ₂ O)
Cement kiln dust	Lime (CaO); Portlandite (Ca(OH) ₂); Calcium silicates; Gehlenite (Ca ₂ Al(AlSiO ₇))

Table 1. Major compounds involved in carbonation present for each waste group.

Accelerated carbonation of applied industrial alkaline solid wastes include many potential advantages. For instance, carbonation products such as calcium or magnesium carbonates are thermodynamically stable under ambient conditions, that is, in the absence of acidification. Since these wastes have been largely produced around the world, they offer great availability of deposits. Moreover a carbon capture technology based on these wastes may be useful for these highly emitting industries where alkaline wastes are produced (i.e. cement factories, foundries, incinerators etc.). And then it does not require transport at sites, so it is therefore cost effective. Moreover, products may be beneficially reused in a variety of application, such as construction materials. This also because carbonation eliminates environmental impacts due to decreased leaching of heavy metal trace elements such as Pb, Ni, and Cd from residues and stabilizing of the waste leading to an improvement of environmental quality.

Industrial alkaline solid wastes such as sources of calcium or magnesium oxide are ideal CO₂ sequestration materials due to their availability and low cost. These materials are generally rich in calcium-content and often associated with CO₂ point source emissions, so no mining is needed and the consumption of raw materials is avoidable. In addition, these solid wastes tend to be chemically less stable than geologically derived minerals. Carbonation of industrial solid waste does not generally require the extraction of reactive ions from the solid matrix due to the alkaline-containing silicates, oxides and hydroxides as the mainly reactive phase. Since CaO is one of the most concerned chemical compounds in carbonation, its content can give an idea on the carbon capture of these materials. In Fig.3 the relationship of CaO content with the actual CO₂ capture capacity of various alkaline solid wastes in the literature is displayed. It's pointed out that a higher lime content is related to a greater carbon capture potential. The CaO contents were significantly high in the following wastes: steelmaking slag (30–60% wt. CaO), residues from APC (Ca content up to 35%) and bauxite (4.8% Ca), cement kiln dust (~34–50% wt. CaO), oil-shale waste (CaO content up to 50%), and fly ash (53% CaO) from municipal solid waste incinerators.

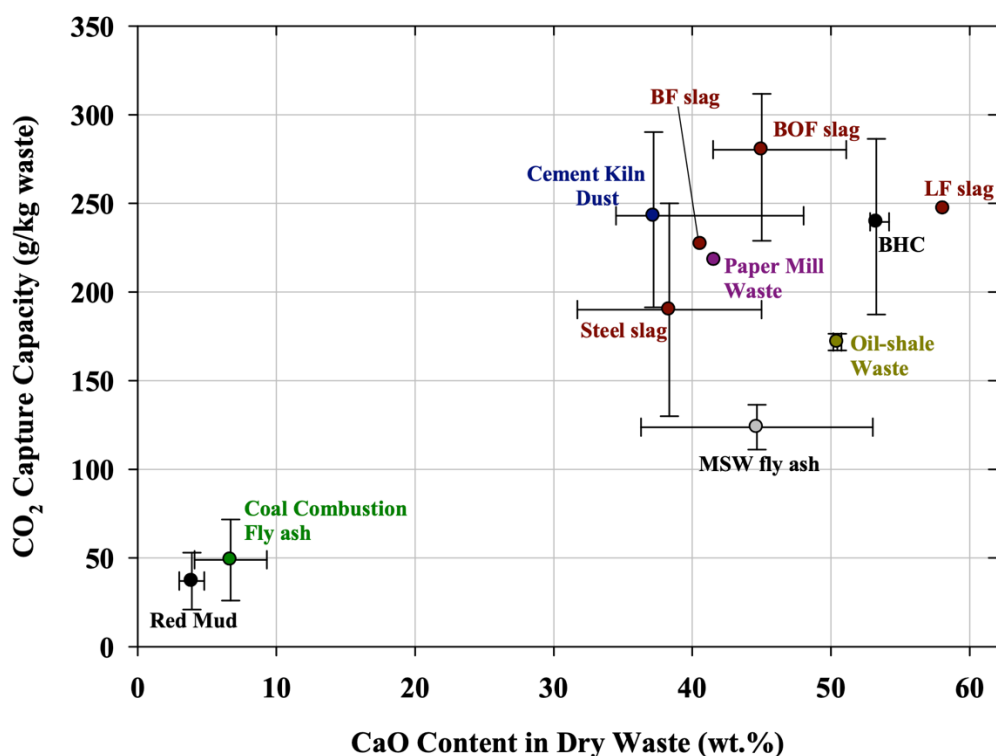


Figure 3. Comparison of actual CO₂ capture capacity with the CaO content in solid for different types of wastes.

1.4 MATHEMATICAL MODELS FOR CO₂ SEQUESTRATION

1.4.1 Theoretical models for CO₂ sequestration

(Steinour et al., 1959) have proposed a formula to calculate the maximum theoretical amount (mass percentage) of CO₂ that can be stored in cement-based materials considering their oxide contents.

In particular it relies on carbonation reactions to synthesized metal carbonates from metal oxides present in cement. It is applied on conventional ordinary Portland cement concrete. They indicated that all the *CaO* present in Portland cement except the one present in the form of *CaSO₄* can be converted to *CaCO₃*, all *MgO* to *MgCO₃*, all *Na₂O* to *NaHCO₃*, all *K₂O* to *KHCO₃*.

For the purpose of the calculations, it was assumed that free water was present and that the solids were freely exposed so reactions could progress as it would if a powder were stirred in a laboratory beaker with water kept saturated with carbon dioxide. Therefore, in dense concrete the reaction would only be superficial.

$$\%ThCO_2 = 0.785 CaO + 1.091 MgO + 1.41 Na_2O + 0.935 K_2O - 0.55 SO_3.$$

Where: *ThCO₂* = theoretical CO₂ storage capacity in mass % *gCO₂/gCement*, *CaO* = *gCaO/gCement* mass fraction of calcium oxide, *SO₃* is the mass fraction of sulfur trioxide, *MgO* is the mass fraction of magnesium oxide, *Na₂O* is the mass fraction sodium oxide, *K₂O* is the mass fraction potassium oxide.

Despite the fact that the Steinour formula was developed to work with cement-based materials, many authors applied it for assessing potential carbon dioxide uptake of different materials which have high alkaline metals oxides content, for instance: (Nam et al., 2012a), (Schnabel et al., 2021), (Chang et al., 2015) applied it to municipal solid waste incineration bottom ashes; (Yuan et al., 2022), (Schnabel et al., 2021) (Pei et al., 2018) applied it to fly ashes from incinerators and power plant; (Schnabel et al., 2021) applied it to steel slags.

The formula has been modified in order to reduce the gap between theoretical and experimental quantities of CO₂ stored measured in laboratory. Indeed, the theoretical equation gives the potential (and so maximum) amount of the CO₂ uptake considering that all alkaline metals oxides would react. Actually, this doesn't happen, as the reactions take place near the surface of the aggregates particles and the formation of calcium carbonate impedes further carbonation. As we will see, there is not correlation between chemical species considered in the Steinour equation modified by the authors and the materials for which it is used.

(Nam et al., 2012a) applied the original Steinour equation for assessing carbon dioxide storage of bottom ashes. They considered chemical composition of municipal solid waste incineration bottom ash can vary depending on the incinerating and operating conditions but decided to apply the original formula previously conceived for Portland cement.

(Schnabel et al., 2021) cites the Steinour Equation by (Huntzinger et al., 2009) for assessing potential carbon dioxide uptake on several materials which they are: steel slags, fly ashes, bottom ashes. They consider that the oxides of *Ca*, *Mg*, *Na*, and *K* undergo carbonation, while the corresponding carbonates as well as sulfur and chlorine compounds (*CaCO₃*, *SO₃*, and *KCl*) lower the CO₂ uptake. They considered that the chemical composition of these materials is different from the one of the Portland cements, so they adapted the formula changing the coefficient of *Na₂O* and *K₂O*, and considering the contribution of *KCl* and of the already present *CaCO₃*.

$$\%ThCO_2 = 0.785 CaO + 1.091 MgO + 0.71 Na_2O + 0.468 K_2O - 0.55 SO_3 - 0.44CaCO_3 + 0.296KCl$$

Where: CaO =mass fraction of the calcium oxide on the total sample mass in %, MgO =mass fraction of the magnesium oxide on the total sample mass in %, Na_2O =mass fraction of the sodium oxide on the total sample mass in %, K_2O =mass fraction of the potassium oxide on the total sample mass in %, SO_3 =mass fraction of the sulfur trioxide on the total sample mass in %, $CaCO_3$ =mass fraction of the calcium carbonate on the total sample mass in %, KCl =mass fraction of the potassium chloride on the total sample mass in %.

The same formula has been considered for assessing potential carbon dioxide uptake on recycled concrete aggregates by (Fang et al., 2017), (Xuan & Poon, 2018); but in this case they modified it by removing the contribution of KCl . So the formula becomes:

$$\%ThCO_2 = 0.785 CaO + 1.091 MgO + 0.71 Na_2O + 0.468 K_2O - 0.55 SO_3 - 0.44 CaCO_3.$$

The same coefficient for K_2O reported by (Steinour et al., 1959.) and (Nam et al., 2012a) was reported also by (Pei et al., 2018)., which used this Steinour equation modified to assess the carbon sequestration of petroleum coke fly ashes. Unlike them, Pei et al. don't consider the contribution of Na_2O , instead they considered the contribution of $CaCO_3$. The formula becomes:

$$\%ThCO_2 = 0.785 CaO + 1.091 MgO + 0.935 K_2O - 0.55 SO_3 - 0.44 CaCO_3 .$$

(Yuan et al., 2022) also modified the Steinour equation, for assessing the CO_2 captured by fly ashes aggregates. But they only considered the contribution of CaO , MgO , $CaCO_3$. So the modified formulation is changed as:

$$\%ThCO_2 = 0.785 CaO + 1.091 MgO - 0.44 CaCO_3 .$$

Similarly to (Yuan et al., 2022), even (Chang et al., 2015) considered only the contribution to carbonation of three chemical species, which are: CaO , SO_3 , $CaCO_3$. They did not considered the contribution of MgO due to the relatively low content of MgO in the bottom ashes. They assessed the carbon sequestration on bottom ashes, and reported the following formula:

$$\%ThCO_2 = 0.785 CaO - 0.55 SO_3 - 0.44 CaCO_3 .$$

If we compare the theoretical formulas applied to assess carbon dioxide sequestration for the same material, we don't find any correlation between chemical species considered by each author and the material itself. For instance considering the fly ashes, compared to the more complete formula of (Schnabel et al., 2021), (Pei et al., 2018) al does not consider the influence of Na_2O and KCl , and finally (Yuan et al., 2022)) It does not consider the influence of Na_2O , KCl , K_2O and $CaCO_3$. The same occurs with the bottom ashes, where (Nam et al., 2012a) consider the influence of CaO , MgO , Na_2O , K_2O , SO_3 , while (Chang et al., 2015) take into account $CaCO_3$ but not MgO , Na_2O , K_2O . In the end we can conclude the authors consider different chemical species with different coefficients for assessing the carbon uptake for the same material. Theoretical models have been collected in ANNEX IV to be completed with quantitative data (%mass of chemical species), thus obtain a $\%ThCO_2$ in mass %.

1.4.2 Semi empirical models applied to recycled concrete aggregates

The theoretical amounts of sequestered CO_2 calculated with the Steinour formula do not depend on others factors other than the chemical composition. However, the carbonation rate and CO_2 uptake of alkaline materials would be greatly influenced by factors as: time of reaction, humidity, temperature, CO_2 partial pressure and concentration of CO_2 , CO_2 flow, water to solid ratio, cement content and particle size.

(Fang et al., 2017) (Kaliyavaradhan et al., 2020) worked on carbon dioxide storage of recycled concrete aggregates. They developed semi-empirical models which fits the experimental results according to the change of process parameters.

1.4.2.1 Kaliyavaradhan et al. model

(Kaliyavaradhan et al., 2020) studied the carbonation effects on recycled concrete aggregates. They proposed a semi-empirical model in which the CO_2 uptake depends on the water to solid ratio and the reaction time. The effects of two process parameters, were evaluated on maximum CO_2 uptake capacity of concrete slurry waste using response surface methodology.

In this study, field of application sees that: the w/s ratio is ranging from 0 to 0.7 and reaction time is ranging from 1 to 168 h, while keeping other factors constant (temperature: 20 °C, relative humidity: 65 %, and CO_2 concentration: 20 %).

Response Surface Methodology is an effective statistical method used for optimizing the process conditions and developing the models. It can evaluate the influence of various experimental factors and their interactions on one or more response variables under investigation. The relationship between experimental variables and one or more response (by the model) variables can be established by a regression procedure using a second-order polynomial equation.

$$[y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \beta_{ij} X_i X_j + \varepsilon]$$

Where: y = predicted response, β_0 = intercept, β_i = linear effect coefficient, β_{ii} = quadratic effect coefficient, β_{ij} = interaction effect coefficient, X_i , X_j = independent variables, ε = residual.

In this study, field of application sees that: the w/s ratio is ranging from 0 to 0.7 and reaction time is ranging from 1 to 168 h, while keeping other factors constant (temperature: 20 °C, relative humidity: 65 %, and CO_2 concentration: 20 %).

This method produced a statistical model to predict the maximum CO_2 uptake:

$$Uptake_{CO_2}(sample) = +5.607 + 90.906 (w/s) + 0.193 (t) - 0.0687 (w/s * t) - 257.917 (w/s)^2 - 0.00206(t)^2 - 0.00172 (w/s)^2 * (t) + 0.0006 (w/s) * (t)^2 + 195.039 (w/s)^3 + 0.0000063 (t)^3$$

Where: $Uptake_{CO_2}(sample)$ = mass of CO_2 sequestered by the dry mass of sample predicted by the model, in %; w/s = water to solid ratio [kg/kg]; t = time [h].

This model showed predicted results near the ones measured in laboratory. The Absolute Relative Deviation (ARD%) was the parameter chosen as a measure of predictability. It is defined as:

$$ARD(\%) = \frac{\text{Experimental value} - \text{Predicted value}}{\text{Experimental value}} \cdot 100$$

It was found ARD to be 0,8% and 3,0% for two samples. Being less than 10% means this semi empirical model is a very effective tool for predict CO_2 sequestration.

1.4.2.2 Fang et al. model

(Fang et al., 2017) developed a semi empirical model by regression analysis of the gathered data, for the CO_2 uptake of recycled concrete aggregates. They tried to find an equation that relates the CO_2 uptake of the sample (from experimental results) and the potential CO_2 uptake of the sample (related to the Steinhour equation), carbonation duration (t), particle size (d), relative humidity, CO_2 concentration ($[CO_2]$) and pressure (P) or flow rate (v) (P for pressurized carbonation and v for flow-through carbonation). A is a factor depending on the carbonation route (batch and pressurized reactor or open and flow through reactor).

The CO_2 uptake (g) presented was normalized to 1 kg of RCAs (noted as $CO_2 \text{ uptake}(\text{sample})$ (g/kg)).

The following general equation expresses the relationship between the CO_2 uptake ($CO_2 \text{ uptake}$), theoretical CO_2 uptake potential ($Potential_{CO_2}(\text{sample})$), carbonation duration (t), particle size (D), relative humidity (RH), CO_2 concentration $[CO_2]$ and pressure (P) or flow rate (Q).

$$Uptake_{CO_2}(\text{sample}) = A * f(t, d, RH, [CO_2], P, Q) \cdot Potential_{CO_2}(\text{sample})$$

According all previous considerations on test conditions, for the parameters including time, relative humidity, size of aggregates, CO_2 gas concentration and carbonation methods (open or closed reactor), their relationships with CO_2 uptake can be written as:

$$CO_2 \text{ uptake}(\text{sample}) = A * f(t, d, RH, [CO_2], P, Q) \cdot Potential_{CO_2}(\text{sample})$$

- **Carbonation potential** $Potential_{CO_2}(\text{sample})$

Natural carbonation starts right after the concrete is prepared. In order to calculate the CO_2 uptake during accelerated carbonation, we need to assess the initial CO_2 uptake before accelerated carbonation, defined as $m_{CO_2}^0$. This step was to set up a reference point, so that the increment in CO_2 uptake is the result of the accelerated carbonation. Accordingly, X^0 represents the mass ratio of the initial CO_2 uptake to cement:

$$X_{CO_2}^0 = \frac{m_{CO_2}^0}{C_m \cdot m_{RCA}}$$

Where: $X_{CO_2}^0$ = mass ratio of the initial CO_2 uptake to cement [%]; C_m = cement mass inside the RCA sample [%]; $m_{CO_2}^0$ = initial mass of CO_2 already carbonated [g]; m_{RCA} = mass of the RCA sample [g]. Here, is set = 1kg.

So, the potential uptake in RCA can be expressed as:

$$Potential_{CO_2}(sample) = m_{RCA} \cdot C_m \cdot (\%ThCO_2 - X_{CO_2}^0).$$

$\%ThCO_2$ is the theoretical maximum CO_2 uptake obtained by the Steinoour formula. This formula was applied to $C_m = 12,11 - 22,47 \%$; $X_{CO_2}^0 = 8,4 - 12,47 \%$.

- **Carbonation extent (CO_2 uptake) by RCA as a function of time**

It is generally known that the relationship between carbonation depth and time of a monolithic concrete is:

$$x_c \propto \sqrt{t}$$

with x_c =carbonation depth, t =time. The above equations are therefore further modified to establish the relationship between CO_2 uptake and carbonation duration:

$$CO_2 \text{ uptake}(sample) \propto \frac{\sqrt{t}}{(a + \sqrt{t})}$$

$CO_2 \text{ uptake}(sample)$ (g/kg) is the CO_2 uptake of 1 kg dry RCAs for a duration of t (hours).

According to their experiments, a = regression constant =1,2; $R^2=0,8$. The field of application of these experiments are 0-24h.

- **Carbonation extent (CO_2 uptake) by RCA as a function of particle size**

The particle size of RCAs affects CO_2 uptake in several aspects. One of them is that larger particles have lower area per unit of mass, meaning that RCAs with larger particle sizes have a slower CO_2 take up rate (per unit mass). The size fractions used in the current study, are: 10–20, 5–10, 2.36–5 and <2.36 mm.

$$CO_2 \text{ uptake}(sample) \propto bD^c$$

Where: D =diameter [m], $B=5,23$ $C=-0,34$; B , C are coefficients obtained by the regressing analysis.

- **Carbonation extent (CO_2 uptake) by RCA as a function of relative humidity**

Past research studies on natural carbonation of concrete indicated that the relative humidity (RH) affected the CO_2 diffusivity as well as the rate of CO_2 uptake. It had been reported that the carbonation rate decreased significantly when the RH was either too high or too low and the optimal RH for carbonation was between 50 and 70%. In this study the effect of RH was studied at three major situations: dry gas ($RH < 5\%$), middle RH ($50 \pm 5\%$), and moistened gas ($RH > 95\%$).

The relationship between RH and CO_2 uptake can be expressed as:

$$CO_2 \text{ uptake}(sample) \propto [1 - e(RH - 0,5)^2]$$

Where: RH = Relative Humidity, $e=4$ regression constant

- **Carbonation extent (CO₂ uptake) by RCA as a function of CO₂ concentration**

The study used the flow-through test to study the 24-h CO₂ uptake under three (CO₂) values (natural (0.03%, 10, 100%). The effect of (CO₂) on rate of carbonation can be expressed as following equation according to regression analysis:

$$CO_2 \text{ uptake}(\text{sample}) \propto [CO_2]^g$$

Where: $[CO_2]$ = carbon dioxide concentration [%], $g= 0,072$ regression constant.

- **Carbonation extent (CO₂ uptake) by RCA as a function of pressure (for batch reactors)**

The carbonation pressure affects the CO₂ uptake. The equation was determined by varying the CO₂ pressure, keeping constant the concentration at 100%. In particular the pressures monitored varied from 1 to 6 bar.

The following equation is to describe the influence of the carbonation pressure on CO₂ uptake:

$$CO_2 \text{ uptake}(\text{sample}) \propto (1 + hP^i)$$

P is the additional positive pressure of the carbonation process ($P=0$ means atmospheric pressure); h, i = Regression constants $h = 0,311$; $i = 0,112$

- **Carbonation extent (CO₂ uptake) by RCA as a function of gas flow rate (for open reactors)**

When using the flow through carbonation test to carbonate RCAs, the gas flow rate Q would affect the CO₂ uptake. A series of tests have been conducted in open reactors where CO₂ was mixed with N₂. The concentration of carbon dioxide was varied from 10-30%. The regression analysis of all three fitting curves suggested an expression for the effect of gas flow rate:

$$CO_2 \text{ uptake}(\text{sample}) \propto jQ^k$$

Where: Q = flow rate of gas mixture [L/min]; j, k = regression constants. $j = 0,74$; $k = 0,18$.

- **Final Formula and model predictions:**

According all previous considerations on test conditions, for the parameters including time, relative humidity, size of aggregates, CO₂ gas concentration and carbonation methods (open or closed reactor), their relationships with CO₂ uptake can be written as:

$$CO_2 \text{ uptake}(\text{sample}) = A * f(t, d, RH, [CO_2], P, Q) \cdot Potential_{CO_2}(\text{sample})$$

Becomes:

$$CO_2 \text{ uptake}(\text{sample}) = A \cdot [1 - 4(RH - 0,5)^2] \cdot 5,23D^{-0,34} \cdot \frac{\sqrt{t}}{(1,2+\sqrt{t})} \cdot [CO_2]^{0,072} \cdot C_m \cdot (\%ThCO_2 - X_{CO_2}^0) \cdot f(P, Q).$$

Where in pressurized and batch reactor conditions $f(P, Q) = 1 + 0,311P^{0,112}$. And in open reactor with gas flow conditions, we have that $f(P, Q) = 0,74Q^{0,18}$.

The value of $A = 0.012$ and $A = 0.01$ are recommended for the pressurized test (batch reactor) and flowthrough test (open reactor), respectively.

$CO_2 \text{ uptake}(\text{sample})[g/kg]$ uptake of the dry mass of the sample. The CO_2 uptake (g) presented was normalized to 1 kg of recycled concrete aggregates, C_m = cement mass inside the RCA sample [%], D =particle size [m], t =carbonation time [h], RH = relative humidity [%], $[CO_2]$ = carbon dioxide concentration [%], P = carbon dioxide additional pressure [bar], Q = gas flowrate [L/min].

Based on the above test data and analysis, the optimal conditions for accelerated carbonation of recycled concrete aggregates are: 100% CO_2 gas concentration with 0.1 bar additional pressure or 10% CO_2 gas concentration with 5 L/min flow rate.

1.5 FURTHER DEVELOPMENTS

1.5.1 Rotating packed bed reactor theoretical model

(Chen et al., 2020) developed a pilot-scale RPB (Rotating packed bed reactor) theoretical model to evaluate the performance of simultaneous removal of NO_x - SO_2 - CO_2 by a flue gas. By utilizing the alkaline solid wastes (fly ash). O_3 Was added to oxidize nitrogen and sulfur oxides. Namely NO , SO_3 and then converted to the higher valence and water solubility species. This is displayed in Fig.5. The high alkalinity and large amount of calcium ions (Ca^{2+}) present in fly ashes cause the CO_2 absorption and precipitation of calcium carbonate (CaCO_3). The schematic of the general process is presented in the following image Fig.4.

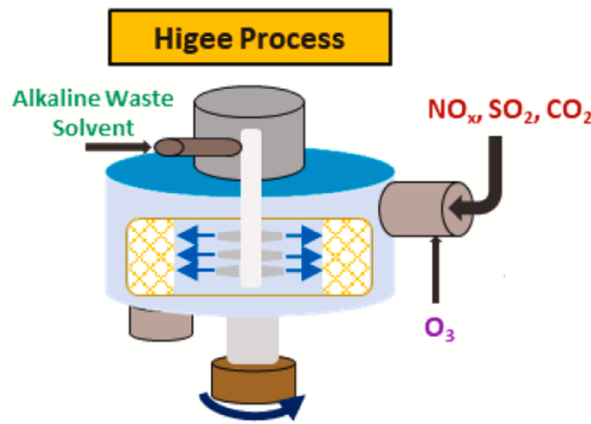


Figure 4. Schematic of the general process

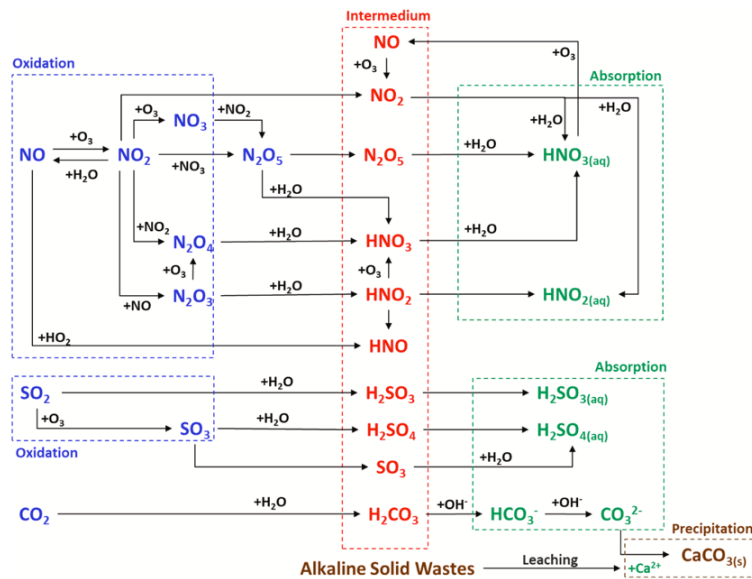


Figure 5. Process chemistry of simultaneous removal of CO_2 , NO_x and SO_3 using O_3 oxidation and alkaline solid wastes.

1.5.1.1 Experimental setup and RPB characteristics

The system was equipped with power, tank, blower, RPB reactor, pump and heat exchanger. The rotation type was horizontal rotation with a countercurrent flow of mixed gas and liquid. The packing zone equipped stainless steel wire as a packing material, had an inner diameter of 0.558 m, an outer diameter of 0.61 m, mean diameter of 0.584 m and axial height of 0.286 m. The weight, density and voidage of the packing medium were 4.5 kg, 7990 kg/m³ and 0.99 m³/m³. The volume of packed bed was 0.007 m³. Six holes of liquid distributor with a diameter of 0.02 m were designed. The designed maximal capacities of gas flow rate and liquid flow rate were 20 m³/min and 0.032 m³/min. A gaseous composition analyzer was used for measuring the concentration of NO_x, SO₂ and CO₂ in flue gas. A schematic of the process flow is displayed in Fig.6.

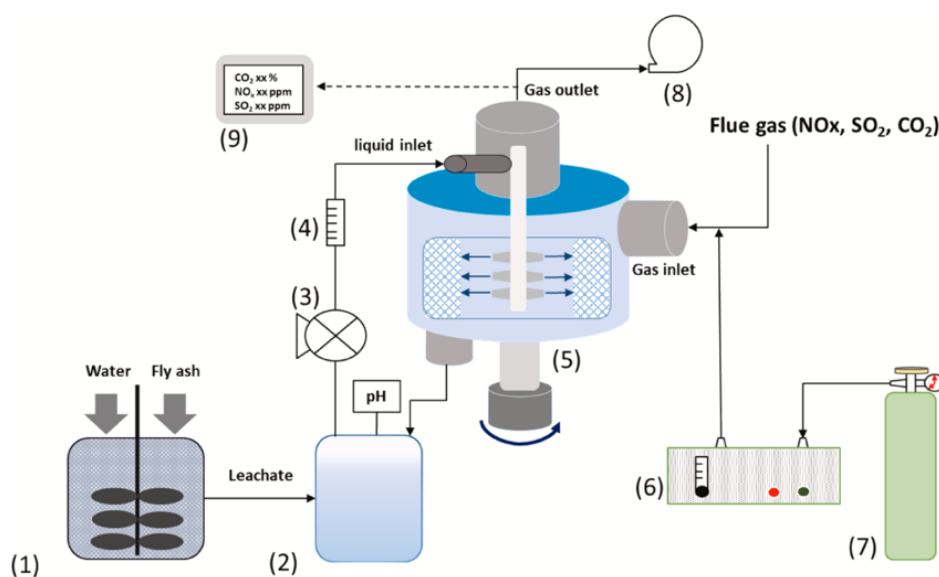


Figure 6. Schematic flow sheet of the process: (1) Mixing tank; (2) Solvent tank; (3) Pump; (4) Flow meter; (5) Rotating packed bed; (6) O₃ generator; (7) Oxygen cylinder; (8) Blower; (9) Gas analyzer

1.5.1.2 Application field

In the flue gas, the major pollutants are NO, SO, and CO₂ with the concentration ranges of 100-220 ppm, 40-300 ppm and 8-15 vol%, respectively. The temperature, humidity and average density of flue gas were observed to be 90-110 °C, 14-20% and 0.92 kg/m³ respectively.

1.5.1.3 Results

The obtained results indicated that the conc. of SO₂ decreased quickly within 150 sec. while the concentration of CO₂ declined slowly within 0 to 450 sec. Afterward, the conc. of CO₂ and SO₂ gradually increased and then remained at a stable value of 11.5 vol% and 37 ppm, respectively. The conc. of NO was abruptly fell down from 122 ppm to 1.3 ppm at 750 to 900 sec. and then remained at 1-5 ppm within 1050 to 2700 sec.

1.5.2 Drum reactors pilot-scale experimentation - MSW BA

(Lombardi et al., 2016) proposed a laboratory scale process to remove by carbonation the CO₂ present in a gas stream through reaction with bottom ashes waste. The idea is to compare the performance of two drum reactors: one has a fixed bed and the other has a rotating one.

1.5.2.1 Experimental setup and characteristics

The experimental facility mainly consists of the laboratory scale fixed bed or rotating drum reactors and the measuring systems. BA is loaded into the reactor and the gas is flowed through the BA bed.

The **fixed bed** reactor (R1) is a 27l stainless steel cylindrical tank (internal diameter: 26.5 cm, height: 66 cm). The reactor can be opened from the top, by means of a semispherical cap, for loading and unloading the BA; the gas seal is ensured by a series of twelve bolts. The gas flows into the reactor from the bottom to the top (upflow configuration). That's visible in Fig.7. Inside the reactor three internal supports are welded every 12 cm. In this way it is possible to decide to place the overall amount of BA in just one layer or to divide it into two/three layers. This feature was added to check if the multi-layer configuration allows better performance with respect to the single layer one. With the three-layer configuration we would like to reduce the possibility for the gas to follow preferential paths through the ash bed, by introducing a plenum between one layer and the other.

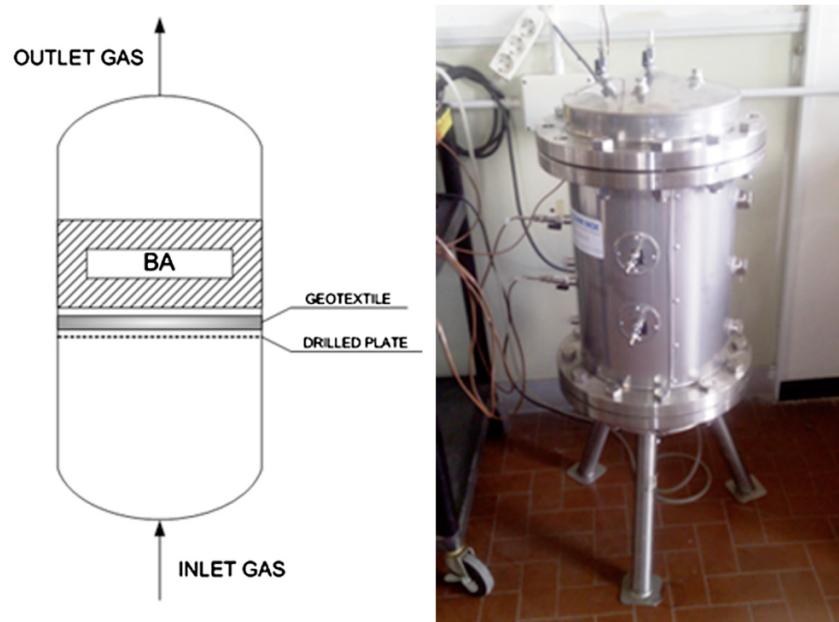


Figure 7. Schematic and external view of Fixed bed reactor

As an alternative to the fixed bed reactor, the rotating drum concept was selected in order to provide continuous mixing of the solid reactive particles with the gas.

The rotating drum concept was selected in order to provide continuous mixing of the solids. The rotating drum reactor (R2) is a 18 L stainless steel cylindrical tank (Fig. 8), with horizontal-axis rotation, provided with thermal insulation (not shown in Fig. 8). The main body has an internal diameter of 21.9 cm and a global length of 43 cm. The gas flows from one side to the other. A large circular opening (with a nominal diameter of 10 cm) with a screw cap is present to easily fill and

empty the reactor with bottom ash. As the reactor rotates, conveyor blades, welded inside the reactor favor the exit of BA at the end of the treatment and promote internal mixing.



Figure 8. External view of the rotating drum reactor

1.5.2.2 Application field

For fixed bed rotatory drum, bottom ashes was sieved under 10 mm and 5mm. Mass of bottom ashes inside the reactor was 6kg that could be divided in 1-3 layers according the selected configuration. The flow of 100% CO₂ was fixed to 400 Nml/min. While for for rotating bed rotatory drum: the amount of BA used for each test was equal to 3-6-9 kg according to the filling ratio selected. Bottom ashes was sieved under 10 mm to remove inerts. Flow of 100% CO₂ of 200-400-600 Nml/min was tested. Three Filling ratio were: 10-20-30%. And rotation speed of 2.5-5.0 rpm was selected. In both cases the time duration of the tests in the range of 8–14 h.

1.5.2.3 Results

According to the various reactor configuration and variables, various results have been achieved.

- Fixed bed reactor: achieved an uptake of 42-47 gCO₂/kg (with bottom ash material from sample lot A) and 27-35 gCO₂/kg (with bottom ash material from sample lot B) through a configuration of 3-layers with 10 mm thickness. When the thickness was changed to 3 layers 5 mm thick, the CO₂ uptake ranged 45-48 gCO₂/kg (for lot A) g/kg and 34 (for lot B).
- Rotating bed reactor: achieved an uptake of 19-23 gCO₂/kg with FR (filling ratio) =30%; 2.5 rpm and sample media 21g; 25-31 gCO₂/kg FR=20% 2.5 rpm media 28g; 33-37 gCO₂/kg FR=10%. 2.5 rpm media 35g; 23-39 gCO₂/kg FR=20%. 5 rpm media 26g.

It is clearly observed that working with a lower filling ratio is beneficial allowing an improved contact between BA and the gas flow

1.5.2.4 Conclusions

For fixed bed reactor, no significant differences were observed between the carbon dioxide specific removal average trends obtained for the different conditions. Trends related to smaller particle size (i.e. 5 mm sieving) are slightly better than those related to larger particle size (i.e. 10 mm sieving). While comparing different layers configurations, the multiple layer arrangement seems not to influence significantly the process performances. While comparing fixed and rotating bed conditions, from this comparison The rotating drum reactor is able to provide improved carbon dioxide removal with respect to the fixed bed reactor. It is clear that the lower filling ratio allows better performances, while the rotating speed variation effect does not seem to be very significant. Comparing the carbon dioxide specific removal achievable by using the rotating reactor in the best operating conditions (35–37 g/kgBA) with that measured for the fixed bed reactor (21–23 g/kgBA), an increase of about 61–66% is observed. The most merging limit of the technology is that: The CO₂ uptake is more influenced by the characteristics of BA rather than by the reactor operating conditions.

1.5.3 Rotating drum reactor pilot-scale experimentation - Biomass BA

(Schnabel et al., 2022) developed and tested a rotating drum reactor integrating hydration and carbonation of biomass bottom ash. In the carbonation experiments, the influence of: rotation rate, fill level and moisture content on the bed motion was studied. The reactant gas was 10 vol% CO₂. It was fed either simultaneously with or subsequently to humidification.

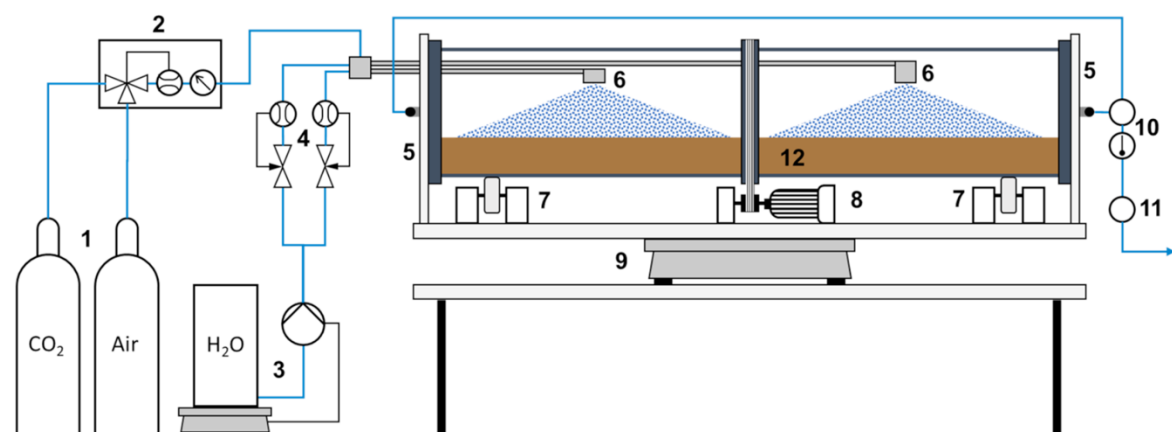


Figure 9. Setup of the carbonation experiments with axial section of the reactor. (1) CO₂ and compressed air supply, (2) gas mixing station for constant flux and concentration of CO₂, (3) water reservoir on a scale for acquisition of the water flow rate and adjustment of peristaltic pump, (4) flowmeters to adjust an equal supply to both nozzles, (5) static front plates equipped with v-seals and gas outlets, (6) two- fluid nozzles for supply of water and reactant gas, (7) bearing wheels, (8) variable-speed motor with drive sprocket, (9) scale measuring the mass of the reactor, (10) temperature and relative humidity sensor, (11) CO₂ sensor, (12) biomass bottom ash or quartz sand.

1.5.3.1 Experimental setup and characteristics

The carbonation experiments were conducted in a rotating drum reactor. The reactor was a stainless-steel cylinder with an inner diameter of 0.3 m and a length of 1.5 m running within v-seal rings in static front plates. The drum was driven by a motor with variable speed. Four stainless steel pipes were conducted through one front plate feeding two two-fluid nozzles. The nozzles served for spraying water and feeding gas into the reactor. They were positioned at 0.38 m and 1.13 m from the reactor front-end and directed to the material bed with a flat spraying characteristic (spraying angle

80° to 130°, depending on the fluid pressure). Gas outlets were integrated in the reactor front and back ends. The exhaust was routed to a flow-through cell equipped with sensors for temperature, relative humidity and CO₂ with tolerances of ± 0.5 °C, $\pm 4.5\%$ RH and ± 0.5 vol% CO₂. Temperature and humidity were recorded every minute and CO₂ concentration was logged every 10 s. The schematic of the process is displayed in Fig.9.

The reactant gas was continuously supplied by a gas mixing station mixing CO₂ and compressed air. Water was supplied by a peristaltic pump to both nozzles. The water supply was determined gravimetrically by weighing the water reservoir and recording the mass every 10 s. The water supply to the nozzles was monitored with two flowmeters and adjusted by a valve. The latter ensured equal flow through each nozzle.

1.5.3.2 Application field

The gas was CO₂ at fixed concentration of 10% v/v. The fill level (vol% of the void volume) varied between 5 and 20 vol%. Reactor rotation rate (in rpm) varied 0.5 - 7 rpm and L/S ranged between 0.0, 0.1, 0.2. Mass tested was 19.1 kg dry matter corresponding to a fill level of 20 vol%. The gas flow rate was kept at 50 L/min (referring to standard conditions, i.e. dry gas, 101.325 kPa, 0 °C), and it was fed for a total of 120 min. Considering the flow rate, CO₂-level, duration of feed, and reactor loading the cumulative CO₂ supply was 62 g CO₂/kg bottom ash. Bottom ash material was sieved with 2mm mesh seize.

1.5.3.3 Results

Overall, the CO₂ uptake by BBA varied between 22 and 31 g/kg, which is far from full conversion of the alkaline components predicted from the elemental composition of the material. The bottom ash had a significant carbonate content already before the carbonation (31–40 g/ kg) which increased to 42–76 g/kg after carbonation. The material sequestered 1/3 to 1/2 of the total CO₂ supply (62 g/ kg). The favorable bed motion was identified at a rotation rate of 7 rpm and a fill level of 20 vol%. The maximum CO₂ uptake of 31 g/kg was achieved at a moisture content of L/S 0.1 within a reaction time of 2 h.

1.5 CRITICAL REVIEW

1.6.1 Methodology adopted for critical review

1.6.1.1 Premise

This bibliographic research was carried out for a master thesis of Environmental Engineering course of Università degli studi di Padova. It is part of an activity in which the bibliographic research is followed by a critical analysis of notable results and these will be utilized as a basis for experiments carried out in laboratory.

The investigation allowed to quantify the potential carbon dioxide sequestration by alkaline wastes despite the variety of methodologies through which the experiments were carried out.

1.6.1.2 Objective

Assess the potentiality of carbon dioxide capture of alkaline waste materials, find methodologies to carry out a laboratory experimentation, understand which parameters influence carbonation.

1.6.1.3 Analysis methodology

The methodology adopted for the systematic bibliographic analysis is the one defined by “*The PRISMA 2020 statement: An updated guideline for reporting systematic review*”. (Page et al., 2021).

The execution of this methodology allows to identify in a rigorous way, a list of articles present in the scientific literature, coherent with the intended objective. The identification of the list of items is obtained through a step-by-step procedure, based on the following phases:

- Identification;
- Screening and Eligibility;
- Inclusion.

This methodology is illustrated by figure 1.

Identification

The following key-words have been used for the bibliographic research:

(“*Accelerated*” AND “*carbonation*”) AND (“*aggregates*” OR “*fly ashes*”) AND (“*CO₂*”).

This means that the key words *Accelerated carbonation* and *CO₂* have to be present as well as one of the words word *aggregates* or *fly ashes*.

The search for words was limited to their presence in the title of the publication, in the abstract or in the keywords of the article.

Database consulted was Scopus. The research was concluded in the March of 2022. 194 records have been found. Of these, 6 records have been removed before screening. The reasons are the: fact that they cannot be downloaded, they were duplicated, or they were available only in Korean language. Moreover, 17 records have been identified from other databases, which are: ScienceDirect, researchgate.net or papers cited by authors of papers already included.

Screening and Eligibility

In the screening phase, all abstracts of the papers have been read. The ones which do not concern with the aim of this study have been excluded. 146 papers have been excluded, the reasons are: aim of paper out of scope, the study utilizes temperatures too high and out of our concern, the paper focus on mechanical, physical or chemical properties. For instance the paper focus on carbonation depth (physical property) or to compression strength (mechanical properties).

42 papers resulting from scopus research and 17 from other databases have been assessed for eligibility. In the eligibility phase, 20 reports coming from scopus have been removed. The reasons are: quantitative CO₂ capture was not assessed, or formulas to convert CO₂ uptake were not present. From 17 papers coming from other databases, 5 of them have been excluded, the reasons are: paper results cite other authors already analyzed, or paper results expressed in terms of carbonate content (for instance through total inorganic carbon content parameter), there were not equations to convert results in $g^{CO_2}/kg \text{ sorbent}$.

Inclusion

In this phase, 22 papers coming from scopus and 12 coming from other sources have been fully analyzed and included in this study.

1.6.2 Papers analyses and data collection

Then the included papers have been analyzed through the following features: typology of reactor, sorbent typology (i.e. the substrate material), experimentation routes and variables (wet or dry route, Liquid to solid ratio applied, relative humidity, CO₂ concentration, CO₂ flow, temperature, pressure, granulometry), operative conditions (mild or intensive), scale of the experiment (mass of the sorbent and time of exposure), characterization of the sorbent (typology of analyses conducted), methods to assess the carbonation uptake, results (gCO₂/kg dry weight), objective of the paper. These features have been collected in a table, for more informations, see ANNEX I, ANNEXIII.

Then a quantitative analysis have been carried out, to compare numerically the features collected in the previous research step, and it is available in ANNEX III. At last, the schematic of the experimental setup utilized in literature was collected, they are visible in ANNEX VII.

1.6.2.1 Reactors classification

Reactor division was chosen between open and closed reactor configuration. Closed reactor configuration represents a reactor in whose chamber the gas exchange is prevented. Before injecting gas inside, the chamber was vacuumed by a vacuum pump. Concentration and pressure of gases inside can be changed before closing the valves. Open reactor configuration is composed by a chamber connected with a vacuum pump and gas cylinders in which gas flow was controlled by a flowmeter. Carbonation chambers are equipped with P, T sensors, inlet and outlet valves, pressure gauge.

1.6.2.2 Materials for accelerated carbonation classification

Materials in which accelerated carbonation tests have been carried out have been classified in eight classes.

- Fly Ashes class includes granulated fly ashes from incinerators so APC (Air Pollution Control) residues, and ashes from MSW (municipal solid waste) combustion processes; fly

ashes from coal fired power plant; fly ashes from hydrocarbons fired power plant; fly ashes from biomass combustion; fly ashes from carbon coke and petroleum combustion. 9/34 of the papers that have been included in this study worked on this material.

- Fly ashes-monolites class includes petroleum coke fly ashes is added as supplementary cementitious material in blended cement mortars. Have been found 2 studies on this material.
- Slags class includes granulated steel slags from EAF (electric arc furnace) furnace; stainless steel slags; granulated blast furnace slag (iron slags); steel slags from Basic Oxygen Furnace. Have been found 5 studies on this material.
- Bottom ashes class includes granulated bottom ashes from MSW incinerators, bottom ashes from biomass combustion. Have been found 3 studies on this material.
- Natural aggregates class includes siliceous aggregates, not other specifications about their origin. Have been found only one study on this material.
- Concrete-monolites class includes sample of concrete prepared according to classical mix design (water, cement, aggregates) with expanded slags aggregates; Portland limestone cement paste, without aggregates. Have been found 3 studies on this material.
- RCA class includes Recycled Concrete aggregates: recycled concrete aggregates produced by crushing old concrete debris; RCA obtained from crushing a designed concrete mix produced by a ready mixed concrete plant; RCA obtained from crushing laboratory concrete cubes; RCA derived from demolition concretes, which are crushed on recycling plants, RCA obtained from crushing old foundation piles. Have been found 8 studies on this material.
- RCA-Monolite class includes concrete blocks made up by water, cement, and RCA as aggregates. Have been found 2 studies on this material.

1.6.2.3 Scale of the experiment

Experimentations carried out in previous studies were performed in laboratory scale. In particular, time range was 0.5-168h, where times over 24h experiments have been carried out mostly on recycled concrete aggregates and concrete monoliths to assess the effects of carbonation on the sample. 13/34 studies performed test on range time 0-2h; 10/34 on range >2-8h; 15/34 on range 8-24h; 7/34 on time >24h. obviously many studies carried out tests on different times. Sample mass ranges 1-6000g, where masses over 300g generally are associated to concrete monoliths samples. 3/34 studies found in literature worked on range 44mg-10g mass sample; 4/34 on 11-100g; 5/34 on 101-300g; 3/34 on 301-500g; 3/34 over 500g. it's important to note that in 21/34 cases mass of sorbent was not found.

1.6.2.4 Operations route and variables

Accelerated carbonation treatment have been carried out through different operations route and variables. Experimentation route has been divided in dry and slurry, where in dry route experimentations, sample material has been dried in oven at 105°C for 4 hours before to be exposed to carbonation; while in slurry route sample material has been mixed with demineralized water and stirred to achieve the desired liquid to solid ratio (L/S in L/kg) before to be exposed to carbonation. 20 on 34 included studies worked with an applied L/S=0. 14/34 studies worked through the wet route, with different L/S ratios, which range found is 0,1-3 L/kg. Relative humidity range in the chamber was 0.04-100%, and it was maintained by solutions of $NaCl$, $Ca(NO_3)_2$, $Mg(NO_3)_2$. CO_2 flows

range was 0.01-10 L/min. 7/34 studies performed carbonation through an open reactor. The carbonation experiments were performed with a pressure range of 1-9 bar inside the static chamber. 25/34 studies included in the review carried out tests with atmospheric pressure condition, while 15/34 utilized a pressure greater than 1 bar. Granulometry of materials varied according to the typology of material. Fly ashes displayed the lowest values and ranged from a minimum value of 0.0000152 mm to 10mm. Bottom ashes ranged 0.125-10mm. RCA granulometry ranged 1-20mm. Slags granulometry ranged <0.1-9.53mm.

1.6.2.5 Characterization of material sorbent

Characterization of sorbent found in literature varied greatly according to the objectives of the studies. Physico-chemical characterizations found in literature include: Leaching test according UNI EN 12457-2, TC (total carbon content), LOI (Loss on ignition), XRD (X-ray diffractometric), TG (Thermogravimetric), SEM (scanning electric microscope) analyses, GC (Gas Chromatography), ANC (acid base neutralization) tests, XRF (X-ray fluorescence microscopy), DSC (differential scanning calorimetry), moisture content, water absorption capacity, pH and electric conductivity analyses, particle density analyses, SSA (specific surface area).

1.6.2.5 Results

The CO_2 uptake results from the analyzed papers were converted into the $gCO_2/kg \text{ dry sorbent}$ unit of measurement through the formulas required depending on the specific case study. They have been organized in box plot according to their operative conditions. They are visible in paragraph 2.1.3 and in ANNEX V, VI.

1.6.2.6 Methods to measure carbonates content

The most utilized method to measure carbonates content was TGA (15/34) coupled with DTG (differential thermogravimetry analysis) or DTA (Differential thermal analysis), followed by Mass gain method (5/34) and calcimetry analyses (3/34), XRF measurements (1/34), ideal gas law (2/34), inorganic carbon measurements (2/34).

1.6.2.7 Operative conditions

Operative conditions have been classified according to the values of temperature (T), pressure (P), and % CO_2 concentration in “very mild”, “mild”, “intensive”, “very intensive”. Which correspond respectively to T=room temperature < 35°C, P=1bar and CO_2 =0-20% for the first category; to T<35 °C, P=1 bar and CO_2 =20-100% for the second one; to T>35°C or P>1 and CO_2 < 20% for the third and to T>35 °C or P>1bar and CO_2 =20-100% for the last one. Temperature range found in this research is 20-400 °C, while pressure range utilized was 0.1-9 bar. Higher pressure tests (8-9 bar) usually are coupled with high temperature (T>60°C) and have been carried out especially for fly ashes and steel slags materials.

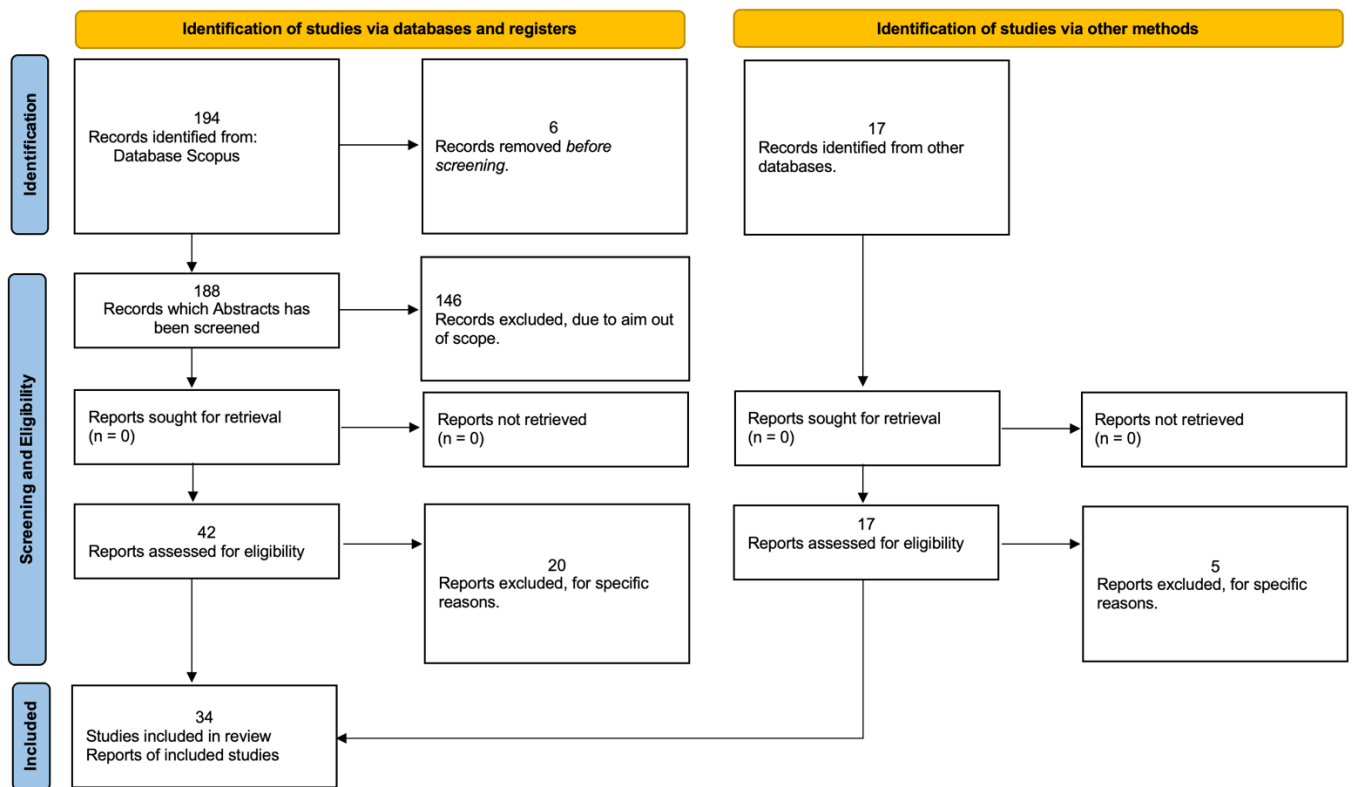


Figure 10. PRISMA 2020 flow diagram for new systematic reviews which included searches of databases, registers and other sources.

PART II – SCIENTIFIC PAPER

“Accelerated carbonation of alkaline waste: from a bibliographic review to an experimental assessment to evaluate the CO₂ capture potential”

2.1 INTRODUCTION

2.1.1 The role of the carbon accelerated carbonation in the Climate change and carbon neutrality policies

According to latest IPCC report [IPCC, 2021] The increase in global mean surface temperature, which reached 0.87 ° C in 2006-2015 compared to 1850-1900, has increased the frequency and magnitude of shocks, increasing evidence of how a temperature increase of 1.5 ° C or more could impact natural and human systems. Without a marked acceleration in emissions cuts, the plans to achieve the climate goals set in Paris in 2015 are linked to the possibility of capturing and storing excess carbon dioxide (CO₂) emissions in the atmosphere.

The net-zero emissions target is part of the measures designed to limit global warming, as underlined in the text of the Paris Agreement on climate change in 2015 and set as a goal within 2050 by the European commission.

This concept includes not only the reduction of greenhouse gases emissions, but also the capture of those that will inevitably continue to be released into the atmosphere.

Therefore, it is necessary to capture the CO₂ emissions produced in excess by human activities (such as from power plants, waste thermal plants and heavy industry) and then store them utilizing merging Carbon Capture and Storage (CCS).

Among CCS methods, the accelerated carbonation process has recently captured the interest of more and more researchers. The aim of the process is to store gaseous CO₂ in a stable form making it react with alkaline earth-metal oxides to precipitate stable carbonates (Pan et al., 2012).

Considering that 1,076,515 tons of bottom ashes and non hazardous slags have been produced in Italy in 2019 and assuming an average absorption of 20 gCO₂/kg dw then 21530 tons of carbon dioxide per year could be fixed. While increasing the scale, in UE in 2019 and 2020 800 million tons of C&D waste and 19 million tons of bottom ashes have been produced, with the previous capture assumption, a technology based on this process can mineralogically fix 16.38 million tons of CO₂.

2.1.2 The accelerated carbonation in the field of waste residues

Carbonation occurs in natural conditions, but at those conditions it is a slow process, involving the reaction of CO₂ with metal oxides present mainly on the material surface. This is because the concentration in the atmosphere of carbon dioxide is 0.04% v/v and the materials rich in alkaline-earth-metals oxides are not sufficiently porous to guarantee the diffusion of co₂ to react with all reactive species present in the bulk. Thus, natural carbonation requires long periods (even years) to exploit its whole carbonation potential. Therefore, recently, many studies have focused on accelerating the carbonation process duration. In present times, the research focuses on assessing and maximizing the CO₂ uptake by optimizing the variables which affect the process such: materials and

granulometry, moisture content, temperature, pressure, gas flow rate, CO₂ concentration and liquid-to-solid ratio applied. (dos Reis et al., 2020) (Schnabel et al., 2021) (Jiang et al., 2013).

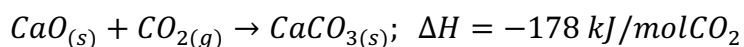
2.1.3 Accelerated carbonation materials

Materials particularly suitable for accelerated carbonation are alkaline wastes. Metallurgical slags, bottom incineration ashes, fly ashes from air pollution control devices and cement-based construction & demolition waste are generally rich in Ca and Mg oxides, hydroxides and silicates which make the material alkaline. Construction and demolition waste is one of the macro items of the European Waste Catalogue (EWC). It accounts for 37.1% of all waste generated in Europe in 2020, corresponding to about 800 million tonnes, making construction and demolition activities the largest source of waste in UE [EC Europe]. In Ue during 2018 approximately 19 million tons of bottom ash was produced [cewep.eu]. Looking worldwide about 130 million tons of various steel slags are produced every year (Schnabel et al., 2021).

Accelerated carbonation treatments of alkaline wastes can bring other benefits than the carbon dioxide capture. It can increase the environmental quality of the residues by reducing the release of heavy metal oxides (Cappai et al., 2012). And improve the mechanical properties of aggregates for a possible utilization as construction material (Gunning et al., 2009) (Pei et al., 2018). Furthermore, developing a full-scale CO₂ capture plant applied to large carbon dioxide emitters such as incinerators, foundries and cement factories utilizing their same wastes, would mean that, in addition to being a tool for achieving carbon neutrality, the method for capturing the carbon dioxide through accelerated carbonation is also a process inserted in a circular economy context.

2.1.4 Carbonation Chemistry

Natural carbonation occurs such that the alkaline earth-metal oxides present in the solid phase react with CO₂ to form earth-metal carbonates. The process requires the presence of water to occur and is based on acid-base reactions in which carbonate acid is neutralized by a base (alkaline earth-metal oxide). Due to the wide range in composition of alkaline wastes, oxides like *CaO*, *Ca₂SiO₃*, *MgO*, *K₂O*, *Na₂O* react with carbon dioxide and contribute to the carbon sequestration. Since lime is one of the most abundant metal-oxides in alkaline wastes (Pan et al., 2012) and calcium carbonate is one of the most carbonates concerned in carbonation studies, its reaction with carbon dioxide is given below. The general formula that summarizes the entire calcium carbonate formation process states that lime (*CaO_(s)*) reacts with carbon dioxide according to the following reaction:



Since enthalpy has a negative value, the reactions are exothermic, so it produces heat. According to Le Chatelier principle, if temperature increases, the product formation is inhibited.

Scientific literature identifies the following factors as influencing factors for carbonation: L/S ratio, CO₂ concentration, pressure, temperature, time of treatment.

Carbonation is a phenomenon that is influenced by the moisture content presented in the aggregates as reported by (dos Reis et al., 2021). That's because moisture influences the permeation properties of the aggregates, indeed water presence is necessary for the oxides hydration. They reported that higher water-content than 8% can decrease the carbon dioxide uptake. Indeed, water can fill material pores which can slow down the access of CO₂ that occurs by diffusion.

Many authors performed carbonation tests through a wet route and demonstrated its effectiveness. In particular, the range 0.2-0.3 for the L/S ratio was demonstrated as the most effective by (Bacocchi et al., 2009) concerning carbon capture in aqueous route. As a matter of fact, (Bacocchi et al., 2009) found that liquid to solid ratios higher than 0.3 decreased the carbon dioxide uptake for fly ashes.

This probably due to the fact that CO₂ diffusion in the water layer decreases when increasing the water quantity. This optimal range of liquid to solid ratio was reported even by (Ukwattage et al., 2015)(Schnabel et al., 2021) (Nam et al., 2012b). Most of the experiments found in literature have been carried out with 100% concentration of CO₂.(Zhang & Shao, 2016); (El-Hassan & Shao, 2014). However, (Sereng & Dangla, 2020) performed accelerated carbonation on recycled concrete aggregates with “very mild” operative conditions (static chamber, atmospheric pressure and 15% CO₂ concentration) and stated that CO₂ concentration impacts the uptake. Indeed, the higher the concentration, the greater its storage. But the increase in concentration over 15% is not significant on CO₂ uptake. Even (Fang et al., 2017) found that the effect of CO₂ concentration on the uptake becomes insignificant after (CO₂) has reached value typically over 20%. Moreover, according to (Baciocchi et al., 2009) results, fast kinetics can be obtained with a flow with 10% CO₂ concentration. Regarding pressure, (Fang et al., 2017) have found that a moderate increase in CO₂ pressure (0.1-1 bar) inside the reaction chamber, significantly increased the carbonation rate. Otherwise, a further increase of 2-4 bar only a slight improvement in CO₂ uptake was observed. Many authors (Rostami et al., 2011; Schnabel et al., 2021), (Fang et al., 2017) found that the carbonation rate is higher in the initial hours and then gradually decreased until approached a stable CO₂ uptake level. In fact, the studies that aimed to evaluate the absorption of CO₂ had a duration of less than 24h (Chang et al., 2015) (Lombardi et al., 2016)(Nam et al., 2012b). Instead, those who had as objective to understand the carbonation of concrete and how to improve its mechanical properties (Kaliyavaradhan et al., 2020) (Suescum-Morales et al., 2021) carried out carbonation tests with duration greater than 24h.

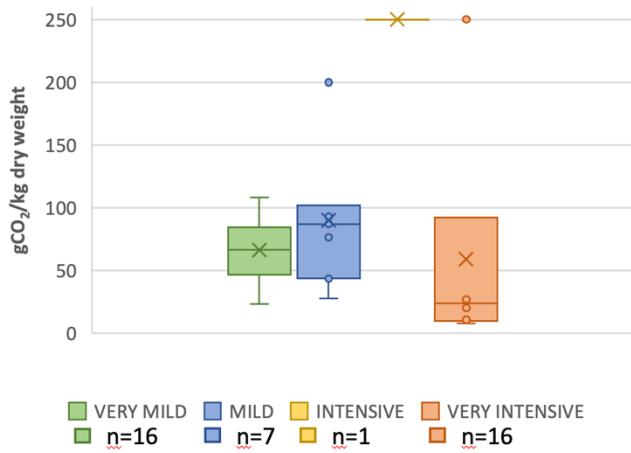
2.1.3 Critical review

For the purpose of this study, it was decided to conduct a systematic review according to the methodology defined by “*The PRISMA 2020 statement: An updated guideline for reporting systematic review*”. (Page et al., 2021).

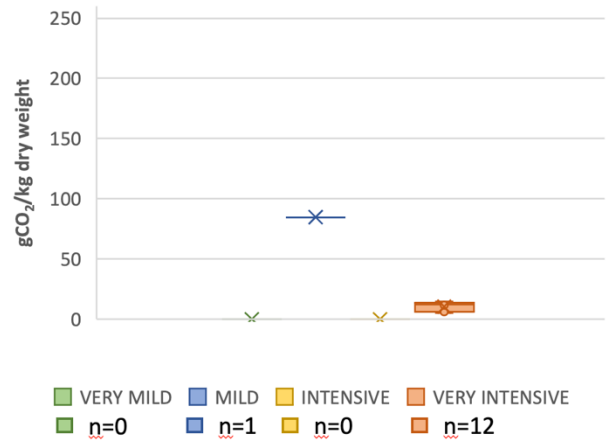
From this bibliographic analysis, it's pointed out that there is no standardized method to carry out the accelerated carbonation treatment. Many kinds of operation routes, variables and measurement methods have been tested. Despite the fact that the most used carbonate measurement method is TGA, it is highlighted that this has the limit of measuring a very limited portion of the test, and having to analyze wastes, there is a great risk of error given the heterogeneous nature of the material. It should be noted that the studies were often incomplete for some information, in particular data for particle size are missing in 8/34 cases; sample mass 21/34 cases.

In Fig.11 results of carbon uptake for the different material classes and for the different operative conditions are shown. Are also shown the number of records found per box plot.

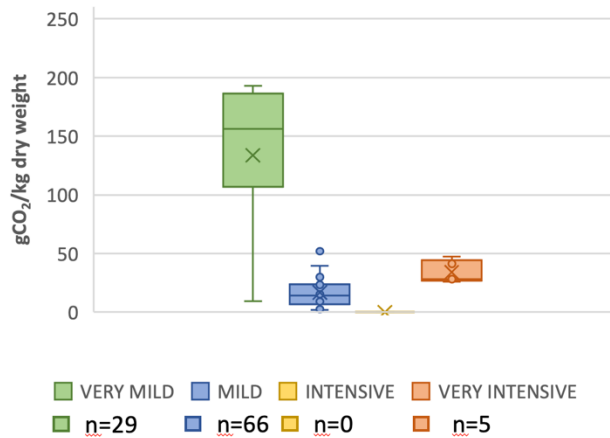
FLY ASHES (granular form)



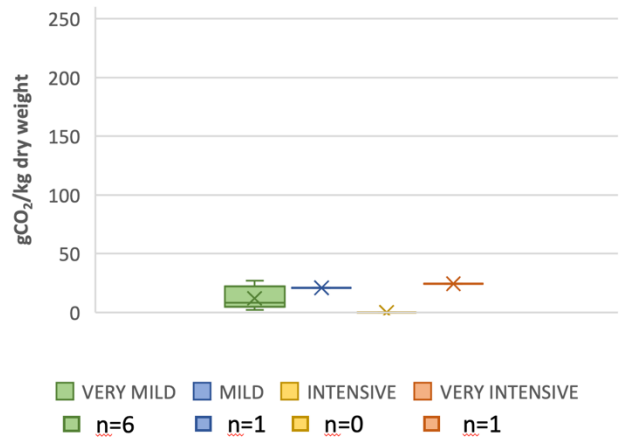
FLY ASHES (monolithic form)



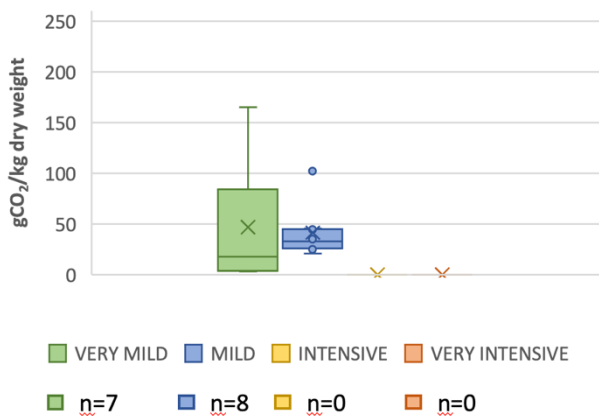
RCA (granular form)



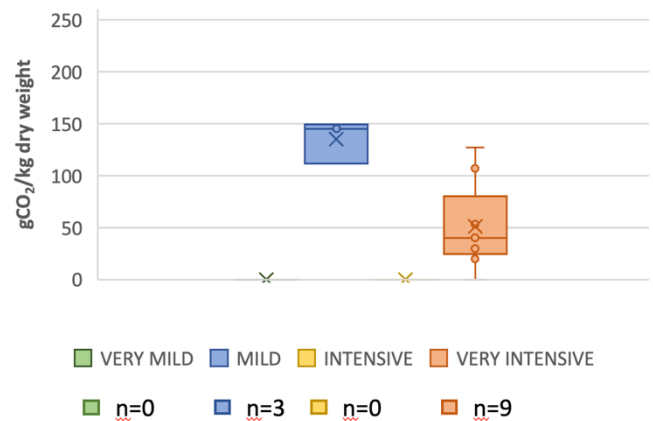
RCA (monolithic form)



BOTTOM ASHES (granular form)



SLAGS (granular form)



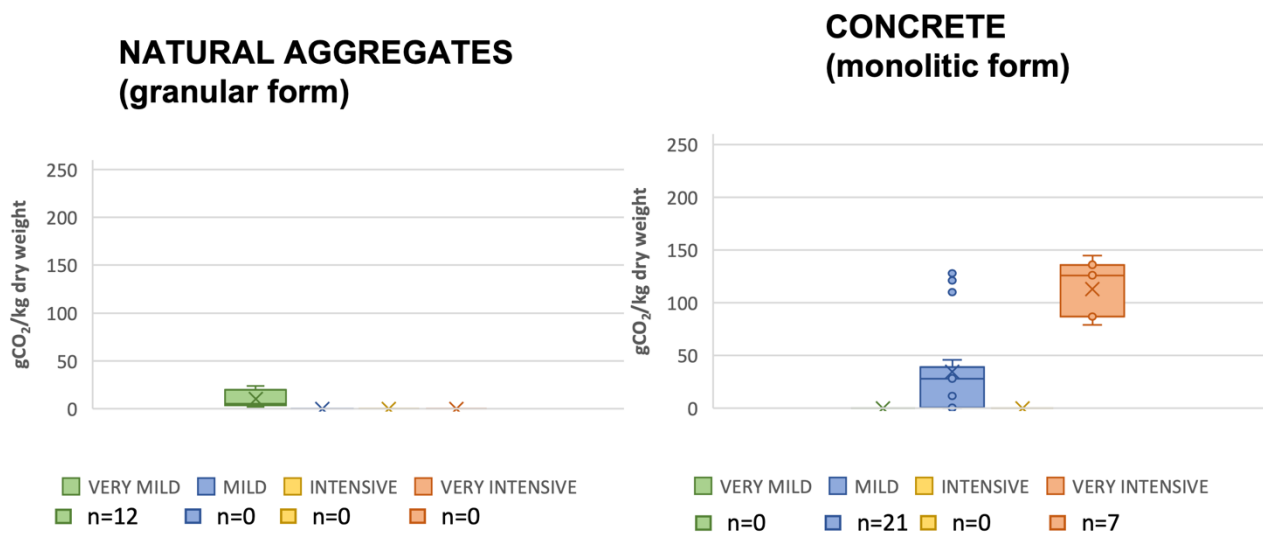


Figure 11. Results of CO₂ uptake after accelerated carbonation treatment of different classes of materials found in literature. n=x is the number of data found.

The Fig11. highlights the following notable results:

- From the statistics the CO₂ sequestration values into the sorbent materials cover a large range, demonstrating the variability and conditions analyzed in the experiments. The maximum value of CO₂ captured was found for granular fly ashes, 250 and 200 gCO₂/kg_{dw}, in mild, intensive, and very intensive operative conditions respectively.
- RCA granular is the material for which most data have been found, for a total of 100, in 8 on 34 included studies on which the research was conducted. Data found for very mild conditions 29 range 9.5-192.7 gCO₂/kg_{dw}; while in mild conditions 66 data range 2.076-55 gCO₂/kg_{dw} which seems more consistent.
- Moreover, RCAs granular seem to capture better in very mild conditions. But it must be noted that very mild conditions plot is influenced by 23 on 29 total results of experiments in which the carbonation treatment lasted 168h. However, the criteria utilized to build the plot did not consider the carbonation duration.
- The material is able to capture CO₂ better when it is in granular form, as it is visible for recycled concrete aggregates and fly ashes. This due the fact that in granular form the specific surface is greater than in the monolithic one, thus the diffusivity of CO₂ inside the material is improved.
- According to this bibliographic research the best material according to the carbon dioxide potential is the granular fly ashes. This due the high presence of alkaline oxides presents inside.

The high range of values recorded by the papers is underlined. Results are characterized by a great variability, because the parameters which influence the CO₂ uptake (time, L/S ratio, reactor, granulometry, mass of the sample) were not taken into consideration in the plots construction. The fact that there is no standardized method to perform accelerated carbonation and evaluate the absorption of CO₂ can explain the great variability of the results.

2.1.4 Introduction to our experimental study

This study investigates the possibility of applying the accelerated carbonation process on a mix of alkaline wastes with the aim of assessing their potential for CO₂ sequestration. Different conditions have been tested to provide the basis for a future scale up of the process on the materials used. For the purpose of this work, several accelerated carbonation tests were performed at three different grain sizes (0/6mm, 6/16mm and 16/31mm) according to 3 different liquid to solid ratios (L/S=0.0 L/kg, L/S=0.2 L/kg and L/S=0.3 L/kg), for three different times (2, 4, 8h) to 4 different masses (200g, 100g, 50g, 25g) under fixed operating conditions (relative humidity, CO₂ concentration, pressure and temperature).

2.2 MATERIALS AND METHODS

2.2.1 Experimental design

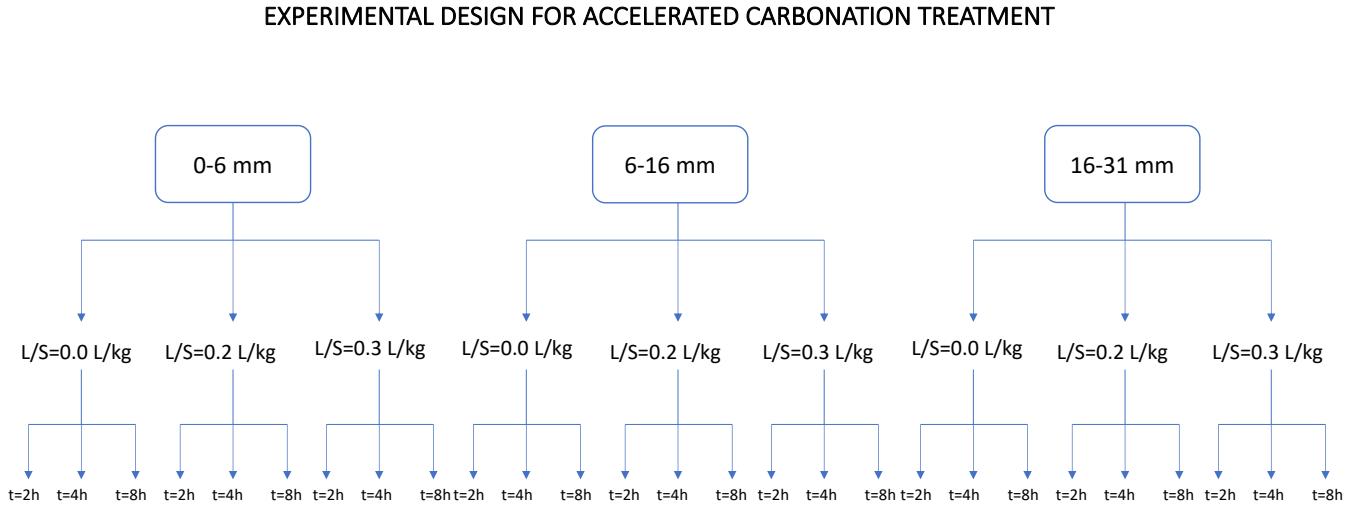


Figure 12. Schematic of the Experimental Design. Granulometry of the samples, liquid to solid ratio applied and carbonation treatments duration are shown.

The schematic of the experimental design is graphically displayed in Fig.12.

The treatments consisted of two different routes: a single step aqueous-route process carried out through two different liquid to solid ratios (L/S): 0.2 and 0.3 L/kg; and a single step dry-route process carried out with L/S=0.0 L/kg. Each process was carried out for every fraction i.e. 0/6; 6/16; 16/31 mm and for three increasing value of incubation time. (i.e., 2, 4, 8h). Each treatment was performed in triplicate. Each replicate consisted on a mass of 200g in aluminum vessels and placed in the CO₂ incubator for the selected amount of time. Since this study is the basis for a possible application it makes sense to test a significant quantity of material, and it was decided 200g. This test was also carried out with sample mass of 100; 50; 25g each.

The experimental setup has been designed according to what have already been done before in literature. While the novelty introduced in this study is that the carbonation is performed on a mix of waste materials previously treated to be inertized and intended for use in construction or disposed in landfill. Now the applied variables chosen will be discussed. Since the carbonation reaction occurs in water environment, it makes sense to add a certain amount of water to the sample before to apply the carbonation treatment. In particular, water is necessary for leaching of calcium and metal oxides and solvation of CO₂.

In this study 3 liquid to solid ratios have been chosen. First is the $\frac{L}{S} = 0.0 \text{ L/kg dry matter}$, where no water was added and we have still to consider that the samples had a certain moisture. In the second route experiment 2 different liquid to solid ratios have been used: 0.2 and 0.3 L/kg dry matter . It was fixed a 10% CO₂ concentration in volume in air to simulate the

concentration of carbon dioxide presents in the chimney fumes of an incinerator (since Padua's incinerator has 7-8% CO₂ concentration in volume) [relazione tecnica inceneritore padova 2020]. This gives value to carry out accelerate carbonation through these concentrations to simulate the contact with chimney gases of an incinerator. It was decided to keep the pressure atmospheric to be consistent with the pressure of incineration fumes. Moreover, the incubator (N-BIOTEK NB-203) can't increase the pressure inside the static chamber. Temperature was kept at room temperature $25^{\circ}\text{C} \pm 5^{\circ}\text{C}$, that corresponds to analogue operative conditions of experimentations carried out by (Baciocchi et al., 2009) (El-Hassan & Shao, 2014) (dos Reis et al., 2020, 2021). Since the carbonation reaction is exothermic ($\Delta H = -178 \text{ kJ/molCO}_2$) it makes sense to carry out the experimentation not at high temperatures. Furthermore, higher the temperature lower is the solubility of CO₂ in water, inhibiting the carbonates formation. (Baciocchi et al., 2009) (Berber et al., 2020) (Yuan et al., 2022) Tests have been carried out with incubation periods of 2, 4, 8h because it was demonstrated by (Wang et al., 2019)(Xuan & Poon, 2018) that most of the CO₂ captured occurred in the first hours of carbonation. Since the focus of this study is to assess the carbon dioxide sequestration, it was decided to perform accelerated carbonation tests for the duration of 2, 4, 8h. Different masses of the test portion subjected to accelerated carbonation test were tested (200, 100, 50, 25g) to evaluate the influence of the fluid dynamic configuration. In particular increasing the sample mass, thickness material increase, therefore diffusion of CO₂ is hindered. Tests were carried out through 8h treatment to better evaluate the difference in carbon capture between the different samples.

2.2.2 Materials

The samples of mineral/alkaline waste for this study were collected from an industrial waste treatment plant located near Verona (IT). They are the result of an inertization process of a mix of wastes which are: foundry slags, municipal solid waste incinerated bottom ashes and construction and demolition waste. Materials have undergone a process of metal separation, sieving and stored in piles. Here, the material is moved and left to mature for about 30 days on open air and irrigated. This process has the aim of increasing the stabilization to the release of contaminants by transforming oxides in hydroxides. At the end of maturation process alkaline wastes are screened in different grain size, namely 0/6, 6/16 and 16/31 mm, respectively. Laboratory samples are illustrated in **Fig.3**, **Fig.4** and **Fig.5**. Samples have been collected for the 3 fractions with about 30 kg in mass for each. All the samples were quartered and stored in closed buckets to prevent natural weathering and further natural carbonation.

Initial characterization of the aggregates is displayed in Tab.2 and included the determination of water content, pH, electric conductivity, total dissolved solids, DOC, anions, and metals according to UNI EN 12457-2 2004 leaching test. For raw data of the moisture content test, see ANNEX IX. For Carbonates content was assessed by calcimetry according to DM 13 09 1999 Met. V.1. Calcimetry tests have been performed on unaltered state and wet samples before the accelerated carbonation treatment to quantify the already carbonates content. The methodology for determining the carbonate content and the consequent carbon dioxide uptake performed is described in detail elsewhere.

Parameter	Unit	0/6 mm	6/16 mm	16/31 mm
pH	-	11.89	11.4	11.8
TS	%	85.51	97.86	95.96
Carbonates	gCO ₂ /kg _{dw}	15.79	12.00	2.69
Electric conductivity	μS/cm	3510	2410	1820
TSS	mg/l	2700	1860	1410
DOC	mg/l	155	56	2.22
Fluorides	mg/l	<0.0088	0.0222	0.157
Chlorides	mg/l	687	445	9.1
Sulphates	mg/l	55	121	18.5
As	mg/l	0.00113	0.00122	0.000411
Sb	mg/l	0.0272	0.059	0.00101
Ba	mg/l	0.262	0.092	0.087
Cd	mg/l	<0.00016	0.000107	<0.000099
Cr	mg/l	0.042	0.0209	0.0255
Hg	mg/l	0.0000680	0.000234	<0.000068
Mo	mg/l	0.085	0.119	0.069
Ni	mg/l	0.033	0.0142	0.000656
Pb	mg/l	0.044	0.0139	0.000490
Cu	mg/l	2.07	0.00134	0.00468
Se	mg/l	0.00133	0.0514	0.00115
Zn	mg/l	0.106	0.0514	0.00384

Table2. Chemical characterization of the material under analyses. These results were obtained from performing UNI 12457-2:2004 Leaching test. Carbonates content were evaluated through calcimetry analyses. TS stands for total solids, TSS for total dissolved solids, DOC for dissolved organic carbon.

The material of the fraction 16/31 mm is particularly inhomogeneous, where it is possible to clearly distinguish inerts, glass, metal slags and concrete in Fig.13. While for 6/16 and 0/6 mm fractions material appears more homogeneous. Heterogeneity of the material is an important feature, because it may determine important variations on the results as it will be discussed in the paragraph 3: results.

Since it is composed of aggregates of a mix of alkaline waste, in which aggregates have been crushed and washed, it was found a certain amount of them have already carbonated in the recycling plant. Old recycled concrete aggregates obtained from demolished concrete structures are usually partially carbonated as result of exposure to air and rain during the lifetime of the structure. This was noted also by (Fang et al., 2017) which studied carbonation effects on recycled concrete aggregates.

Moreover, has been found that the coarser the fraction, the lower is the initial carbonates content. This probably meant that after the maturation period in the industrial waste treatment plant, only portion of the waste surface naturally carbonated. Initial carbonates content amounts for 15.79 gCO₂/kg, 12.00 gCO₂/kg dw and 2.69 gCO₂/kg dw for 0/6; 6/16 and 16/31 mm fractions respectively as can be seen in Tab.2.



Figure 13. 200g Samples from the left to the right 0/6 mm, 6/16mm; 16/31 mm.



Figure 14. 200g dry samples of 6/16 and 16/31 mm fractions

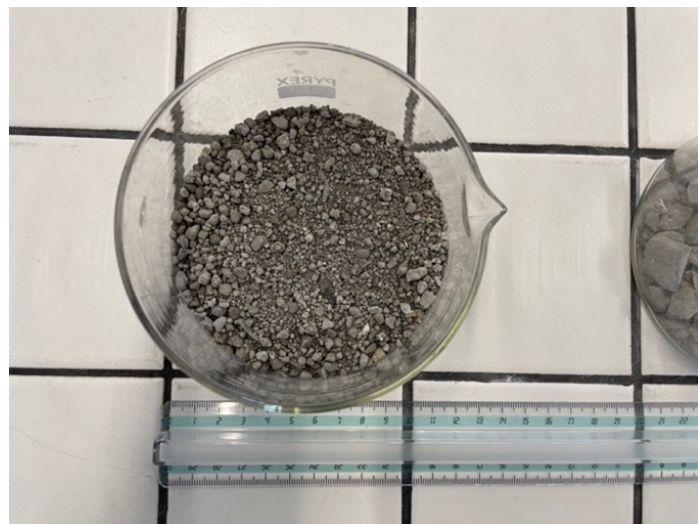


Figure 15. 200g dry sample with granulometry 0/6mm.

2.2.3 Accelerated carbonation test

It was decided to perform an accelerated carbonation in “very mild” conditions, that means in a batch reactor (static chamber) incubator N-BIOTEK NB-203 with the following operative conditions: fixed CO₂ concentration of 10% v/v , constant relative humidity of 98,5%, atmospheric pressure, and temperature $25 \pm 5^\circ\text{C}$. The static chamber was able to keep constant the operative conditions just mentioned. Temperature, relative humidity, temperature and pressure measurements are displayed in ANNEX X. Experimental setup is represented in Fig.16.

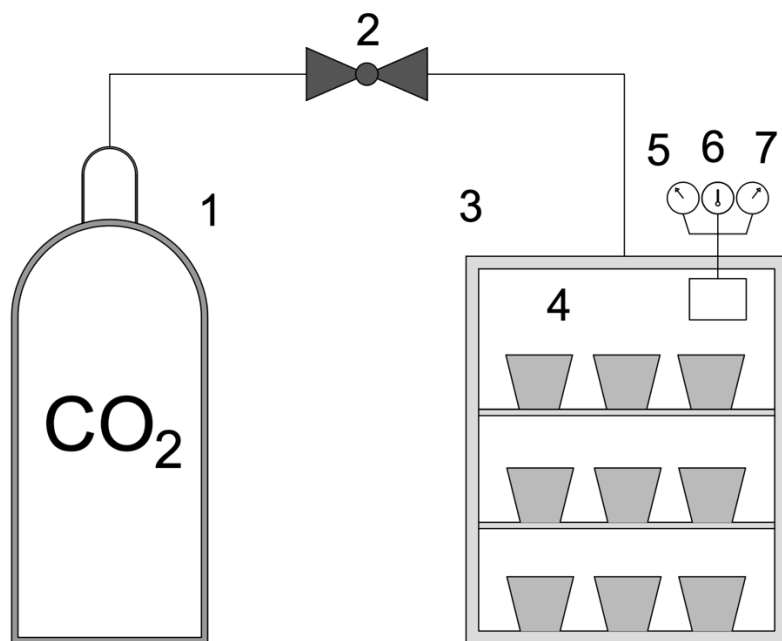


Figure 16. Experimental setup for carbonation tests: (1) CO₂ gas cylinder; (2) Valve; (3) Static chamber; (4) Samples; (5) Relative Humidity sensor; (6) Temperature sensor; (7) CO₂ partial pressure sensor.

For each granulometric fraction the experimental design allowed to test different incubation periods (2, 4, 8h) with different applied liquid to solid ratio and mass of the test portion (see Paragraph 2.3, 2.4 for detailed information).

In particular, to simulate the influence of the degree of moisture of the material, different liquid to solid ratio were applied. This was done by adding to 600g of each fraction a certain amount of demineralized water to according to the chosen L/S ratio. The slurry mix was put inside a 2 L bottle and then mixed by an agitator (MPM Instruments M100-MB) for 30 min at 8 rpm. Then, the slurry has been filtered through a filter paper (90 gsm) until it was drained. The leachate water was preserved in closed in plastic containers to be further analyzed. The filtering operation and then removal of a certain amount of water was chosen to avoid the creation of a water head in the sample that would have prevented the diffusion of CO₂. It must be noted that the water retained by samples was as bigger as the granulometry was fine. It can be seen in the Tab.3.

	Mass (g)	H ₂ O (mL)	Leachate (mL)	Leachate/H ₂ O (%)
0/6-02	627,83	125,57	20	15,93
0/6-03	610,48	183,14	50	27,30
6/16-02	638,19	127,64	70	54,84
6/16-03	553,22	165,97	130	78,33
16/31-02	617,13	123,43	90	72,92
16/31-03	560,34	168,10	155	92,21

Table 3. Leachate of the slurry filtered through the filter paper is displayed. Mass of the tal quale sample, water volume added according to the selected L/S ratio, volume of leachate passed through the filter paper are shown for each sample fraction, i.e. 0/6, 6/16 and 16/31mm

Then the so-prepared material was placed in aluminum vessels (Fig.17) in triplicate with a mass of 200g each and inserted in the incubator for the selected amount of time (2, 4, or 8h). Photos of the experimental setup and samples have been reported in ANNEX XIII.



Figure 17. 0/6 samples with a mass at least of 200g placed in aluminum vessels.

2.2.4 Chemical analyses

At the end of carbonation treatment, 3 calcimetry tests have been carried out for each replicate undergone accelerated carbonation.

The extent of carbonation was determined through calcimetry analyses as described in DM 13 09 1999 Met. V.1. For coarse fractions (6/16 mm and 16/31 mm) aggregates size was first reduced by hammer to increase the initial low homogeneity of the material.

A total of 5 ml HCl (34-37% v/v UN1789) was added to 5g of sample in the Dietrich-Fruehling apparatus, where it was possible to read the volume in mL of CO₂. For calculations carried out see ANNEX VIII. The effect of carbonation on several chemicals (As, Ba, Be, Cd, Co, Cr, Hg, Mo, Ni, Pb, Cu, Sb, Se, V, Zn, Na, K, Mg, Ca, fluorites, sulphates, nitrates, chlorides) mobility was assessed by performing the EN 12457-2:2004 leaching test on the untreated aggregates of the finest fraction (0/6mm) before and after carbonation treatment. Raw data are displayed in ANNEX XII. It was decided to assess the effects of carbonation on leaching behavior of the finest fraction because it resulted the most effective for carbon capture and so for the future development of a full-scale plant. Leachate “hardness” tests have been carried out according to APAT CNR IRSA 2040 Met.B Man 29 2003 on the liquid filtered by paper filter to assess the quantity of carbonates that have been washed when demineralized water was added and then removed from the samples. Results of this test are displayed in ANNEX XI.

2.2.5 Statistical analysis.

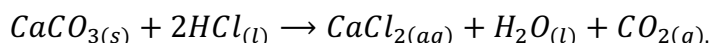
Data have been collected in Excel and starting from the amount of CO₂ read in the Dietrich-Fruhling calcimeter it was calculated the amount of $gCO_2/kg \text{ dry sorbent}$ that was sequestrated by the sample during accelerated carbonation. With Grubb test outliers were eliminated and an ANOVA Tukey test was carried out to assess if two groups of data are significantly different (confidence limit 95%, p=0.05).

It was noted that, when performing calcimetry tests, the calcimeter presented some spills of gas by submerging it in water. This caused a loss in gas pressure which meant a drop in the level of CO₂ that can be read with the instrument.

Dietrich -Fruhling calcimeter has been tared according to the following procedure.

50mL have been selected as volume of CO₂ to be reached by reaction of pure CaCO₃ in stoichiometry excess of HCl. For the experiment CaCO₃ (MW=100.09 g/mol) and HCl (34-37% UNI1789 PM) have been utilized.

The reaction is formulated with the following expression:



Through the formula $PV = nRT$, starting from the volume of CO₂ to be expected (50 ml), number of moles of $CaCO_3$ to react is calculated. It corresponds to 0,00204 mol, since the stoichiometric ratio between $CaCO_3$ and CO_2 moles is 1:1. For calculations were used P (pressure) in atm, n (number of moles) in mol, R gas constant equal to 0,08205734 l atm/K mol, V (volume) in l, and T (temperature) in K. Moles of calcium carbonates have been converted in mass (g) by multiplying it by its molecular weight ($PM = 100,087$). So, it was obtained that 0,2045g of $CaCO_3$ were necessary to react and produce 50mL of carbon dioxide.

9 tests have been carried out, and the calibration factor has been calculated as $Calibration Factor = V_{exp}/V_{th}$, where V_{exp} stands for mean experimental volume, and V_{th} for mean theoric volume. It was obtained a $Calibration Factor = 1,809042033$.

Tests results and calculations have been summarized in the following Tab.4.

CaCO ₃ sample	mass (g)	Experimental volume (mL)	mol CaCO ₃	mol CO ₂	Theoric volume(mL)
	0,25	28	0,0025	0,0025	61,1103
	0,21	33	0,0021	0,0021	51,3327
	0,25	35	0,0025	0,0025	61,1103
	0,24	32	0,0024	0,0024	58,6659
	0,18	29	0,0018	0,0018	43,9994
	0,24	27	0,0024	0,0024	58,6659
	0,19	30	0,0019	0,0019	46,4438
	0,26	33	0,0026	0,0026	63,5547
	0,23	30	0,0023	0,0023	56,2215
Mean		30,778			55,678

Calibration factor (V_{exp}/V_{th})	1,809
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Table 4. Calculations to determine the Calibration Factor

All experimental volume of CO₂ collected by calcimetry analyses have been multiplied by the calibration factor.

2.3 RESULTS AND DISCUSSION

2.3.1 Influence of operative conditions on the process

In this chapter the analysis of the experimental results is presented, and the influence of: L/S ratio, particle size, carbonation time and mass of the sample on the CO₂ uptake and in general on the carbonate content is discussed.

2.3.1.1 L/S

Fig.18 shows the influence of L/S ratio applied to 0/6, 6/16 and 16/31mm granulometry according to different timing tests.

In Fig.18a is pointed out that for 0/6 mm, L/S=0.0 L/kg caused a significant better uptake with respect the others liquid to solid ratios, achieving 8.75 gCO₂/kg dw, 10.15 gCO₂/kg dw and 13,92 gCO₂/kg dw in 2, 4, 8h respectively. Even if after 2h treatment the carbon dioxide uptake results of the samples with L/S=0.0 L/kg were comparable with the ones with L/S=0.0 L/kg, it's not possible to state the same for the results found after 4 and 8h. For there latter samples with applied L/S=0 L/kg show a significantly higher uptake with respect to samples with applied L/S=0.2 and 0.3 L/kg, the ones which behave in a similar way to each other. Fig.18b Highlights that 6/16 mm fraction, showed a better uptake with an applied L/S=0.2 L/kg with respect the others two L/S (0.0 and 0.3 L/kg respectively). This made it possible to capture 0.79, 3.24 and 2.73 gCO₂/kg dw. However, this uptake didn't increase proportionally to the carbonation treatment duration. Moreover, when L/S=0.3 L/kg was applied, the samples didn't show any uptake. Fig.18c points out that the fraction 16/31 mm increased its carbon sequestration potential with increasing L/S ratio. Indeed, after the 2h treatment it achieved a CO₂ uptake value of 16.81 gCO₂/kg dw with an applied L/S of 0.3 L/kg. However, this result is considered not consistent, since after 4 and 8h the uptake wit L/S=0.3 L/kg decreased, reaching 0 gCO₂/kg dw after 8h. The same can be said for the other L/S ratio applied.

Although it is widely demonstrated in the literature that carbonation is improved with an adequate L/ S ratio (Yuan et al., 2022) (Berber et al., 2020) the same cannot be said from our results. After the accelerated carbonation treatment, it has been found that increasing the liquid to solid ratio had a negative effect on CO₂ uptake for 0/6 fraction, while for the other fractions results are inconsistent with the treatment duration.

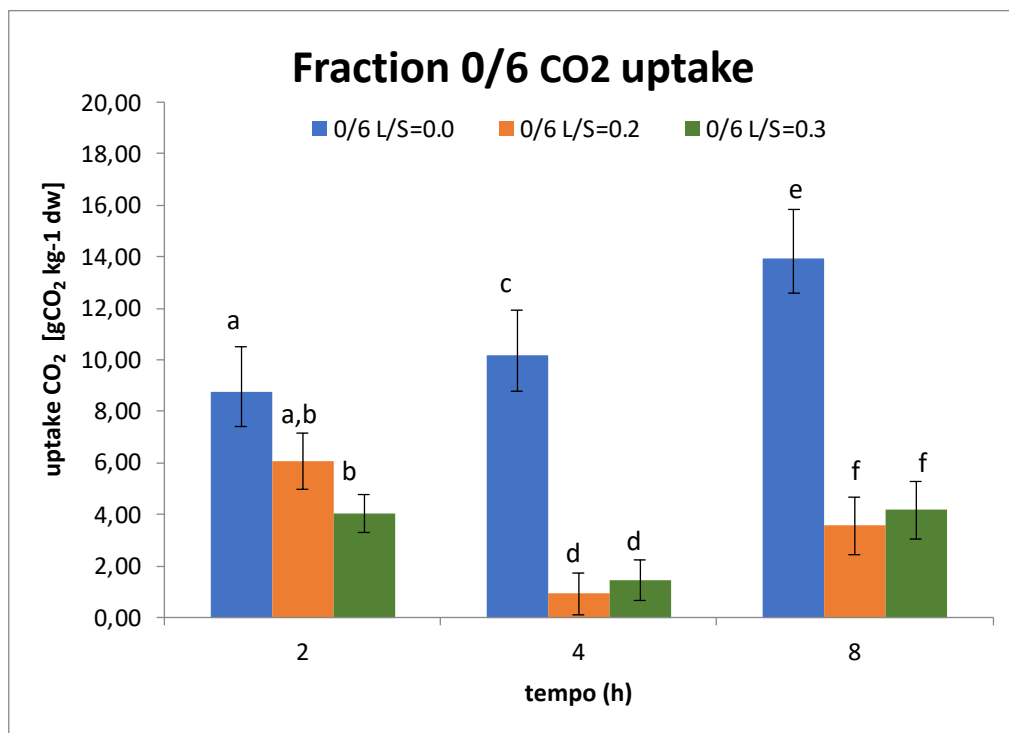


Figure 18a. CO₂ uptake of the fraction 0/6 mm with applied different L/S ratios

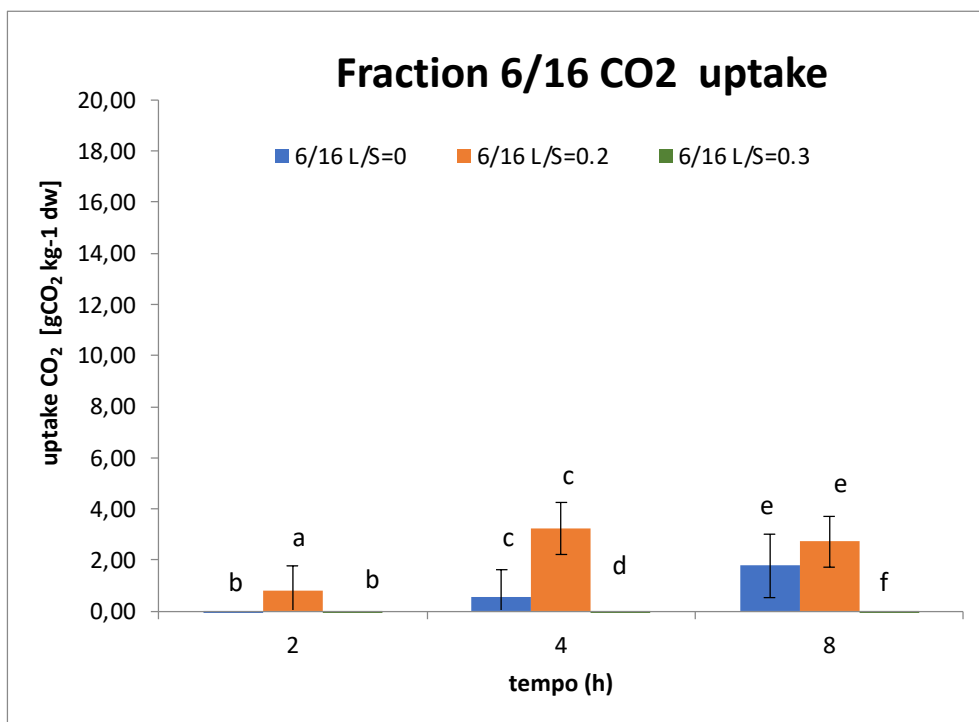


Figure 18b. CO₂ uptake of the fraction 6/16 mm with applied different l/s ratios

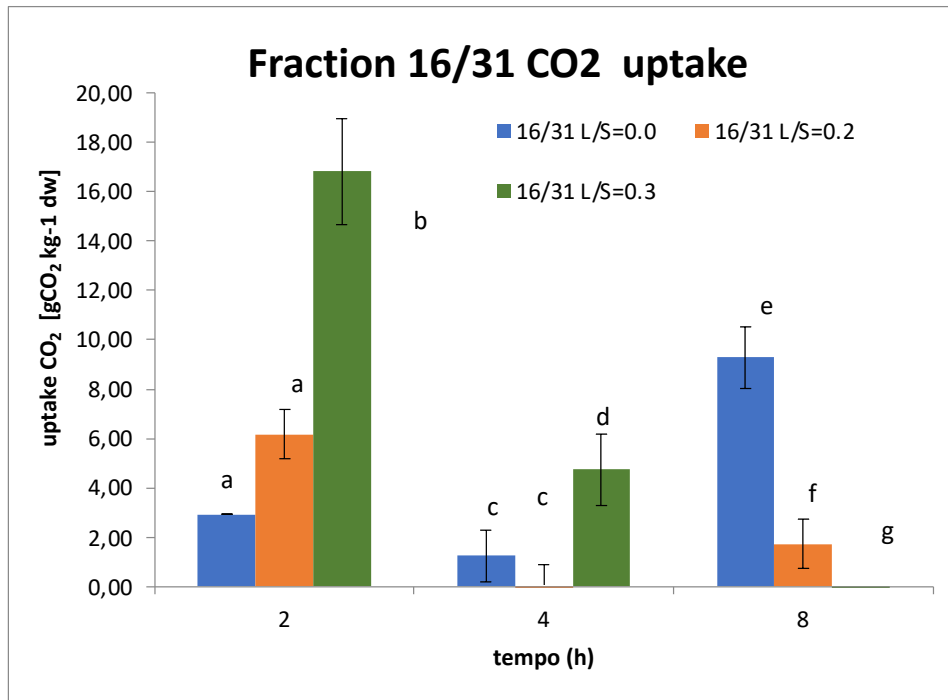


Figure 18c. CO₂ uptake of the fraction 16/31 mm with applied different l/s ratios

This observed behavior may be determined by two causes. The first is related to the decreased CO₂ diffusion potential within the material, likely due to the agglomeration and the thickening of the water layer around the solid particles. The second one could be due to the washout process probably occurred when demineralized water was firstly added and then removed from the sample before the accelerated carbonation treatment. Through washout effect chemical species involved in the carbonation process are removed from the particle surface. (Berber et al., 2020) indicates that increasing the L/S ratio on fly ashes sample material, the resulting agglomeration process inhibited CO₂ diffusion and the following carbonation reaction. This can explain the inefficiency of the accelerated carbonation treatment on 0/6 mm fraction performed in this study.

Thus, to further investigate possible influence of the washout in the decrease of carbonation potential, analyses of the leachate in terms of chemical species involved in carbonation is suggested.

Since the results on graph on Fig.18a show the highest carbon uptake for 0/6mm fraction was achieved through L/S=0 L/kg, we can infer that the inner moisture of the samples left by the maturation treatment that took place in the industrial treatment plant was enough to allow a good extent of carbonation without adding further water.

Results for the other granulometries, shown in Fig.18b and Fig.18c. seemed to be not consistent. In particular, the results of applying a L/S does not entail any benefits on carbon dioxide uptake. However, as will be further discussed, the carbonation potential of coarser fractions resulted significantly lower than the finest fraction.

2.3.1.2 Granulometry

Fig.19 shows the influence of granulometry on the carbonation uptake when accelerated carbonation is performed in 2, 4, 8h on samples with an applied L/S ratio of 0.0 L/kg. It highlights that finest fraction (i.e. 0/6 mm) had a significantly greater uptake potential than the coarser fractions (i.e. 6/16

mm and 16/31mm) with respect all the different times tested. This is verified by the Tukey test analyses carried out on these data. Indeed, while the uptake of the 0/6 fraction is never comparable to the carbon sequestration occurred in the others in all the times tested (i.e. 2, 4, 8h), the uptake of the 6/16 and 16/31 fractions is comparable after 4h accelerated carbonation treatment. Moreover, the coarsest fraction (16/31 mm) always showed a greater uptake compared to the 6/16 mm one.

It is clearly visible that granulometry 0/6 shows a positive increment to carbonation treatment that is directly proportional to time. We can't say the same for the other two fractions. 6/16 fraction uptake doesn't show any correlation to treatment duration. Indeed after 2 hours of exposure the carbonates content decreased, which means that the uptake was not significant. A slight increase in carbonates content can be visible after 4 and 8h, when a maximum uptake of 1.78 gCO₂/kg dw was measured. 16/31 fraction showed a smaller uptake in 4h than in 2h, meaning that this material doesn't respond directly to CO₂ exposure.

This trend could be due the specific surface increase and consequently more material amount per unit weight is exposed to carbonation. This result was validated by other authors findings, even performing carbonation to different materials: for instance, (Nam et al., 2012b) on bottom ashes and (Fang et al., 2017) on recycled concrete aggregates. Being carbonation a superficial phenomenon, a decrease in particle size will increase the absorption of CO₂ as reported by (Xuan & Poon, 2018).

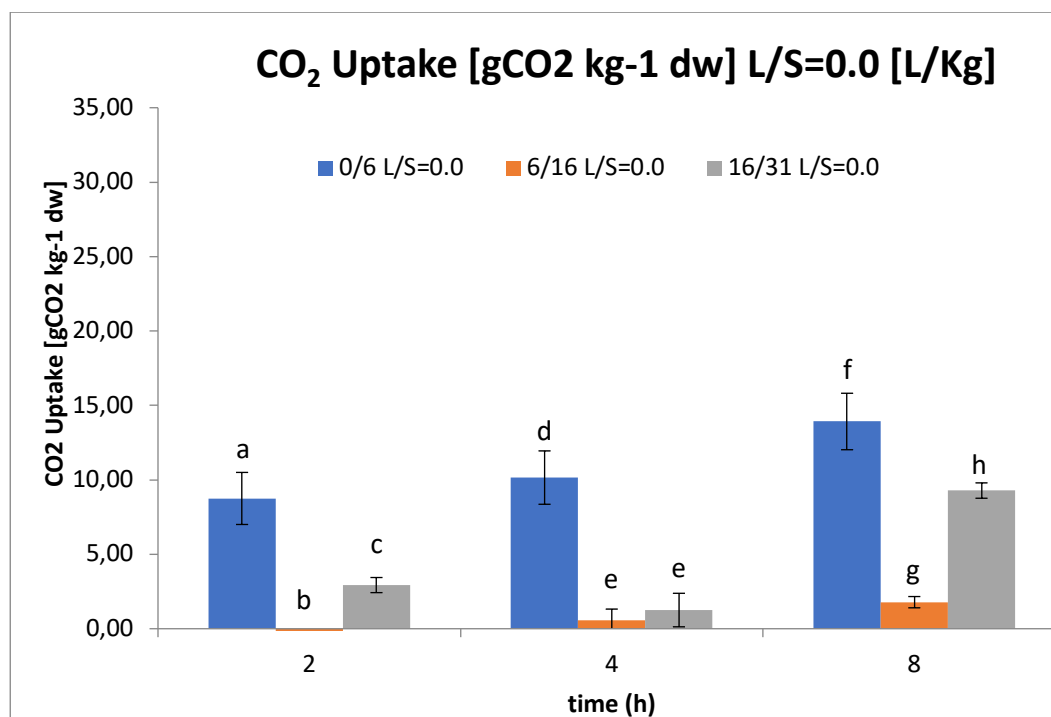


Figure 19. Influence of granulometry on the CO₂ uptake when accelerated carbonation is performed in 2, 4, 8h on samples of 0/6, 6/16 and 16/31mm granulometries with an applied L/S ratio of 0.0 L/kg.

Such differences may also be ascribed to the non-homogeneity of the collected waste samples: in particular the coarser granulometries (6/16 and 16/31 mm) could have been characterized by different (i.e., lower) concentrations of earth-metal oxides involved in the carbonation process. (Lombardi et al., 2016) also reported that non consistent results in terms of CO₂ uptake are due to the heterogeneity of the sample material. The fact that the 6/16 and 16/31 particle sizes do not show an absorption proportional to time could not suggest them as effective substrates for an upscale in real conditions.

2.3.1.3 Time

The Fig.20 displays the influence of accelerated carbonation treatment time on the different sample fractions (i.e. 0/6 mm, 6/16mm, 16/31mm) with an applied L/S ratio of 0.0 L/kg.

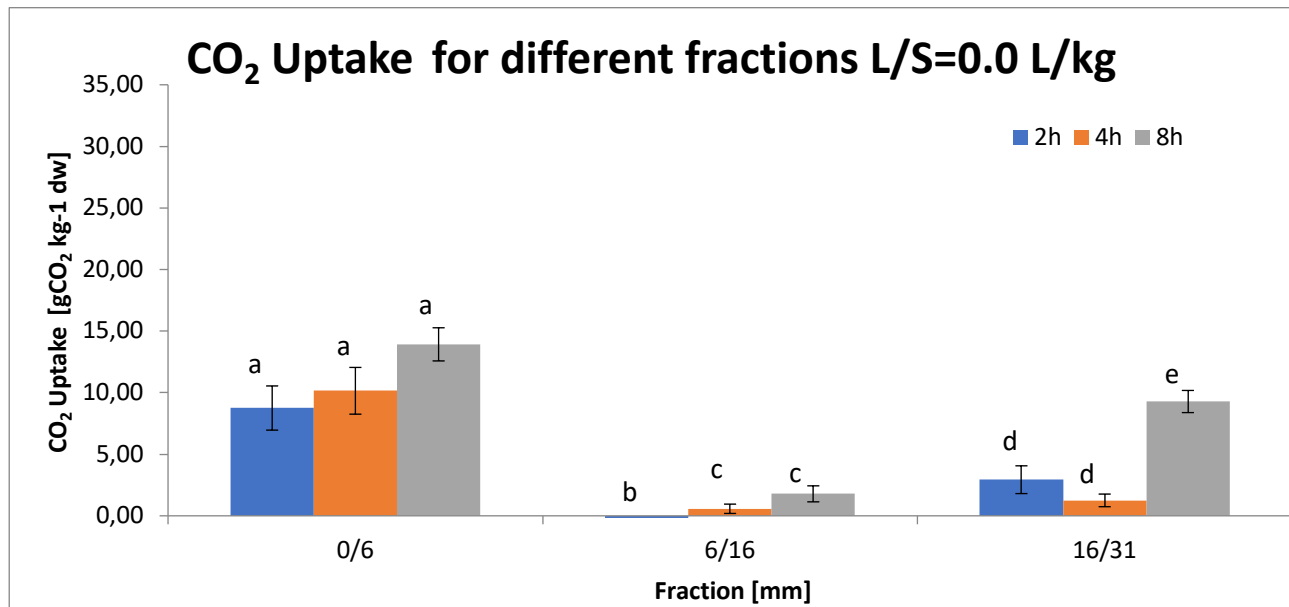


Figure 20. Carbon dioxide sequestration of the different granulometries according to 2, 4, 8h accelerated carbonation treatment with a L/S=0.0 L/kg applied.

From Fig.20 can be noted that for 0/6 fraction, there is a rapid increase in uptake after 2h treatment. A carbon sequestration of 8.75, 10.15, 13.92 gCO₂/kg dw resulted after 2, 4, 8h respectively. In our experimentation 62.86% of the total CO₂ uptake occurred in the first 2h, while the other 37.14% took place in the following 6h. Indeed, the uptake occurred in the treatment period 2-4h and 4-8h does not show significantly different data (since the letters are the same for each group of data) with respect the 0-2h period. This means that the carbonation rate is higher in the initial hours and then gradually decreased until approached a stable CO₂ uptake level. (Rostami et al., 2011) found that CO₂ uptake consequent accelerated carbonation process on concrete samples saw a rapid increase in the first 10 min, during which more than 40% of the total carbon uptake over 2 h took place. Then the reaction continued at a reduced rate and carbon uptake accumulated to about 9% after a period of 2 h, for a total accelerated carbonation treatment of 4h. Further (Xuan & Poon, 2018) reported that after 5h, where the carbonation rate was faster, it slows down to a lower rate to an approximately stable CO₂ uptake level. The same result was achieved by several authors (Yuan et al., 2022) (Schnabel et al., 2021)(Ukwattage et al., 2013) which performed accelerated carbonation tests on alkaline wastes among which recycled concrete aggregates, metal slags and incineration ashes.

For 6/16 and 16/31 mm fraction the uptake rate was lower than the 0/6mm. As can be noted in the Fig.20, for the fraction 6/16 no uptake after 2h was reported. While for 16/31 fraction can be observed that the uptake after 4h was lower than the one after 2h. The fact that after a longer accelerated carbonation treatment time there is a lower uptake was also found by (Xuan & Poon, 2018) which reports the fact that the recycled concrete aggregates had a lower CO₂ uptake after 5h compared to 3h treatment. Since for 6/16 and 16/31mm after 2h accelerated carbonation treatment the uptake was 0 and 2.93 g/kg dw, after 4h was 0.57 and 1.25 g/kg dw, and after 8h was 1.78 and 9.28 g/kg dw respectively, it can be concluded that 6/16 and 16/31 mm fraction are not suitable to be exploited in an upscale project. Nevertheless, since fraction 0/6mm demonstrated to be effective in absorbing CO₂

in the first hours of carbonation treatment, it could be suggested to utilize it in order to perform accelerated carbonation treatments in upscale conditions through short-duration tests (i.e. 2h).

2.3.1.4 Mass

In Fig.21 are displayed the results of performing 8h accelerated carbonation tests on 0/6mm fraction with an applied $L/S = 0.0$ L/kg for different sample mass, i.e. 200g, 100g, 50g, 25g.

It has been found that the sample mass is discriminating for results in term of the carbonation extent: less sample mass leads to greater results in uptake. As a matter of fact, by decreasing the mass of the sample, the thickness of the sample inside the vessels decreases. This allows the gas to spread more easily within the finest fraction. In this way the reactive surface of the material has been increased and a greater carbonation extend achieved.

Carbon uptake shows a significant increase when the mass is reduced from 200 to 100g, but not significant when reducing from 100 to 50g. After 8h carbonation, 200g of sample mass captured 13.92 gCO₂/kg dw, while decreasing the mass to 100g an uptake of 19.59 gCO₂/kg dw was observed. This means that decreasing by a half the mass, a capture increase of 40% was observed. When the mass tested was 50g the CO₂ uptake was 24.67 gCO₂/kg dw and when mass halved again the carbon capture amounted to 25.64 gCO₂/kg dw. CO₂ uptake results for 50g mass sample showed comparable results with the ones achieved by 100g and 25g but not with the 200g initial mass. In literature were not found studies which studied the mass sample influence on carbonation extent, and thus there are not comparable results.

Since the oligodynamic configuration is discriminant for the carbonation uptake, for the purposes of an upscale these results suggest using a thin layer of material to improve the carbonation reaction.

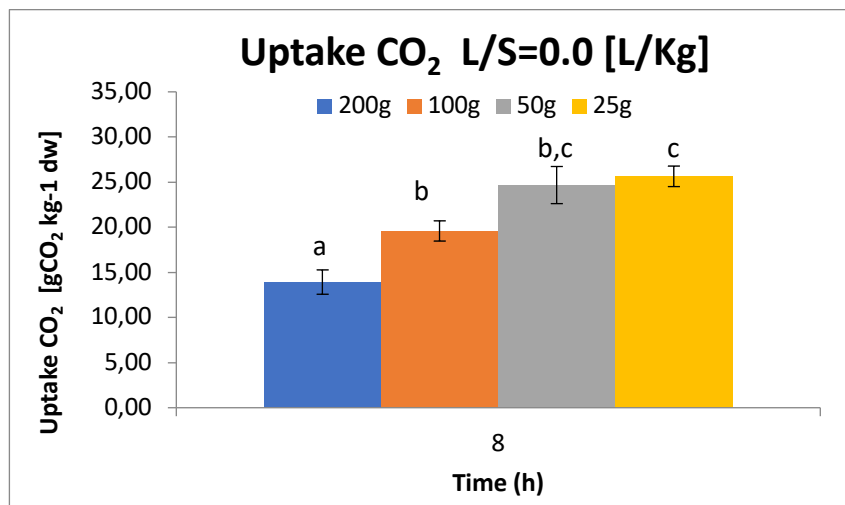


Figure 21. CO₂ uptake related of the 0/6 mm fraction after 8h accelerated carbonation treatment of samples with different mass, i.e.: 200, 100, 50, 25g.

2.3.2 Comparison with scientific literature

REFERENCE	MATERIAL	FLOW (L/min)	TIME (h)	%CO ₂	L/S (L/kg)	MASS (g)	UPTAKE gCO ₂ /kg dw	OPERATIVE CONDITIONS	CO ₂ ASSESSMENT	REACTOR
Jiang, et al. 2013	FA	0,01	1,2	12	0	100	41	Very Mild	TGA; DSC	Static
Suescum- Morales, et al. 2021	slurry mortar: RCA + cement + water	0	24-168	5	0.08	4000	3.3-27.1	Very Mild	TGA; DTA	Static
Sereng, et al. 2020	RCA	0	24	15	water content 2-13%	Not found	8.6-12.6	Very Mild	Mass gain method; TGA	Static
Fang, X et al. 2017	RCA	1-5-10	3-5-7-24	10	0	Not found	9.5-9.8-9.6-10.5	Very Mild	TGA	Static
Nam, et al. 2012	BA	0	Not found	10-30	0.1-0.3	Not found	3-165	Very Mild; Mild	Not found	Static
Berber, et al 2020	FA	100	0,5	20	0.2-0.3	200	22-108	Very Mild	Not found	Rotating

Table 5. Comparison of experimentations found in literature carried out through similar conditions to the ones presented in this study. FA= fly ash; BA= bottom ash; RCA= Recycled concrete aggregates; TGA= Thermogravimetric analysis; DSC= differential scanning calorimetry; DTA=differential thermal analysis

Our experimental results are comparable with the ones obtained from accelerated carbonation experiments in which operative conditions were very mild (temperature =ambient <35°C; pressure =atmospheric and %CO₂=0-20%) performed on recycled concrete aggregates, which are visible in Tab.5. In particular, (Sereng & Dangla, n.d.) performed carbonation tests on RCA in very mild conditions with a concentration of CO₂ equal to 15% and they found a capture range of 8.6-12.6 gCO₂ /kg dw in 24 hours. (Fang et al., 2017) performed carbonation tests on RCA in mild conditions with a concentration of CO₂ equal to 10%. Their results ranged between 9.5-10.5 gCO₂ /kg dw in 3-24 hours. If we consider concentration less than 10%, (Suescum-Morales et al., 2021) found results ranging 1.5-16,55 gCO₂ /kg dw performing carbonation on RCA with 5% CO₂ concentration in very mild conditions. CO₂ concentration play a discriminant role in the carbonation extent, as reported by (dos Reis et al., 2020) an increase of the partial pressure determines an important increase in CO₂ uptake. Regarding time and mass, (Jiang et al., 2013) and (Baciocchi et al., 2009) performed accelerated carbonation tests on fly ashes samples with comparable masses (i.e., 100g and 200g respectively) and times: 80 and 20 min respectively. They found a CO₂ capture of 41 and a range of 22-108 gCO₂ /kg dw. The last one carried out their tests through a rotating drum reactor, that can explain the higher values achieved.

It's important to note that the operative conditions together with material, time and sample mass variables play a determinant role to achieve higher values of carbon dioxide capture. That's why it was decided to compare previous studies with similar ones. For instance if we take in consideration acceleration tests performed with similar CO₂ concentration but with different operative condition and variables we found that (Baciocchi et al., 2009) obtained 250 gCO₂ /kg dw uptake in an acceleration carbonation test on fly ashes with 10% CO₂, but it was carried out in intensive conditions (pressure =1bar, temperature=300-500°C) on a sample with mass 44mg. Others considerations are that tests of this study were performed in a 98.5% relative humidity atmosphere conditions, and (Morandeau et al., 2013) reported that increasing the relative humidity of the chamber where carbonation treatment has been carried out, significantly decreased the carbonation rate. They found an optimal relative humidity value for carbonation was in the range of 50-70%, and when it was either too low or too high it would negatively affect the carbonation reactions, thus the carbon uptake. Results in terms of uptake and carbonates content are graphically represented in ANNEX XIV.

2.3.3 Mobility of pollutants after the the accelerated carbonation tests

All control (0/6-00-t0) and carbonated samples (0/6-00-t8h; 0/6-02-t8h; 0/6-03-t8h) were subjected to batch leaching tests at L/S 10 according to UNI EN 12457-2 2004 to determine the influence of the accelerated carbonation treatment on contaminant mobility. Results are displayed in Tab.6.

UNI EN 12457-2					
Chemical	Unit	0/6-00-t0	0/6-00-t8	0/6-02-t8	0/6-03-t8
pH	/	11,89 ± 0,07	9,43 ± 0,16	11,28 ± 0,04	11,53 ± 0,03
Barium	mg/l Ba	0,09 ± 0,01	0,05 ± 0,00	0,04 ± 0,00	0,06 ± 0,01
Cadmium	µg/l Cd	<1 ± 0,00	<1 ± 0,00	<1 ± 0,00	<1 ± 0,00
Chromium	µg/l Cr	54,00 ± 15,59	203,00 ± 9,64	115,33 ± 4,73	34,67 ± 6,66
Molybdenum	mg/l Mo	0,03 ± 0,03	0,11 ± 0,00	0,03 ± 0,01	<0.013 ± 0,01
Nickel	µg/l Ni	14,33 ± 0,58	8,43 ± 0,38	12,00 ± 1,00	9,40 ± 0,60
Lead	µg/l Pb	21,67 ± 7,09	<5 ± 0,00	<5 ± 0,00	11,37 ± 7,70
Copper	mg/l Cu	0,89 ± 0,03	0,77 ± 0,02	0,82 ± 0,07	0,63 ± 0,03
Antimony	mg/l Sb	0,03 ± 0,01	0,08 ± 0,01	0,06 ± 0,01	0,04 ± 0,00
Zinc	mg/l Zn	0,11 ± 0,08	0,00 ± 0,00	0,02 ± 0,00	0,04 ± 0,00
Magnesium	mg/l	0,02 ± 0,01	5,80 ± 3,55	0,15 ± 0,10	0,01 ± 0,00
Calcium	mg/l	52,67 ± 2,52	224,33 ± 5,51	81,67 ± 5,03	59,67 ± 1,53
Sulfates	mg/L SO ₄ ⁼	115,33 ±	828,67 ±	416,33 ±	160,00 ±
		21,57	39,72	61,01	11,27
Chlorides	mg/L Cl ⁻	462,33 ±	528,00 ±	375,67 ±	345,33 ±
		17,90	19,67	104,71	14,74

Table 6. Leaching test results according to UNI EN 12457-2 on 0/6 mm fraction before (t0h) and after (t8h) 8h accelerated carbonation treatment with different L/S ratios applied (00 stands for L/S= 0.0 L/kg; 02 stands for L/S= 0.2 L/kg; 03 stands for L/S= 0.3 L/kg). Results are expressed as the mean value of the triplicate leaching test ± the standard deviation.

0/6 mm fraction samples with L/S=0.0 L/kg had the most effective performance in terms of carbon dioxide uptake. The maximum CO₂ uptake of 13,9 gCO₂/kg dw after 8h accelerated carbonation treatment caused a drop in the leachate pH from 11.9 (native material) to 9.5 (carbonated material). The leachate concentrations of Ba, Ni decreases by a half, while for Pb and Zn the concentration decreases drastically from a mean of 22 to <5 µg/L and from 0.1 mg/l to 0.03mg/l. However, the leachate concentrations of total Cr, Mo, Sb, Mg, Ca, sulfates increased with CO₂ uptake. Same results for Cr, Mo, Sb, Pb and Zn have been found by (Schnabel et al., 2021) which performed accelerated carbonation tests on biomass bottom ashes. Same we can say for Pb, Zn Sb (Bacocchi et al., 2009) on fly ashes.

Moreover Mo, Cd, Pb, Zn, Cu were immobilized in full accordance with the what explained by the solubility-pH graphs, so on how pH influenced leachability of these contaminants (Meima et al., 1998).

Results of leaching tests on 0/6-02-t8h and 0/6-03-t8h samples displayed similar values among them. pH slight decrease (i. e. from 11.89 to 11.28 and 11.53) could indicate a lesser extent of occurred carbonation with respect to the L/S=0.0L/kg sample. This agrees with the lower CO₂ uptake values obtained from the calcimetries. For both has been found that Mo, Cu, concentration in leachate keeps constant, while Pb, Zn, chlorates decreases and sulfates concentration increases. All these contaminant concentrations variations are in agreement with the variations occurred due to

carbonation. Cr makes an exception; it increases for $L/S=0.2$ L/kg samples while in the $L/S=0.3$ L/kg it remains constant. We can conclude that the carbonation treatment had the expected effects on leachate.

2.4 Conclusions

- There is no standardized method to perform accelerated carbonations tests in literature. Thus, results found in terms of uptake ($\text{gCO}_2/\text{kg dw}$) for different materials showed a great range, due to the applied boundary conditions. Our study is limited to evaluate material, particle size, L/S , time and mass of the sample with fixed operative conditions (i.e. temperature, CO_2 concentration) in order to estimate the better conditions to get the maximum carbon sequestration.
- In any fractions studied, the applied water according to different L/S ratios didn't increase the CO_2 uptake of materials which already have an inner moisture content. Thus, it has been found that the best L/S ratio is 0.0 L/kg. Since this result is not in agreement with literature, it probably means that the maturation treatment occurred in the industrial waste treatment plant already carbonated the material utilized in this study.
- Fractions 6/16 and 16/31mm didn't show a visible trend of carbonation that increases with treatment time or with increasing liquid-to-solid ratio. They are therefore unsuitable for use in a subsequent upscale level plant.
- Decreasing the mass of the sample there is a significant increase in CO_2 capture.
- A maximum uptake of $25.64 \text{ gCO}_2/\text{kg dw}$ was achieved by the 0/6 mm fraction, through a sample of 25g of mass, with $L/S=0.0$ L/kg applied, after a 8h accelerated carbonation treatment in a static incubator with very mild conditions, i.e.: 25°C temperature, 98.5% relative humidity, and 10% CO_2 at atmospheric pressure.
- The results in uptake of the 0/6mm fraction agree with other studies carried out in similar conditions.

PART III - ANNEX

In this part the annex of the thesis are present. In this section is possible to find any table and graph utilized for the calculations which made possible this study.

ANNEX I: Included papers table comparison

Here the table utilized for analyze and compare the included papers is reported. It includes 34 articles as reported in the paragraph of the critical review. It was utilized to compare qualitatively the papers, to find discriminant factors and variables and to find common features present in previous studies.

ID	REFERENCES	Typology	Subent typology	Operative conditions	Intervall of operative conditions	Scale of the experiment	Characterization of the subent	Monitoring variables	Results	Objective of paper	Inclusion
1	Coppe, G., Ciro, S., Mariani, A., Piretti, M., 2012. Application of accelerated carbonation on RWK (Recycled Water Keel) for metal immobilization and CO ₂ sequestration. J. Hazard. Mater. 207-208, 159-164. https://doi.org/10.1016/j.jhazmat.2011.04.013	Batch reactor slurry	APC medium Air pollution control APC residues from municipal solid waste combustion	This applied accelerated carbonation treatment consisted of a single step in which the APC residues were subjected to a CO ₂ curing process (at atmospheric pressure)	Mild	Material not found each the precast element (100 kg of carbonation)	■ metal and anion content ■ carbonate ■ sodium content (TC) ■ sodium ion (Na) ■ mineralogical properties (X-ray diffraction analysis (XRD), thermogravimetric (TG) and differential thermal analysis (DTA))	Thermogravimetric (TG) and differential thermal analysis (DTA)	200 g CO ₂ absorbed/kg sorbent	Application of APC residues (pollution control residues from RWK) for metal immobilization and CO ₂ sequestration. The results are presented in terms of sequestration of CO ₂ .	
2	Xuan, D., Poon, C. S. (2018). Separation of carbon dioxide by RFA and enhancement of properties of RFA by accelerated carbonation. New Trends in Efficient and Sustainable Construction Materials, 103-114. https://doi.org/10.1016/B978-0-408-10248-9.00014-4	Batch reactor Open reactor	Recycled cement aggregates	2 accelerated carbonation methods are used: 1) pressurized carbonation in a chamber with 100% CO ₂ ; 2) flow through carbonation method. Flow rate of CO ₂ is 1.5 bar, 2 l/min. For the recovery of CO ₂ from the effluent, a 10% NaOH solution is used. The recovery of CO ₂ is 95.5%.	Mild	Time for carbonation: 24h; mass of sorbent not found	■ size of aggregate, apparent density, air absorption (F 15%), cement content (F 12.4%), ■ thermogravimetric (TG) and differential thermal analysis (DTA))	CO ₂ sequestration has been measured. The results are presented in terms of CO ₂ uptake. The results are presented in terms of CO ₂ uptake.	Mild conditions: different values according to the carbonation time (5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100, 105, 110, 115, 120, 125, 130, 135, 140, 145, 150, 155, 160, 165, 170, 175, 180, 185, 190, 195, 200, 205, 210, 215, 220, 225, 230, 235, 240, 245, 250, 255, 260, 265, 270, 275, 280, 285, 290, 295, 300, 305, 310, 315, 320, 325, 330, 335, 340, 345, 350, 355, 360, 365, 370, 375, 380, 385, 390, 395, 400, 405, 410, 415, 420, 425, 430, 435, 440, 445, 450, 455, 460, 465, 470, 475, 480, 485, 490, 495, 500, 505, 510, 515, 520, 525, 530, 535, 540, 545, 550, 555, 560, 565, 570, 575, 580, 585, 590, 595, 600, 605, 610, 615, 620, 625, 630, 635, 640, 645, 650, 655, 660, 665, 670, 675, 680, 685, 690, 695, 700, 705, 710, 715, 720, 725, 730, 735, 740, 745, 750, 755, 760, 765, 770, 775, 780, 785, 790, 795, 800, 805, 810, 815, 820, 825, 830, 835, 840, 845, 850, 855, 860, 865, 870, 875, 880, 885, 890, 895, 900, 905, 910, 915, 920, 925, 930, 935, 940, 945, 950, 955, 960, 965, 970, 975, 980, 985, 990, 995, 1000, 1005, 1010, 1015, 1020, 1025, 1030, 1035, 1040, 1045, 1050, 1055, 1060, 1065, 1070, 1075, 1080, 1085, 1090, 1095, 1100, 1105, 1110, 1115, 1120, 1125, 1130, 1135, 1140, 1145, 1150, 1155, 1160, 1165, 1170, 1175, 1180, 1185, 1190, 1195, 1200, 1205, 1210, 1215, 1220, 1225, 1230, 1235, 1240, 1245, 1250, 1255, 1260, 1265, 1270, 1275, 1280, 1285, 1290, 1295, 1300, 1305, 1310, 1315, 1320, 1325, 1330, 1335, 1340, 1345, 1350, 1355, 1360, 1365, 1370, 1375, 1380, 1385, 1390, 1395, 1400, 1405, 1410, 1415, 1420, 1425, 1430, 1435, 1440, 1445, 1450, 1455, 1460, 1465, 1470, 1475, 1480, 1485, 1490, 1495, 1500, 1505, 1510, 1515, 1520, 1525, 1530, 1535, 1540, 1545, 1550, 1555, 1560, 1565, 1570, 1575, 1580, 1585, 1590, 1595, 1600, 1605, 1610, 1615, 1620, 1625, 1630, 1635, 1640, 1645, 1650, 1655, 1660, 1665, 1670, 1675, 1680, 1685, 1690, 1695, 1700, 1705, 1710, 1715, 1720, 1725, 1730, 1735, 1740, 1745, 1750, 1755, 1760, 1765, 1770, 1775, 1780, 1785, 1790, 1795, 1800, 1805, 1810, 1815, 1820, 1825, 1830, 1835, 1840, 1845, 1850, 1855, 1860, 1865, 1870, 1875, 1880, 1885, 1890, 1895, 1900, 1905, 1910, 1915, 1920, 1925, 1930, 1935, 1940, 1945, 1950, 1955, 1960, 1965, 1970, 1975, 1980, 1985, 1990, 1995, 2000, 2005, 2010, 2015, 2020, 2025, 2030, 2035, 2040, 2045, 2050, 2055, 2060, 2065, 2070, 2075, 2080, 2085, 2090, 2095, 2100, 2105, 2110, 2115, 2120, 2125, 2130, 2135, 2140, 2145, 2150, 2155, 2160, 2165, 2170, 2175, 2180, 2185, 2190, 2195, 2200, 2205, 2210, 2215, 2220, 2225, 2230, 2235, 2240, 2245, 2250, 2255, 2260, 2265, 2270, 2275, 2280, 2285, 2290, 2295, 2300, 2305, 2310, 2315, 2320, 2325, 2330, 2335, 2340, 2345, 2350, 2355, 2360, 2365, 2370, 2375, 2380, 2385, 2390, 2395, 2400, 2405, 2410, 2415, 2420, 2425, 2430, 2435, 2440, 2445, 2450, 2455, 2460, 2465, 2470, 2475, 2480, 2485, 2490, 2495, 2500, 2505, 2510, 2515, 2520, 2525, 2530, 2535, 2540, 2545, 2550, 2555, 2560, 2565, 2570, 2575, 2580, 2585, 2590, 2595, 2600, 2605, 2610, 2615, 2620, 2625, 2630, 2635, 2640, 2645, 2650, 2655, 2660, 2665, 2670, 2675, 2680, 2685, 2690, 2695, 2700, 2705, 2710, 2715, 2720, 2725, 2730, 2735, 2740, 2745, 2750, 2755, 2760, 2765, 2770, 2775, 2780, 2785, 2790, 2795, 2800, 2805, 2810, 2815, 2820, 2825, 2830, 2835, 2840, 2845, 2850, 2855, 2860, 2865, 2870, 2875, 2880, 2885, 2890, 2895, 2900, 2905, 2910, 2915, 2920, 2925, 2930, 2935, 2940, 2945, 2950, 2955, 2960, 2965, 2970, 2975, 2980, 2985, 2990, 2995, 3000, 3005, 3010, 3015, 3020, 3025, 3030, 3035, 3040, 3045, 3050, 3055, 3060, 3065, 3070, 3075, 3080, 3085, 3090, 3095, 3100, 3105, 3110, 3115, 3120, 3125, 3130, 3135, 3140, 3145, 3150, 3155, 3160, 3165, 3170, 3175, 3180, 3185, 3190, 3195, 3200, 3205, 3210, 3215, 3220, 3225, 3230, 3235, 3240, 3245, 3250, 3255, 3260, 3265, 3270, 3275, 3280, 3285, 3290, 3295, 3300, 3305, 3310, 3315, 3320, 3325, 3330, 3335, 3340, 3345, 3350, 3355, 3360, 3365, 3370, 3375, 3380, 3385, 3390, 3395, 3400, 3405, 3410, 3415, 3420, 3425, 3430, 3435, 3440, 3445, 3450, 3455, 3460, 3465, 3470, 3475, 3480, 3485, 3490, 3495, 3500, 3505, 3510, 3515, 3520, 3525, 3530, 3535, 3540, 3545, 3550, 3555, 3560, 3565, 3570, 3575, 3580, 3585, 3590, 3595, 3600, 3605, 3610, 3615, 3620, 3625, 3630, 3635, 3640, 3645, 3650, 3655, 3660, 3665, 3670, 3675, 3680, 3685, 3690, 3695, 3700, 3705, 3710, 3715, 3720, 3725, 3730, 3735, 3740, 3745, 3750, 3755, 3760, 3765, 3770, 3775, 3780, 3785, 3790, 3795, 3800, 3805, 3810, 3815, 3820, 3825, 3830, 3835, 3840, 3845, 3850, 3855, 3860, 3865, 3870, 3875, 3880, 3885, 3890, 3895, 3900, 3905, 3910, 3915, 3920, 3925, 3930, 3935, 3940, 3945, 3950, 3955, 3960, 3965, 3970, 3975, 3980, 3985, 3990, 3995, 4000, 4005, 4010, 4015, 4020, 4025, 4030, 4035, 4040, 4045, 4050, 4055, 4060, 4065, 4070, 4075, 4080, 4085, 4090, 4095, 4100, 4105, 4110, 4115, 4120, 4125, 4130, 4135, 4140, 4145, 4150, 4155, 4160, 4165, 4170, 4175, 4180, 4185, 4190, 4195, 4200, 4205, 4210, 4215, 4220, 4225, 4230, 4235, 4240, 4245, 4250, 4255, 4260, 4265, 4270, 4275, 4280, 4285, 4290, 4295, 4300, 4305, 4310, 4315, 4320, 4325, 4330, 4335, 4340, 4345, 4350, 4355, 4360, 4365, 4370, 4375, 4380, 4385, 4390, 4395, 4400, 4405, 4410, 4415, 4420, 4425, 4430, 4435, 4440, 4445, 4450, 4455, 4460, 4465, 4470, 4475, 4480, 4485, 4490, 4495, 4500, 4505, 4510, 4515, 4520, 4525, 4530, 4535, 4540, 4545, 4550, 4555, 4560, 4565, 4570, 4575, 4580, 4585, 4590, 4595, 4600, 4605, 4610, 4615, 4620, 4625, 4630, 4635, 4640, 4645, 4650, 4655, 4660, 4665, 4670, 4675, 4680, 4685, 4690, 4695, 4700, 4705, 4710, 4715, 4720, 4725, 4730, 4735, 4740, 4745, 4750, 4755, 4760, 4765, 4770, 4775, 4780, 4785, 4790, 4795, 4800, 4805, 4810, 4815, 4820, 4825, 4830, 4835, 4840, 4845, 4850, 4855, 4860, 4865, 4870, 4875, 4880, 4885, 4890, 4895, 4900, 4905, 4910, 4915, 4920, 4925, 4930, 4935, 4940, 4945, 4950, 4955, 4960, 4965, 4970, 4975, 4980, 4985, 4990, 4995, 5000, 5005, 5010, 5015, 5020, 5025, 5030, 5035, 5040, 5045, 5050, 5055, 5060, 5065, 5070, 5075, 5080, 5085, 5090, 5095, 5100, 5105, 5110, 5115, 5120, 5125, 5130, 5135, 5140, 5145, 5150, 5155, 5160, 5165, 5170, 5175, 5180, 5185, 5190, 5195, 5200, 5205, 5210, 5215, 5220, 5225, 5230, 5235, 5240, 5245, 5250, 5255, 5260, 5265, 5270, 5275, 5280, 5285, 5290, 5295, 5300, 5305, 5310, 5315, 5320, 5325, 5330, 5335, 5340, 5345, 5350, 5355, 5360, 5365, 5370, 5375, 5380, 5385, 5390, 5395, 5400, 5405, 5410, 5415, 5420, 5425, 5430, 5435, 5440, 5445, 5450, 5455, 5460, 5465, 5470, 5475, 5480, 5485, 5490, 5495, 5500, 5505, 5510, 5515, 5520, 5525, 5530, 5535, 5540, 5545, 5550, 5555, 5560, 5565, 5570, 5575, 5580, 5585, 5590, 5595, 5600, 5605, 5610, 5615, 5620, 5625, 5630, 5635, 5640, 5645, 5650, 5655, 5660, 5665, 5670, 5675, 5680, 5685, 5690, 5695, 5700, 5705, 5710, 5715, 5720, 5725, 5730, 5735, 5740, 5745, 5750, 5755, 5760, 5765, 5770, 5775, 5780, 5785, 5790, 5795, 5800, 5805, 5810, 5815, 5820, 5825, 5830, 5835, 5840, 5845, 5850, 5855, 5860, 5865, 5870, 5875, 5880, 5885, 5890, 5895, 5900, 5905, 5910, 5915, 5920, 5925, 5930, 5935, 5940, 5945, 5950, 5955, 5960, 5965, 5970, 5975, 5980, 5985, 5990, 5995, 6000, 6005, 6010, 6015, 6020, 6025, 6030, 6035, 6040, 6045, 6050, 6055, 6060, 6065, 6070, 6075, 6080, 6085, 6090, 6095, 6100, 6105, 6110, 6115, 6120, 6125, 6130, 6135, 6140, 6145, 6150, 6155, 6160, 6165, 6170, 6175, 6180, 6185, 6190, 6195, 6200, 6205, 6210, 6215, 6220, 6225, 6230, 6235, 6240, 6245, 6250, 6255, 6260, 6265, 6270, 6275, 6280, 6285, 6290, 6295, 6300, 6305, 6310, 6315, 6320, 6325, 6330, 6335, 6340, 6345, 6350, 6355, 6360, 6365, 6370, 6375, 6380, 6385, 6390, 6395, 6400, 6405, 6410, 6415, 6420, 6425, 6430, 6435, 6440, 6445, 6450, 6455, 6460, 6465, 6470, 6475, 6480, 6485, 6490, 6495, 6500, 6505, 6510, 6515, 6520, 6525, 6530, 6535, 6540, 6545, 6550, 6555, 6560, 6565, 6570, 6575, 6580, 6585, 6590, 6595, 6600, 6605, 6610, 6615, 6620, 6625, 6630, 6635, 6640, 6645, 6650, 6655, 6660, 6665, 6670, 6675, 6680, 6685, 6690, 6695, 6700, 6705, 6710, 6715, 6720, 6725, 6730, 6735, 6740, 6745, 6750, 6755, 6760, 6765, 6770, 6775, 6780, 6785, 6790, 6795, 6800, 6805, 6810, 6815, 6820, 6825, 6830, 6835, 6840, 6845, 6850, 6855, 6860, 6865, 6870, 6875, 6880, 6885, 6890, 6895, 6900, 6905, 6910, 6915, 6920, 6925, 6930, 6935, 6940, 6945, 6950, 6955, 6960, 6965, 6970, 6975, 6980, 6985, 6990, 6995, 7000, 7005, 7010, 7015, 7020, 7025, 7030, 7035, 7040, 7045, 7050, 7055, 7060, 7065, 7070, 7075, 7080, 7085, 7090, 7095, 7100, 7105, 7110, 7115, 7120, 7125, 7130, 7135, 7140, 7145, 7150, 7155, 7160, 7165, 7170, 7175, 7180, 7185, 7190, 7195, 7200, 7205, 7210, 7215, 7220, 7225, 7230, 7235, 7240, 7245, 7250, 7255, 7260, 7265, 7270, 7275, 7280, 7285, 7290, 7295, 7300, 7305, 7310, 7315, 7320, 7325, 7330, 7335, 7340, 7345, 7350, 7355, 7360, 7365, 7370, 7375, 7380, 7385, 7390, 7395, 7400, 7405, 7410, 7415, 7420, 7425, 7430, 7435, 7440, 7445, 7450, 7455, 7460, 7465, 7470, 7475, 7480, 7485, 7490, 7495, 7500, 7505, 7510, 7515, 7520, 7525, 7530, 7535, 7540, 7545, 7550, 7555, 7560, 7565, 7570, 7575, 7580, 7585, 7590, 7595, 7600, 7605, 7610, 7615, 7620, 7625, 7630, 7635, 7640, 7645, 7650, 7655, 7660, 7665, 7670, 7675, 7680, 7685, 7690, 7695, 7700, 7705, 7710, 7715, 7720, 7725, 7730, 7735, 7740, 7745, 7750, 7755, 7760, 7765, 7770, 7775, 7780, 7785, 7790, 7795, 7800, 7805, 7810, 7815, 7820, 7825, 7830, 7835, 7840, 7845, 7850, 7855, 7860, 7865, 7870, 7875, 7880, 7885, 7890, 7895, 7900, 7905, 7910, 7915, 7920, 7925, 7930, 7935, 7940, 7945, 7950, 7955, 7960, 7965, 7970, 7975, 7980, 7985, 7990, 7995, 8000, 8005, 8010, 8015, 8020, 8025, 8030, 8035, 8040, 8045, 8050, 8055, 8060, 8065, 8070, 8075, 8080, 8085, 8090, 8095, 8100, 8105, 8110, 8115, 8120, 8125, 8130, 8135, 8140, 8145, 8150, 8155, 8160, 8165, 8170, 8175, 8180, 8185, 8190, 8195, 8200, 8205, 8210, 8215, 8220, 8225, 8230, 8235, 8240, 8245, 8250, 8255, 8260, 8265, 8270, 8275, 8280, 8285, 8290, 8295, 8300, 8305, 8310, 8315, 8320, 8325, 8330, 8335, 8340, 8345, 8350, 8355, 8360, 8365, 8370, 8375, 8380, 8385, 8390, 8395, 8400, 8405, 8410, 8415, 8420, 8425, 8430, 8435, 8440, 8445, 8450, 8455, 8460, 8465, 8470, 8475, 8480, 8485, 8490, 8495, 8500, 8505, 8510, 8515, 8520, 8525, 8530, 8535, 8540, 8545, 8550, 8555, 8560, 8565, 8570, 8575, 8580, 8585, 8590, 8595, 8600, 8605, 8610, 8615, 8620, 8625, 8630, 8635, 8640, 8645, 8650, 8655, 8660, 8665, 8670, 8675, 8680, 8685, 8690, 8695, 8700, 8705, 8710, 8715, 8720, 8725, 8730, 8735, 8740, 8745, 8750, 8755, 8760, 8765, 8770, 8775, 8780, 8785, 8790, 8795, 8800, 8805, 8810, 8815, 8820, 8825, 8830, 8835, 8840, 8845, 8850, 8855, 8860, 8865, 8870, 8875, 8880, 8885, 8890, 8895, 8900, 8905, 8910, 8915, 8920, 8925, 8930, 8935, 8940, 8945, 8950, 8955, 8960, 8965, 8970, 8975, 8980, 8985, 8990, 8995, 9000, 9005, 9010, 9015, 9020, 9025, 9030, 9035, 9040, 9045, 9050, 9055, 9060, 9065, 9070, 9075, 9080, 9085, 9090, 9095, 9100, 9105, 9110, 9115, 9120, 9125, 9130, 9135, 9140, 9145, 9150, 9155, 9160, 9165, 9170, 9175, 9180, 9185, 9190, 9195, 9200, 9205, 9210, 9215, 9220, 9225, 9230, 9235, 9240, 9245, 9250, 9255, 9260, 9265, 9270, 9275, 9280, 9285, 9290, 9295, 9300, 9305, 9310, 9315, 9320, 9325, 9330, 9335, 9340, 9345, 9350, 9355, 9360, 9365, 9370, 9375, 9380, 9385, 9390, 9395, 9400, 9405, 9410, 9415, 9420, 9425, 9430, 9435, 9440, 9445, 9450, 945		

13	Suescum-Morales, D., Fernández-Rodríguez, J. M., & Jiménez, J. R. (2022). Use of carbonated water to improve the mechanical properties and reduce the carbon footprint of cement-based materials with recycled aggregates. <i>Journal of CO2 Utilization</i> , 45, 103275. https://doi.org/10.1016/j.jcou.2022.103275	batch reactor; slurry reactor	Natural aggregates, RCA (recycled concrete aggregates)	2 different experiments were carried out. In the curing topology a normal climatic chamber was used. CO2 concentration = atmospheric concentration (0.04%), T=21.2 °C; RH =65 ±10%. In the water curing, topology dry samples were put inside containers where natural water or carbonated water was added. Containers were sealed and kept at T=21.2 °C.	Mild	time of curing: 1,3,7 days; mass of sorbent= 2500g and 3500g	Wave length dispersive X-ray fluorescence spectrometry (XRF-PRI-MUS IV, Rigaku, 4 kW power) was performed to determine the chemical composition of the raw materials (FA, RMA, and cement), to determine the morphology and composition of the "stake-outs", scanning electron microscopy (SEM) and mapping were performed with a JEOL JSM 7800 F.	The CO2 uptake of the hardened mixes can be determined using TGA/DTA	max CO2 uptake = 13.8 and 16.5 gCO2/kg dry sorbent. Results and details on paper sheet.	This study evaluated the effect of the use of carbonated water used as curing medium on the properties at an early curing age, as well as the CO2 sequestration capacity.	
14	Rostami, V., Shah, T., & Rostami, V. (2021). Durability of carbonation curing, <i>in</i>-Construction and Building Materials</i>(7), <i>42</i>55-4268. https://doi.org/10.1016/j.cembsu.2021.100106	batch reactor	concrete pipes	p=1.5 bar, carbonation time=2h; T=24 °C; CO2 purity=99.5%; RH=20%. The mixture of samples were proportioned using ordinary Portland cement (CSA Type-GU) at 286 kg/m3, coarse aggregate 730 kg/m3, fine aggregate 1050 kg/m3 and water 100 kg/m3. In the carbonated samples	Intensive	mass of sorbent not found; time of carbonation=2h	The mixture of samples were proportioned using ordinary Portland cement (CSA Type-GU) at 286 kg/m3, coarse aggregate 730 kg/m3, fine aggregate 1050 kg/m3 and water 100 kg/m3	Mass gain method	87.89; 79 gCO2/kg sorbent	improve durability performance and explore the possibility of using concrete pipe to sequester carbon dioxide	not from CR, cited by another paper
15	Zhang, D., & 888, Shao, Y. (2016). Early age carbonation curing of Portland cement concrete. <i>Journal of Building Materials</i> , 19(13), 134-143. https://doi.org/10.1016/j.jbm.2016.03.048	batch reactor, slurry reactor	OPC (Ordinary Portland Cement)	p=1.5 bar; CO2 purity=99.8%; time of carbonation=12h; RH=65 ±5%; T=25±3 °C	Intensive	mass of sorbent not found; time of carbonation=12h	Mixture proportions of concrete are given.	Mass gain method	With mass gain method: 10.13, 1584 gCO2/kg dry cement; mass curve method: 113.3 gCO2/kg dry cement; increasing pressure to 5 bar: 99.5 to 146.9 gCO2/kg dry cement	improve the performance of OPC	not from CR, cited by another paper
16	Das Reis, Gaydon S., et al. "Coupling of Attrition and Accelerated Carbonation for CO2 Sequestration in Recycled Concrete Aggregates." <i>Cleaner Engineering and Technology</i> , vol. 3, Elsevier Ltd., 2021. https://doi.org/10.1016/j.cet.2021.100106	batch reactor + rotating drum	RCA (Recycled Concrete Aggregates)	p=0.75 bar, CO2 concentration =100%; T=2 °C; samples were initially dried, then immersed in water and then re-dried at T=70 °C to reach adequate moisture grade, moisture, size of particles and rotation speed of the drum were varied	Mild	curing times: up to 7 days; mass of sorbent= 500g	particle size and shape obtained from sieving and particle shape obtained according to the standard French XP P 18-566-2002 (AFNOR 2002). Chemical composition analysis was obtained through a scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (EDS) (SEM31, Sweden). Particle density and water absorption potential were assessed according NF EN 1097-46.	not found	according moisture variation and drum rotation velocity. CO2 captured=16.0 to 39.5 g/kg dry sorbent	improve the performance of recycled concrete aggregates	
17	Schnabel, Kevin, et al. "Technically Exploitable Mineral Carbonation Potential of Four Alkaline Waste Materials and Effect on Compressive Modulus." <i>Greenhouse Gas Utilization</i> , vol. 39, 2020. https://doi.org/10.1016/j.jcou.2020.101181	batch reactor	Fly ashes (biomass bottom and fly ashes from fuel incineration) and steel slags	dry samples are mixed with deionized water (L/S =0.1; 0.2; 0.3; 0.4; 0.5). They have been allowed to hydrate for 30 min or 24 h according fast or slow carbonation. Pressure=atmospheric pressure	Mild	mass of sorbent = 200g; curing time= 2, 168h	The acid neutralizing capacity of the samples (ANC in mol kg-1 dry matter (DM)) was determined by titrating aqueous suspensions using a Mettler Toledo 770 titrator equipped with a titration cell (C103) and a titration cell (C103) was used. The elemental composition of the materials was determined by energy-dispersive X-ray fluorescence analysis (EDS) in single determinations. Thermogravimetric analysis (TGA) were conducted to quantify solid phase composition and to determine the degree of carbonation.	TGA	varying according L/S ratio 43.56; 92.84; 76.56; 111.76 gCO2/kg Portland cement	to examine the CO2 sequestration potential of dry-fly-ash aggregates and the effect of curing time and experimental approaches.	
18	Suescum-Morales, D., et al. "Accelerated Carbonation of Fresh Cement-Based Products Containing Recycled Masonry Aggregates for CO2 Sequestration." <i>Journal of CO2 Utilization</i> , vol. 46, 2021. https://doi.org/10.1016/j.jcou.2021.101461	batch reactor	RCA, Natural aggregates	First experiment carried out in a climatic chamber: CO2 conc=0.04%, T=22 °C, RH=65 ±10%, T=21.2 °C (Pair-Exhaust). The aggregates were previously saturated (because water is essential for the carbonation reaction). After this curing time of 7 days, TGA/DTA and XRD analyses were carried out. Time of carbonation 1-3-7 days was varied for each sample, particle pressure of CO2 was varied.	Mild	time for carbonation: 1-3-7 days; mass of sorbent not found	According to UNE EN 933-1:2009, we get composition of RCA (recycled concrete aggregates) and natural aggregates (NA) as follows: mass of sorbent = 200g; curing time= 2, 168h	TGA	varying in a range 1,573-26.35 gCO2/kg sorbent	This study examines the effects of accelerated carbonation on the mechanical properties, dry bulk density, water absorption, and CO2 sequestration capacity of Portland cement-based material that contains different replacement ratio of natural aggregates (NA) by recycled masonry aggregates (RMA).	
19	dos Reis, Gaydon S., et al. "Effect of the Accelerated Carbonation Treatment on the Recycled Sand Physicochemical Characteristics through the Self Physicochemical Processes." <i>Journal of CO2 Utilization</i> , vol. 39, 2020. https://doi.org/10.1016/j.jcou.2020.101181	batch reactor	RCA (recycled concrete aggregates)	T=22 °C; P=atmospheric pressure, CO2 partial pressure = 0.8-1 bar. Samples are dried and then pre-treated with water to reach the wanted level of moisture. The rotation speed of the drum was increased from 0 to 100 rpm. Initial moisture was varied (4-12%)	Mild	mass of sorbent sample=500g; carbonation time=7h	water absorption capacity, density was given. The morphology of the samples was determined using SEM (SEM TESCAN3, Sweden). Thermogravimetric (TGA) and Differential Thermal Analysis (DTA) analyses were made by using a TA Instruments model SDT Q600 (New Castle, USA)	TGA	according moisture, drum velocity CO2 uptake varies 4.2-47.4 gCO2/kg sorbent	to study accelerated carbonation varying moisture, rotation speed	
20	Wang, Dongfang, et al. "Quantification and Micro-Mechanisms of CO2 Sequestration in Magnesia-Lime Fly-Ash/Solidified Soil." <i>International Journal of Greenhouse Gas Control</i> , vol. 91, Elsevier Science, 2019. https://doi.org/10.1016/j.jggc.2019.103837	open reactor; slurry reactor	APC residues from MSW incineration	The initial materials (200g) were mixed with water (L/S = 0.2-0.3 w/w) for 2 min. Then, a gas flow of 20% CO2 in air was fed into the equipment at a rate of 100L/h for 30 min. Then, additives, such as OPC and/or quarry, fines, were added. Then, the equipment was sealed and the gas flow was continued. After this curing time of 7 days, TGA/DTA and XRD analyses were carried out. Time of carbonation 1-3-7 days was varied for each sample, particle pressure of CO2 was varied.	Mild	carbonation time= 20-30 min; mass of dry sorbent =200g	The Brunauer, Emmett, and Teller (BET)-specific surface area (SSA) and particle size distribution (PSD) were determined using a Malvern 1042 sorptometer and Horiba LA-500 laser-scattering PSD analyzer, respectively. Chemical composition was determined using an inductively coupled plasma atomic emission spectrometer (ICP-AES) (PerkinElmer AAnalyst 800) (Beijing, China). Thermogravimetric (TGA) and Differential Thermal Analysis (DTA) analyses were made by using a TA Instruments model SDT Q600 (New Castle, USA)	analytical determination via mathematical formula considering concentration of chemical species	several results: ranging 23-108 gCO2/kg sorbent	Focusing mainly on the effects produced by the content of municipal solid waste incineration ash in the additives, the granule compositions were varied in order to study the effect of the different CO2 uptake, strength development, leaching behaviour, microstructure, and porosity.	
21	Kim, Jung, and Woo Kwon. "Semi-Dry Carbonation Process Using Fly Ash from Solid Released Fuel Power Plant." <i>Sustainability</i> , vol. 11, no. 3, Molecular Diversity Preservation International, 2019. https://doi.org/10.3390/su11030506	round bottom flask rotating reactor	Fly ashes, slag, lime, magnesia, kaolin powder	fly ash + water (50g) were put inside the round bottom flask reactor, rotation velocity fixed to 40 rpm. Pure CO2 at a flow rate of 10 L/min. T=25 °C. Water added was varied: L/S ratio= 0.25-0.50-0.75-1, reaction time= 60 min.	Intensive	carbonation time: from 0.5 to 24h	The morphologies and structures of the fly ash were analyzed by SEM (JSM-4700F, JEOL, Tokyo, Japan). The crystal structures were investigated by XRD (X'Pert3 Powder, Malvern PANalytical) with JADE 9.0 software. The elemental compositions of the fly ash were determined by ICP-AES (PerkinElmer AAnalyst 800) (Beijing, China). Thermogravimetric (TGA) and Differential Thermal Analysis (DTA) analyses were made by using a TA Instruments model SDT Q600 (New Castle, USA)	XRF pre and post carbonation	101.7 gCO2/kg sorbent	the utilization of the fly ash from a solid refused fuel (SRF) power plant as a solid sorbent material for CO2 capture via semi-dry carbonation reaction was evaluated as a simple process to reduce CO2	
22	Pai, Shu-Li, et al. "Efficacy of Carbonated Petroleum Residue as a CO2 Sorbent in Cement Mortars." <i>Journal of Cleaner Production</i> , vol. 180, Elsevier Science Ltd, 2018. pp. 689-97. https://doi.org/10.1016/j.jclepro.2018.01.055	rotating packing bed reactor; slurry reactor	Fly ashes from petroleum coke power plant	Cole fly ash was mixed with blowdown wastewater in a slurry tank reaching L/S ratio of 10m-3/kg. Subsequently, the fly ash slurry was pumped into the RPB stream which was dragged by the blower. Experimentation was over when it could be confirmed that the carbonation completed as soon as pH dropped below 6.3 which is defined by the pKa between bicarbonate and dissolved carbon dioxide. P=atmospheric (7).	Mild	mass of sorbent: time for carbonation = not found	The compositions of PCFA and clinker in this study were examined by X-ray Fluorescence (XRF) and chemical titration analysis	TGA	84.5 gCO2/kg sorbent	In this study, an innovative valorization of petroleum coke fly ash as supplementary cementitious materials in blended cement mortars was investigated	
23	Ukwattage, N.L., et al. "Steel-Making Slag for Mineral Carbonation." <i>Measurement</i> , vol. 97, Published for IMEKO by the Institute of Measurement and Control, 2017. pp. 15-22. https://doi.org/10.1016/j.measurement.2016.10.057	slurry, batch reactor	steel slags	for slurry tests: L/S= from 0.25 to 3, T=20-50-80 °C; CO2 pressure=1 to 6 MPa. Samples were varied	Intensive	mass of sorbent=100 g; time=48h	X-ray fluorescence spectroscopy (XRF) analysis of the mineral composition	ideal gas law	up to 29.47 gCO2/kg sorbent	In the present study, the test apparatus was used to investigate the effect of three reaction parameters (water-to-solid ratio, temperature and pressure) on the mineral carbonation of steel-making slag	
24	Lombardi, L., et al. "Experimental Evaluation of Two Different Types of Reactors for CO2 Removal from Gaseous Stream by Bottom Ash Accelerated Carbonation." <i>Waste Management</i> , vol. 58, Pergamon Press, 2016. pp. 1-10. https://doi.org/10.1016/j.wasman.2016.09.038	rotating reactor; open reactor	Bottom ashes (MSW incineration)	fixed bed reactor: CO2 gas flow/solid ratio equal to 4 Nl/(h.kgBq). Rotating reactor: filling ratio (10.20, 30% of volume reactor) and rotating speed (2.5; 5 rpm) were varied. P=atmospheric pressure. The filling ratio, defined as the ratio of bottom ash to 100 ml of distilled water (UNE EN 12457-2). The pH was measured at the bottom of the reactor internal volume, and the rotating speed.	Mild	mass of sorbent=6 kg	The moisture content was measured after drying for 24 h at 105 °C, providing an average value of 26.3 ±5.2%. For pH measurement, the BA was dried, reduced in size, sieved at 5 mm and mixed in the ratio of 10 g of BA to 100 ml of distilled water (UNE EN 12457-2). The pH was measured at the bottom of the reactor internal volume, with an average value of 13.08 ±0.14, size of sorbent particles: 5-10 mm.	calimetry	35-37 gCO2/kg sorbent for rotating reactor; 21-23 gCO2/kg sorbent for fixed reactor.	An innovative process, based on carbon dioxide capture and storage by means of accelerated carbonation of bottom ash is proposed and studied for the above purpose.	

[illegible]

[illegible]

ANNEX III: Quantitative analysis table of the included papers

In this table, every line corresponds to a quantitative result in terms of gCO₂/kg dw found in the literature. Data found have been converted in selected units of measurement to be comparable each other. Every paper have been classified according various features which describes how the accelerated experimentations have been carried out.

REF	MATERIAL	OPERATIVE CONDITIONS	PRESSURE (bar)	TEMPERATURE (°C)	TIME (h)	HUMIDITY (RH %)	CO ₂ UPTAKE/g kg sorbent				CO ₂	REACTOR	MOISTURE CONTENT %	
							PUNCTUAL VALUE	MIN VALUE	MAX VALUE	GRANULOMETRY (mm) (Flow (L/min)				Conc %
Appai, G., Cara, S., Muttoni, A., Piradda, L., Poon, C. S. (2018). Sequestration of carbon dioxide in steel slag.	FLY ASHES	MILD	1	20	3		200				0		2.5	
	RCA	MILD	0.1	ROOM	1	50±5	6.5				0	100	x	
	RCA	MILD	0.1	ROOM	3	50±5	12.6				0	100	x	
	RCA	MILD	0.1	ROOM	5	50±5	24				0	100	x	
	RCA	MILD	0.1	ROOM	7	50±5	19.5				0	100	x	
	RCA	MILD	0.1	ROOM	24	50±5	19.5				0	100	x	
	RCA	INTENSIVE	1.1	ROOM	24	50±5	25.8				0	100	x	
	RCA	INTENSIVE	5	ROOM	24	50±5	28.2				0	100	x	
	RCA	MILD				50±5	55		1.00		0	100	x	
	RCA	MILD				50±5	32		3.50		0	100	x	
Sequestration of carbon dioxide in steel slag.	RCA	MILD				50±5	25		7.50		0	100	x	
	RCA	MILD				50±5	27		15.00		0	100	x	
	SLAGS	INTENSIVE	1	200	2		40		<0.25		3	100	x	
	SLAGS	INTENSIVE	1	360	2		20		<0.25		3	100	x	
	SLAGS	INTENSIVE	1	200	1		0		<0.25		3	100	x	
	SLAGS	INTENSIVE	1	200	2		40		<0.25		3	100	x	
	SLAGS	INTENSIVE	1	200	4		44		<0.25		3	100	x	
	SLAGS	INTENSIVE	1	200	6		53		<0.25		3	100	x	
	FLY ASHES	INTENSIVE	>1	400	<0.17	75	250				10	X		
	FLY ASHES	INTENSIVE	>1	30	0.17	75	250				100		0.2	
Hassan, H., Shao, Y. (2014). Carbon storage through concrete block	CONCRETE-MONOLITHES	MILD	1	ROOM	4		11.6		/		0	100	X	
			1	ROOM	4		0.6		/		0	100	X	
			1	ROOM	0		0.3		/		0	100	X	
			1	ROOM	4		28.3		/		0	100	X	
			1	ROOM	4		0.8		/		0	100	X	
			1	ROOM	0		0.4		/		0	100	X	
			1	ROOM	4		29.8		/		0	100	X	
			1	ROOM	0		0.8		/		0	100	X	
			1	ROOM	0		0.6		/		0	100	X	
			1	ROOM	4		30.5		/		0	100	X	
Yang, X., Xuan, D., & Poon, C. S. (2017). Empirical modelling of CO2 uptake by recycled			1	ROOM	4		0.9		/		0	100	X	
			1	ROOM	0		0.6		/		0	100	X	
			1	ROOM	4		31.5		/		0	100	X	
			1	ROOM	4		32		/		0	100	X	
			1	ROOM	2		28.2		/		0	100	X	
			1	ROOM	96		45.8		/		0	100	X	
			1	ROOM	0		0.9		/		0	100	X	
Yang, X., Xuan, D., & Poon, C. S. (2017). Empirical modelling of CO2 uptake by recycled	RCA	MILD	0.1		1	50	7				0	100	X	
			0.1		3	50	12.8				0	100	X	
			0.1		5	50	24				0	100	X	
			0.1		7	50	19				0	100	X	
			0.1		24	50	19				0	100	X	
		INTENSIVE	5		24	50	28				0	100	X	
		MILD	1		3	50	5.8				1	100	X	
			1		5	50	12.2				1	100	X	
			1		7	50	12.4				1	100	X	
			1		24	50	12.4				1	100	X	
			1		3	50	9.5				1	10	X	
			1		5	50	9.8				1	10	X	
			1		7	50	9.7				1	10	X	
			1		24	50	10.5				1	10	X	
			1		3	50	4				1	100	X	
			1		5	50	4.2				1	100	X	
			1		7	50	3.5				1	100	X	
			1		24	50	3.4				1	100	X	

Rostami, V., Shao, Y., & Boyd, A.J. (2011). Durability of concrete pipes subjected to combined steam and	CONCRETE-MONOLITES	INTENSIVE	1.5	24	2	30	87			/				100	X		
			1.5	24	2	30	89			/				100	X		
			1.5	24	2	30	79			/				100	X		
Zhang, D., & Shao, Y. (2016). Early age carbonation curing for precast reinforced concretes.	CONCRETE-MONOLITES	INTENSIVE	5	25	12	65	126	99.5	146	/				100		0.3-0.4	
			2	25	12	65	128			/				100		0.3-0.4	
			3	25	12	65	136			/				100		0.3-0.4	
			4	25	12	65	145			/				100		0.3-0.4	
			5	25	12	65				/				100		0.3-0.4	
Prosser, Grayson, et al. "Coupling of Attrition and Accelerated Carbonation for RCA"	RCA	INTENSIVE	1.4	22	7		47.4			<6				80	X		4
		INTENSIVE	1.4	22	7		41.2			<7				80	X		4
		MILD	1	22	7		17.2			<8				80	X		4
		MILD	1	22	7		30.4			<9				80	X		4
		MILD	1	22	7		30.8			<10				80	X		4
		MILD	1	22	7		12			<11				80	X		9
		MILD	1	22	7		15.3			<12				80	X		4
		MILD	1	22	7		4.2			<13				80	X		2
		MILD	1	22	7		9.2			<14				80	X		10
		MILD	1	22	7		15.5			<15				80	X		4
Schnabel, Kevin, et al. "Technically Exploitable Mineral Carbonation"	FLY ASHES	MILD	1		168		92.84			<3				100		0.3-0.2	
			1		168		43.56			<3				100		0.3-0.3	
			1		168		76.56			<3				100		0.3-0.4	
	SLAGS	MILD	1		1		111.76			<3				100		0.3-0.5	
Josecurran, David, et al. "Accelerated Carbonation of Fresh Concrete"	NATURAL AGGREGATES	MILD	1	21	24	65	1.5			<0.5				5	X		
				21	72	65	5.173			<0.5				5	X		
				21	168	65	5.88			<0.5				5	X		
	RCA	MILD	1	21	24	65	2.076			<0.5				5	X		
				21	72	65	5.452			<0.5				5	X		
				21	168	65	8.317			<0.5				5	X		
				21	24	65	14.005			<0.5				5	X		
				21	72	65	15.846			<0.5				5	X		
				21	168	65	16.549			<0.5				5	X		
Simoes, et al. "Effect of the Accelerated Carbonation Treatment on the	RCA	MILD	1	21	7		2.6			<6				80	x		0
				21	7		4			<6				80	x		2
				21	7		23.5			<6				80	x		4
				21	7		16.2			<6				80	x		5
				21	7		13.5			<6				80	x		6
				21	7		15			<6				80	x		7
				21	7		14.3			<6				80	x		8
				21	7		12			<6				80	x		9
				21	7		8			<6				80	x		10
				21	7		11.8			<6				80	x		12
				21	21		27.5			<6				80	x		4
				21	21		30			<6				80	x		6
				21	21		39.5			<6				80	x		8
Berger, Hakan, et al. "Accelerated Carbonation"	FLY ASHES	MILD	1	ROOM	0.5		45.62			4 < SIZE < 10				20		0.2-0.5	
					0.5		39.71			4 < SIZE < 10				20		0.2-0.5	
					0.5		108.06			4 < SIZE < 10				20		0.2-0.5	
					0.5		54.45			4 < SIZE < 10				20		0.2-0.5	
					0.5		23.42			4 < SIZE < 10				20		0.2-0.5	

Ukwattage, N. L. "Investigation of the Potential of Coal Combustion Fly Ash for Mineral Sequestration of CO ₂ "	FLY ASHES	INTENSIVE	3	40	10	7.66					0,9		100	x	1:5	
et al. "Accelerated Carbonation of Municipal Solid Waste"	BOTTOM ASHES	MILD	1	20	0,5	3	75	0,5	10	7.66	>4,75		20		0,2	
					0,5	4	75	0,5			2,36<SIZE<4,75		20		0,2	
					0,5	5	75	0,5			1,18<SIZE<2,36		20		0,2	
					0,5	18	75	0,5			0,6<SIZE<1,18		20		0,2	
					0,5	50	75	0,5			0,3<SIZE<0,6		20		0,2	
					0,5	84	75	0,5			0,15<SIZE<0,3		20		0,2	
					0,5	165	75	0,5			<0,15		20		0,2	
Chang, E., et al. "CO ₂ Sequestration by Carbonation of Steelmaking Slags in Water"	SLAGS	INTENSIVE	5-9	160	12	127					<0,044		100		10 mL/g	
						107					<0,044					

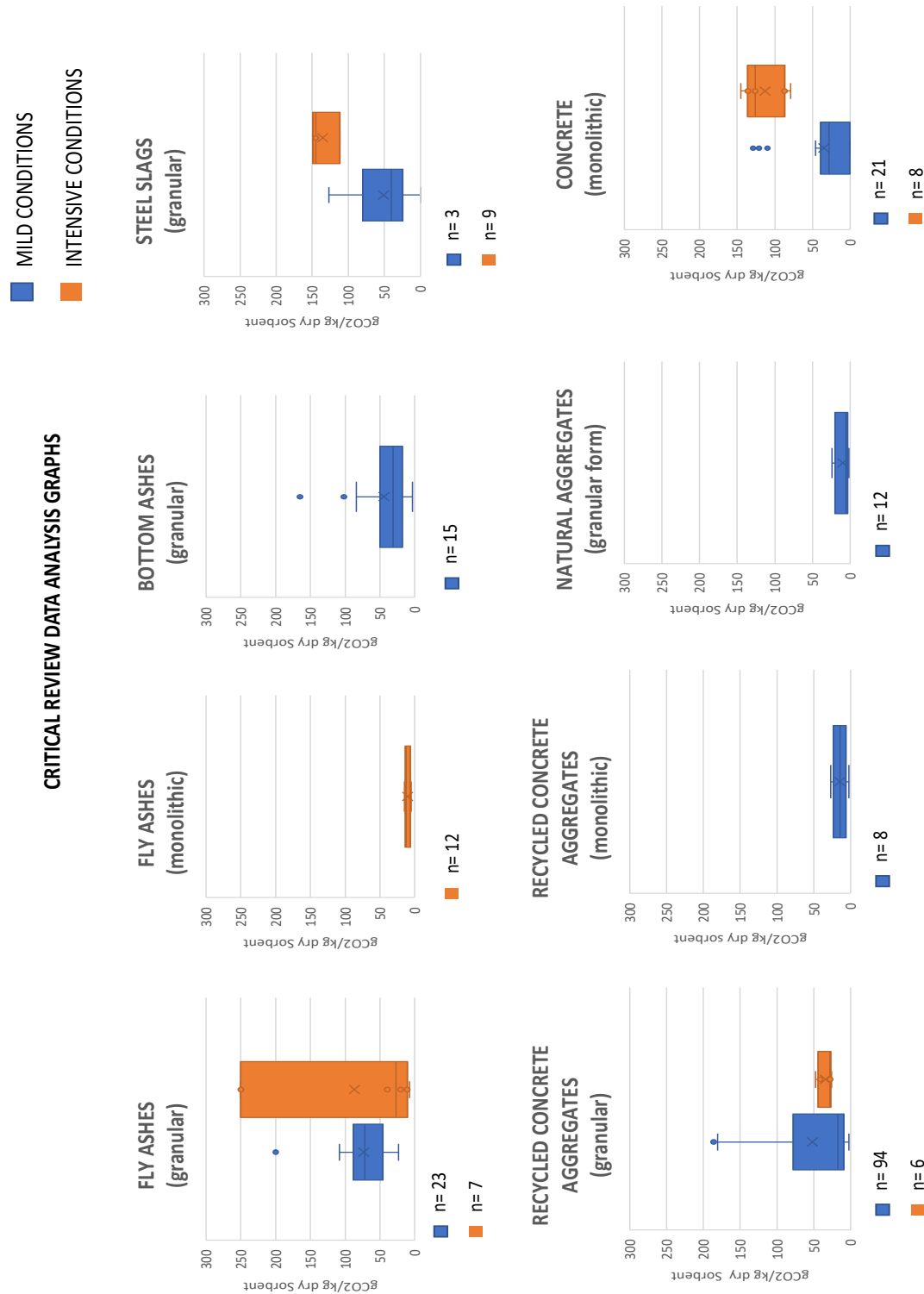
ANNEX IV: Mathematical models calculations

Here theoretical models found in literature are presented. Theoretical models relate the potential CO₂ uptake of the sample given the % mass of chemical species which compose the sample material. The theoretical formulations are present in chapter 1.4 of the introduction. The initial idea was to test their validity through the % mass of chemicals present in our material and to include it inside the thesis. Since we didn't obtain the XRD (X-ray diffractometric) analyses, which could give us the quantitative analyses of the chemical present in the material sorbent, it was not possible to compile these Excel table. Thus, the same fictitious values (1-2-3-4-5) have been inserted in the cells to show the difference in results of the different models.

THEORETICAL MODELS									
Nam, Seong-Young, et al. "Accelerated"	BOTTOM ASHES	CaO	MgO	Na ₂ O	K ₂ O	SO ₃			
	coefficient	0,785	1,091	1,41	0,935	-0,55			
	Mass Fraction (%)	1	2	3	4	5			
	ThCO ₂ uptake (% mass)	8,187							
Xuan, D.; Poon, C. S. (2018). Sequestration of carbon	RCA	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃		
	coefficient	0,785	1,09	0,71	0,47	-0,5495	-0,4396		
	Mass Fraction (%)	/	1	2	3	4	5	6	
	ThCO ₂ uptake (% mass)	1,5899							
Fang, X., Xuan, D., & Poon, C. S. (2017). Empirical modelling of	RCA	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃		
	coefficient	0,785	1,09	0,71	0,47	-0,5495	-0,4396		
	Mass Fraction (%)	/	1	2	3	4	5	6	
	ThCO ₂ uptake (% mass)	1,5899							
Yuan, Q., Yang, G., Zhang, Y., Wang, T., Wang, L.	FLY ASHES	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃		
	coefficient	0,78	1,1	/	/	-0,55	/		
	Mass Fraction (%)	/	1	2	/	/	3		
	ThCO ₂ uptake (% mass)	1,33							
Schnabel, Kevin, et al. "Technically Exploitable Mineral Carbonation"	STEEL SLAGS - FA - BA	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃	KCl	
	coefficient	0,785	1,091	0,71	0,468	-0,5495	-0,4396	0,2958	
	Mass Fraction (%)	/	1	2	3	4	5	6	7
	ThCO ₂ uptake (% mass)	3,6545							
Pei, Si-Lu, et al. "Efficacy of Carbonated Petroleum Coke Fly"	FLY ASHES	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃		
	coefficient	0,785	1,091	/	0,935	-0,5495	-0,4396		
	Mass Fraction (%)	1	2	/	3	4	5		
	ThCO ₂ uptake (% mass)	1,376							
Chang, E. E., et al. "Accelerated Carbonation"	BOTTOM ASHES	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	CaCO ₃		
	coefficient	0,786	/	/	/	-0,55	-0,44		
	Mass Fraction (%)								
	ThCO ₂ uptake (% mass)								

ANNEX V: Box plot Mild and Intensive conditions

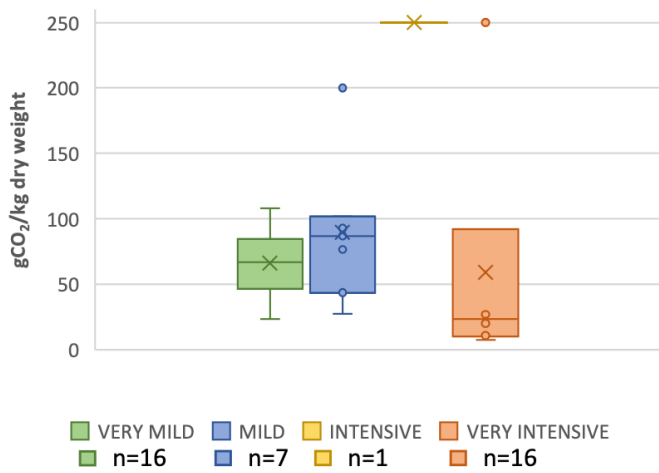
Results of carbon uptake for the different material classes and for the different operative conditions are shown. This classification for the operative conditions indicates such that: Mild conditions correspond to pressure=1 bar, T=room temperature; Intensive conditions correspond to pressure>1 bar, t>35°C. Are also shown the number of records found per box plot.



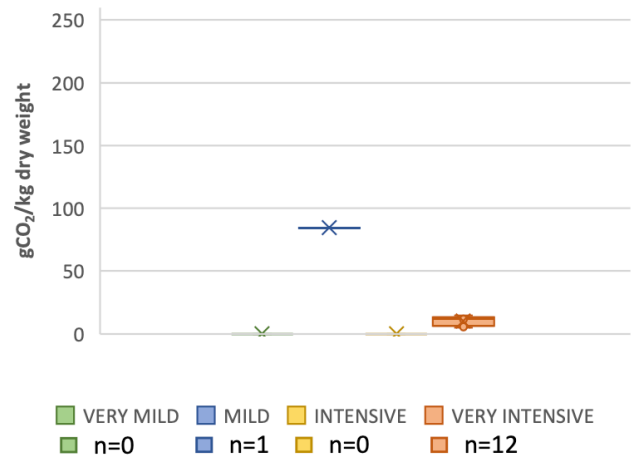
ANNEX VI: Box plot Very Mild, Mild, Intensive, Very intensive conditions

Results of carbon uptake for the different material classes and for the different operative conditions are shown. Also shown the number of records found per box plot are shown. These graphs are present in paragraph 2.1.3 Critical review, inside the scientific paper (Part II) where detailed description is given.

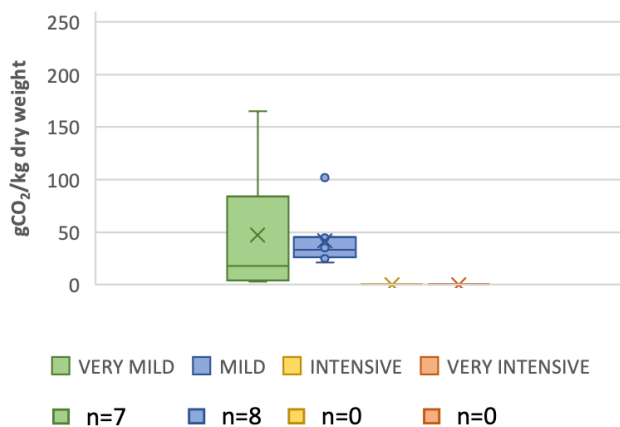
**FLY ASHES
(granular form)**



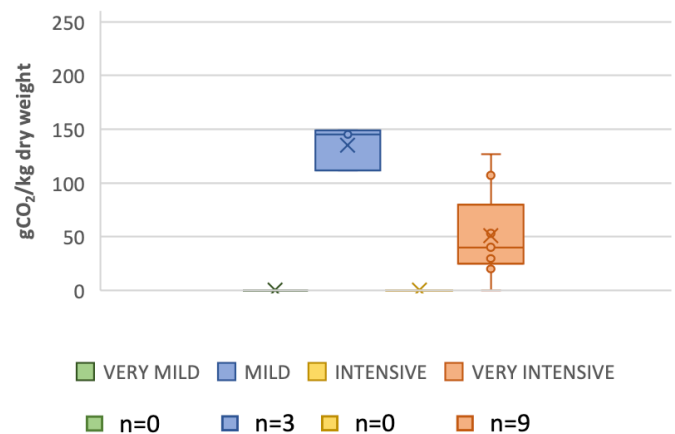
**FLY ASHES
(monolithic form)**



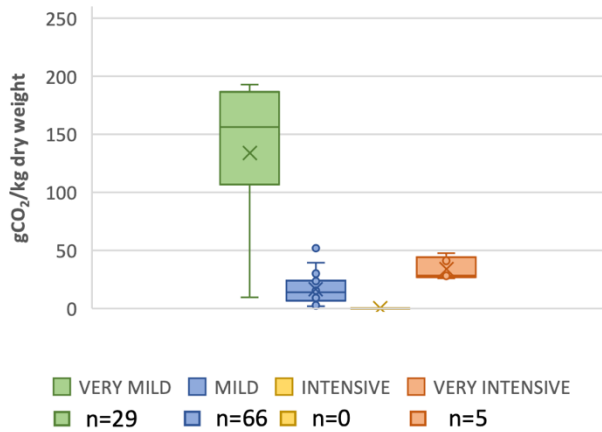
**BOTTOM ASHES
(granular form)**



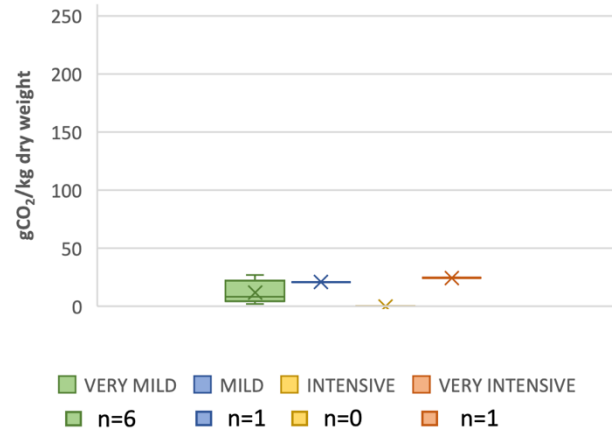
**SLAGS
(granular form)**



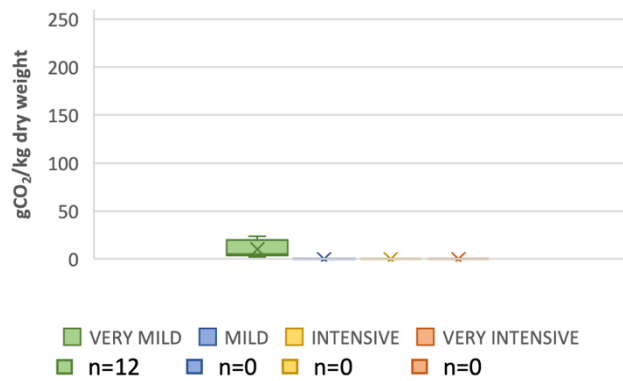
RCA (granular form)



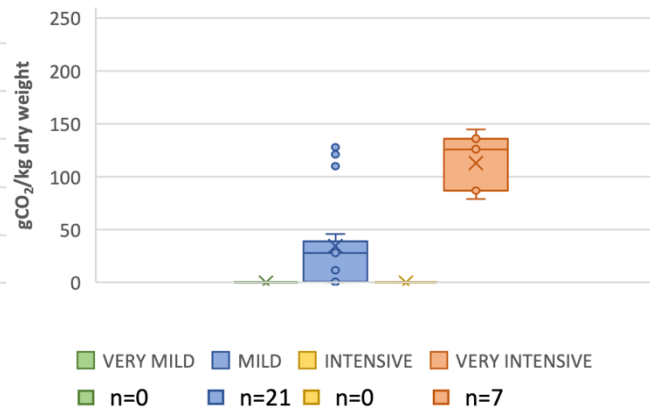
RCA (monolithic form)



NATURAL AGGREGATES (granular form)



CONCRETE (monolithic form)

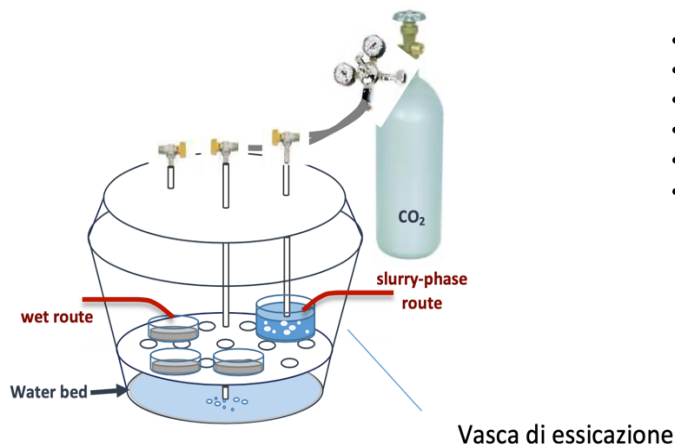


ANNEX VII: Schematic of the Accelerated carbonation setup experiment

The following images refer to the schematic of the experimental setup found of the studies included in the review. Operative conditions are also present.

Cappai, G., Cara, S., Muntoni, A., Piredda, M., 2012. Application of accelerated carbonation on MSW combustion APC residues for metal immobilization and CO₂ sequestration. J. Hazard. Mater. 207–208, 159–164.

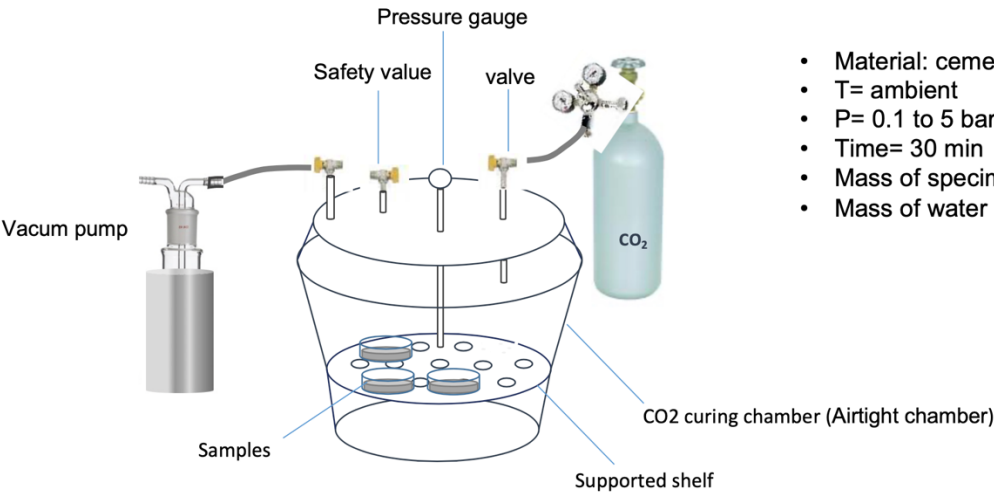
doi:10.1016/j.jhazmat.2011.04.013



- Material: MSW combustion residues
- Process: Single step aqueous route process
- T= ambient (20°C)
- P= Patm
- Time= 3h
- pH monitored

He, P., Shi, C.; Poon, C. S. (2018). Methods for the assessment of carbon dioxide absorbed by cementitious materials. Carbon Dioxide Sequestration in Cementitious Construction Materials (pp. 103–126). Elsevier. doi.org/10.1016/B978-0-08-102444-7.00006-X

1st Method: **Mass gain of the specimen**

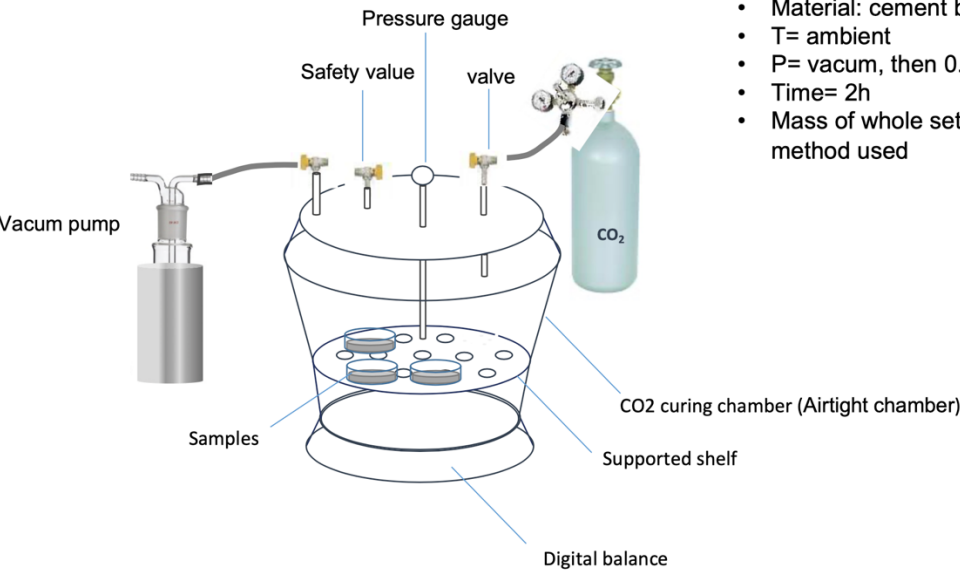


- Material: cement based material, fly ashes
- T= ambient
- P= 0.1 to 5 bar 100% CO₂
- Time= 30 min
- Mass of specimen monitored
- Mass of water evaporated is monitored

Schematic of the CO₂ curing equipment

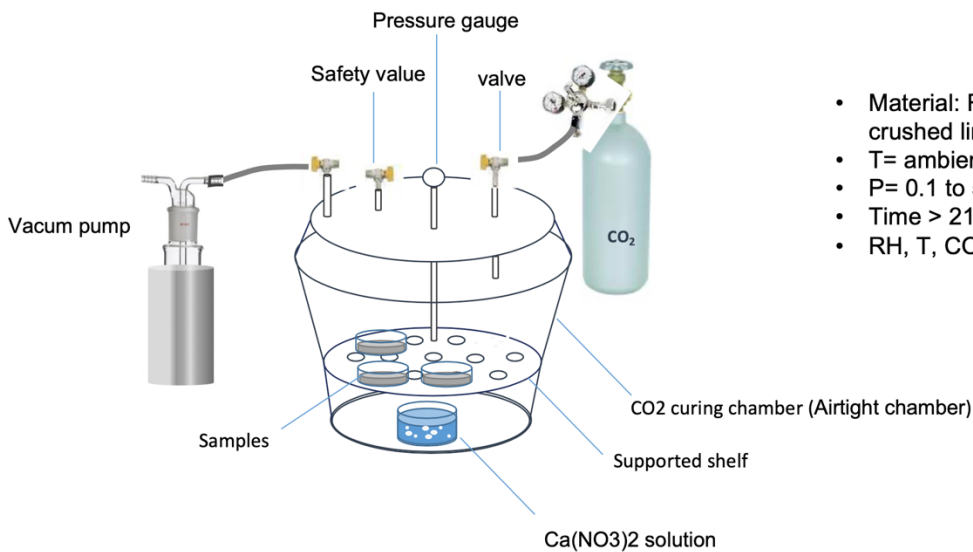
Schematic of the CO₂ curing equipment

Method: **Mass change of the whole test set-up**



- Material: cement based material, fly ashes
- T= ambient
- P= vacuum, then 0.1 to 4 bar 100% CO₂
- Time= 2h
- Mass of whole setup monitored: cumulative mass change method used

Sojobi, A. O., Xuan, D., Li, L., Liu, S.; Poon, C. S. (2021). Optimization of gas-solid carbonation conditions of recycled aggregates using a linear weighted sum method. *Developments in the Built Environment*, doi.org/10.1016/j.dibe.2021.100053



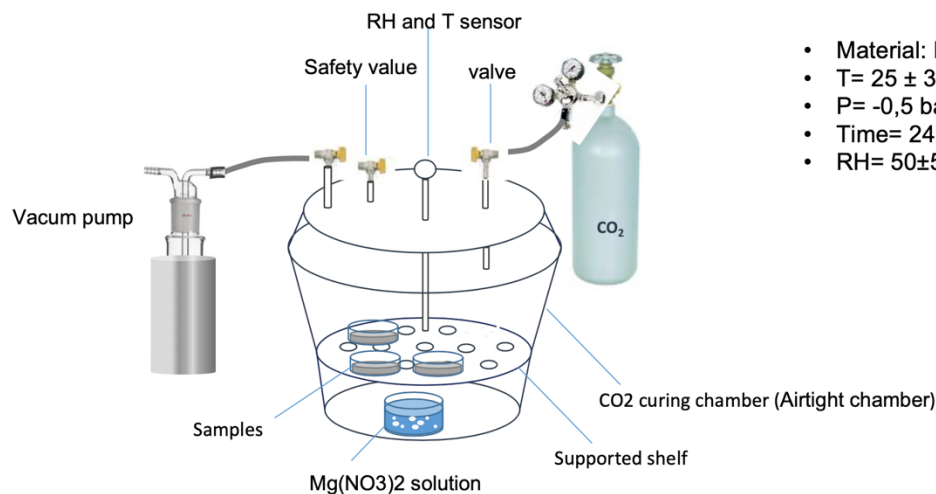
- Material: Recycled gravel aggregate, recycled crushed limestone
- T= ambient (18°C)
- P= 0.1 to 5 bar
- Time > 210 min
- RH, T, CO2 pressure monitored

4

Xuan, D.; Poon, C. S. (2018). Sequestration of carbon dioxide by RCAs and enhancement of properties of RAC by accelerated carbonation. *New Trends in Eco-efficient and Recycled Concrete* (pp. 477–497). Elsevier. doi.org/10.1016/B978-0-08-102480-5.00016-6

Schematics of carbonation chambers of RCA

Method 1: pressurised carbonation in a chamber with 100% CO2 concentration

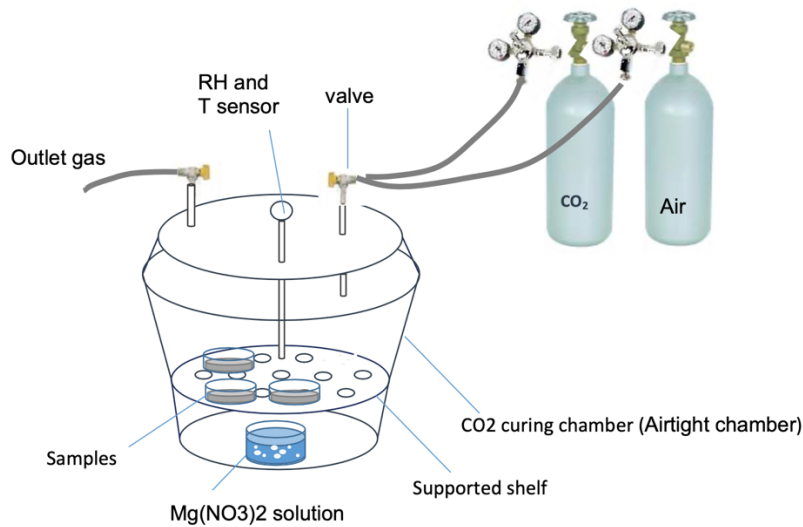


- Material: Recycled cement aggregates
- T= 25 ± 3°C
- P= -0,5 bar vacuum, then 0,1-5 bar 100% CO2
- Time= 24h
- RH= 50±5 %

5

Schematics of carbonation chambers of RCA

Method 2: gas flow carbonation, open reactor.



- Material: Recycled cement aggregates
- T= 25 ± 3°C
- Gas flow air+CO2 = 1-10 L/min
- Time= 24h
- RH= 50±5 %

El-Hassan, H.; Shao, Y. (2014). Carbon Storage through Concrete Block Carbonation. Journal of Clean Energy Technologies, 287–291.
<https://doi.org/10.7763/jocet.2014.v2.141>

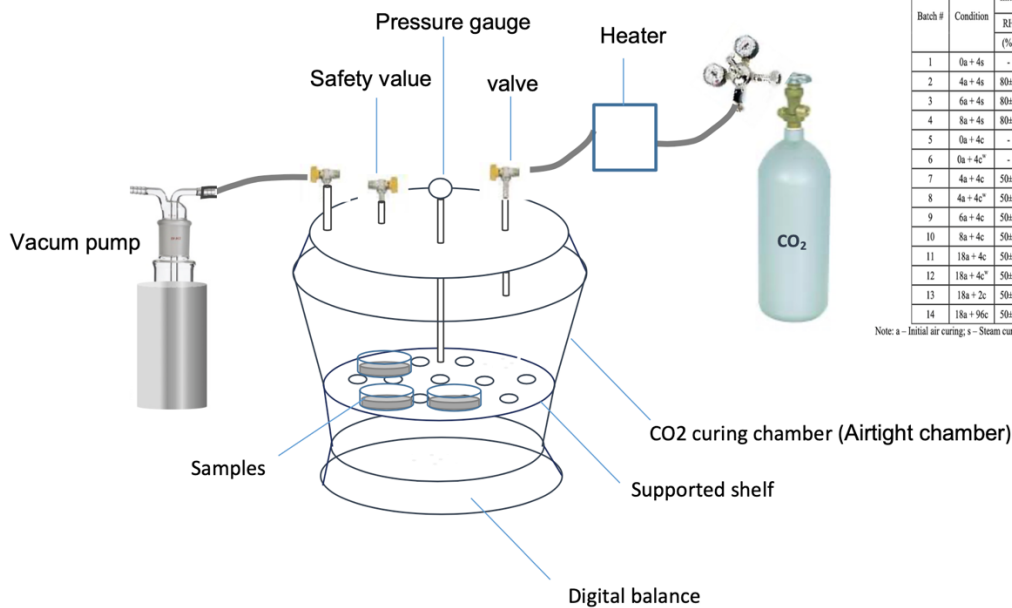
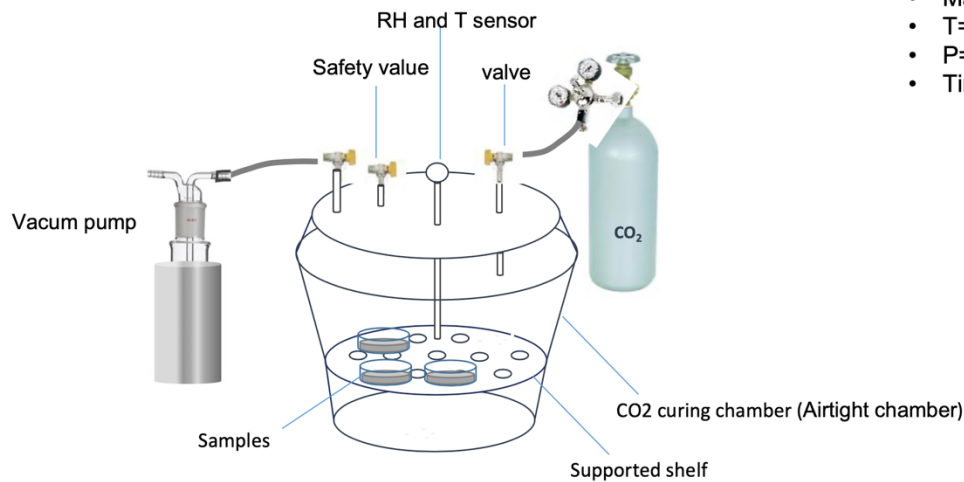


TABLE II: CURING PROCEDURES											
Batch #	Condition	Initial curing			Steam curing			Carbonation curing		Subsequent Hydration	
		RH (%)	T (°C)	t (hours)	RH (%)	T (°C)	t (hours)	t (hours)	Water Spray (g)	t (days)	
1	0a + 4s	-	-	0	95±5	75±5	4	-	-	28	
2	4a + 4s	80±5	22±1	4	95±5	75±5	4	-	-	28	
3	6a + 4s	80±5	22±1	6	95±5	75±5	4	-	-	28	
4	8a + 4s	80±5	22±1	8	95±5	75±5	4	-	-	28	
5	0a + 4c	-	-	0	-	-	-	4	-	28	
6	0a + 4c*	-	-	0	-	-	-	4	1±0.2	28	
7	4a + 4c	50±1	25±0.2	4	-	-	-	4	-	28	
8	4a + 4c*	50±1	25±0.2	4	-	-	-	4	17±2	28	
9	6a + 4c	50±1	25±0.2	6	-	-	-	4	-	28	
10	8a + 4c	50±1	25±0.2	8	-	-	-	4	-	28	
11	18a + 4c	50±1	25±0.2	18	-	-	-	4	-	28	
12	18a + 4c*	50±1	25±0.2	4	-	-	-	4	29±2	28	
13	18a + 2c	50±1	25±0.2	18	-	-	-	2	-	28	
14	18a + 96c	50±1	25±0.2	18	-	-	-	96	-	28	

Note: a – Initial air curing; s – Steam curing; c – Carbonation; RH – Relative humidity; T – Temperature; t – Time; W – Water sprayed after carbonation

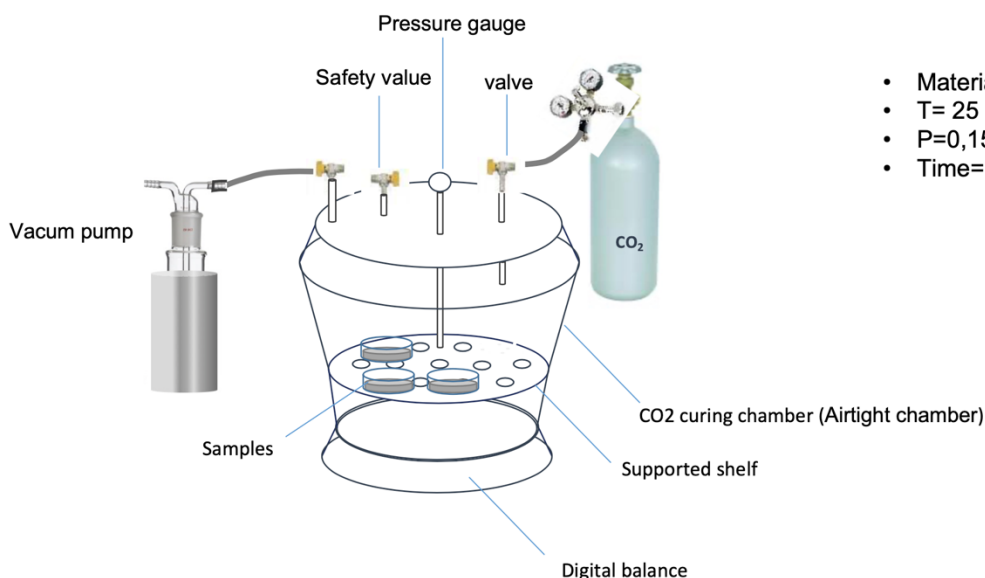
P=1 bar

Tu, Z., Guo, M. Z., Poon, C. S; Shi, C. (2016). Effects of limestone powder on CaCO_3 precipitation in CO_2 cured cement pastes. *Cement and Concrete Composites*, 9–16. <https://doi.org/10.1016/j.cemconcomp.2016.05.019>



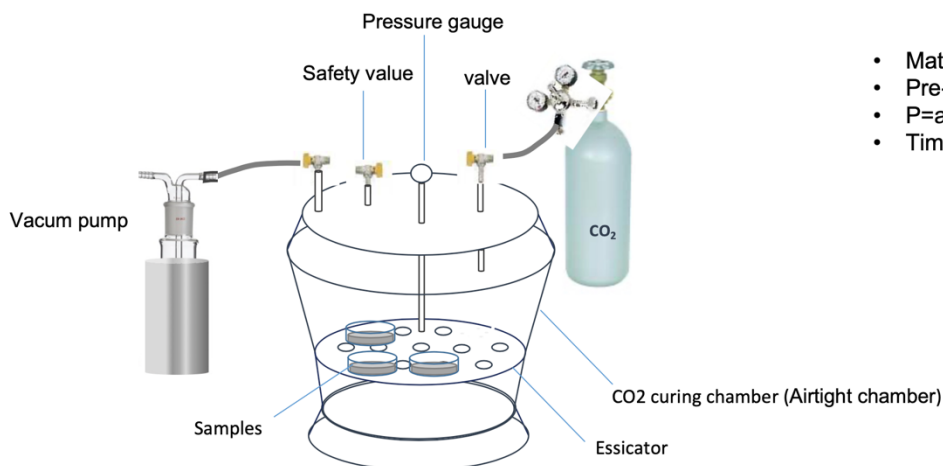
- Material: Portland cement
- $T = \text{ambient}$
- $P = 0,1 \text{ to } 4 \text{ bar}$
- Time = 24h

Fang, X., Xuan, D., & Poon, C. S. (2017). Empirical modelling of CO_2 uptake by recycled concrete aggregates under accelerated carbonation conditions. *Materials and Structures/Materiaux et Constructions*. <https://doi.org/10.1617/s11527-017-1066-y>



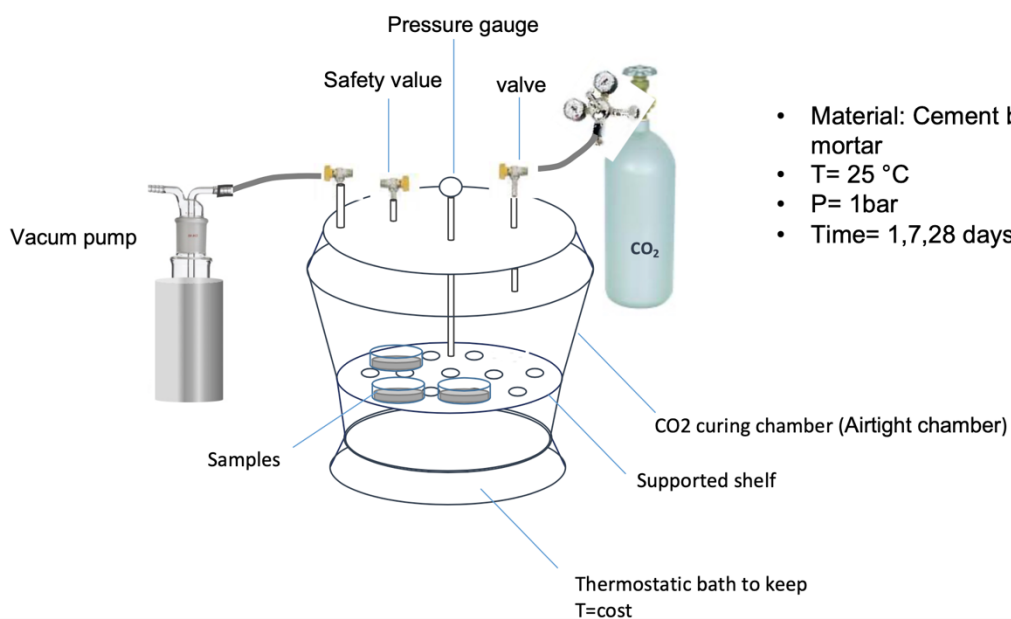
- Material: Portland limestone cement
- $T = 25^\circ\text{C}$
- $P = 0,15 \text{ MPa}$
- Time = 2h

Sereng, M.; Dangla, P. (n.d.). *Accelerated carbonation of recycled concrete aggregates*. <https://www.researchgate.net/publication/352366234>



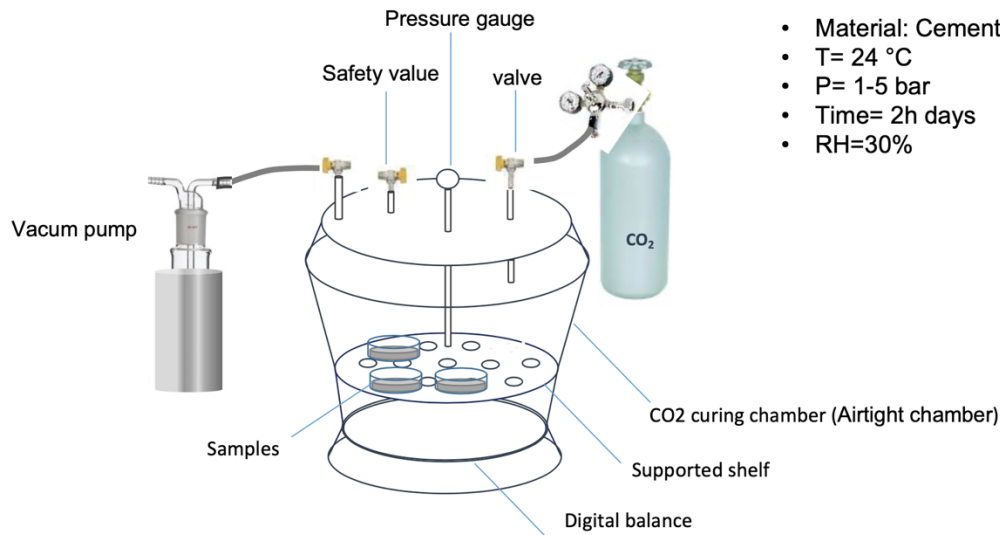
- Material: Recycled cement aggregates
- Pre-treatment in which water content is set
- P=atmospheric
- Time= 24h

Furcas, C., Balletto, G., Naitza, S., & Mazzella, A. (2014). Evaluation of CO₂ uptake under mild accelerated carbonation conditions in cement-based and lime-based mortars. *Advanced Materials Research*, 980, 57–61. <https://doi.org/10.4028>



- Material: Cement based mortar and limestone based mortar
- T= 25 °C
- P= 1bar
- Time= 1,7,28 days

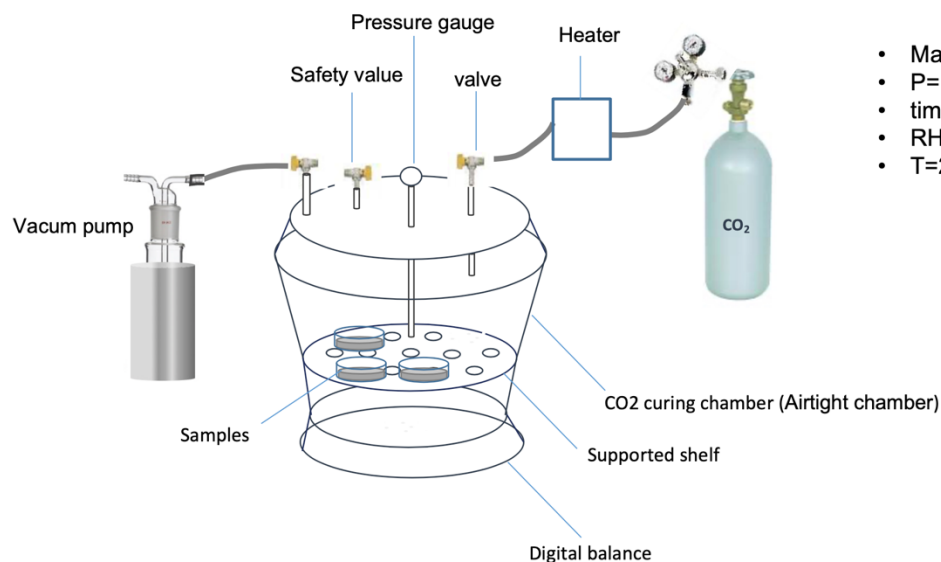
Rostami, V., Shao, Y., & Boyd, A. J. (2011). Durability of concrete pipes subjected to combined steam and carbonation curing. *Construction and Building Materials*, 3345–3355. <https://doi.org/10.1016/j.conbuildmat.2011.03.025>



- Material: Cement based concrete pipes
- T= 24 °C
- P= 1-5 bar
- Time= 2h days
- RH=30%

14

Zhang, D.; Shao, Y. (2016). Early age carbonation curing for precast reinforced concretes. *Construction and Building Materials*, 113, 134–143.



- Material: OPC (ordinary portland concrete)
- P=1-5 bar;
- time of carbonation=12h;
- RH= 65 ± 5%;
- T=25±3°C.

15

Yuan, Q., Yang, G., Zhang, Y., Wang, T., Wang, J.; Romero, C. E. (2022). Supercritical CO₂ coupled with mechanical force to enhance carbonation of fly ash and heavy metal solidification. *Fuel*, 315. <https://doi.org/10.1016/j.fuel.2022.123154>

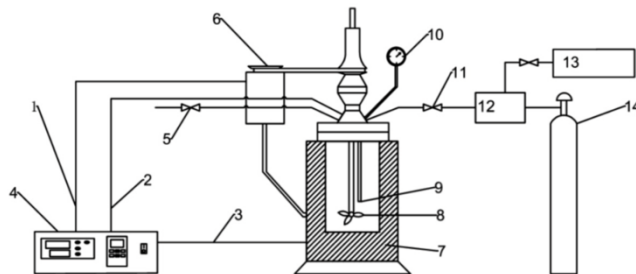


Fig. 1. Schematic diagram of supercritical CO₂ carbonation reaction platform
1: speed control; 2: temperature control; 3: heating control; 4: operation panel;
5: exhaust valve; 6: stirring motor; 7: heating jacket; 8: stirrer; 9: thermocouple;
10: pressure gauge; 11: intake valve; 12: booster pump; 13: compressed air; 14:
CO₂ gas source.

- Material: fly ashes
- Here stirring process is present to increase CO₂ permeability
- P=1-8 Mpa
- T=40-60-80 °C
- T=5 hours

Dos Reis, Glaydson S., et al. "Coupling of Attrition and Accelerated Carbonation for CO₂ Sequestration in Recycled Concrete Aggregates." *Cleaner Engineering and Technology*, vol. 3, Elsevier Ltd., 2021,

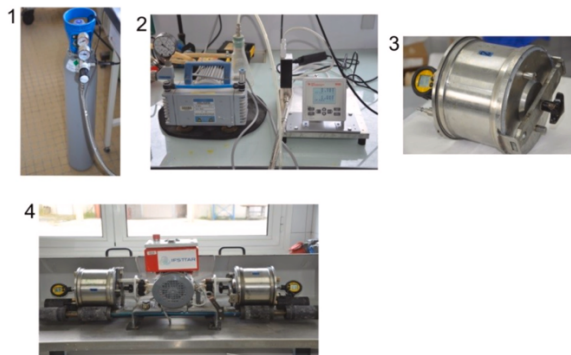
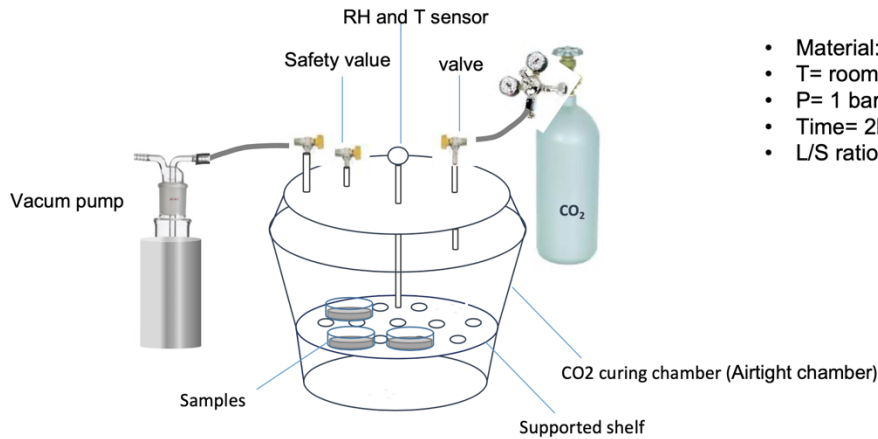


Fig. 1. Set-up of the laboratory-scale rolling carbonation experiments. (1) CO₂ gas cylinder; (2) vacuum pump and CO₂ flow automated controller; (3) hermetic vessel (with pressure sensor) for placing the concrete aggregates; (4) rolling machine.

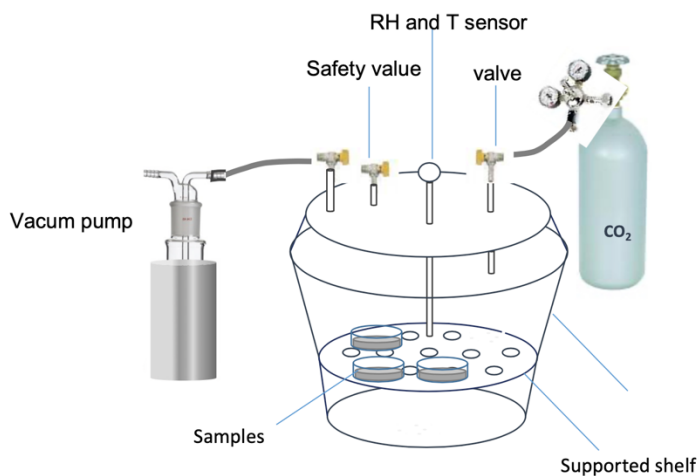
- Material: RCA
- P_{partial} pressure=0,75 bar 100%CO₂ in a P=1bar air test;
- time of carbonation=;
- T=22°C.
- Presence of a rolling machine (rotatory drum)

Schnabel, Kevin, et al. "Technically Exploitable Mineral Carbonation Potential of Four Alkaline Waste Materials and Effects on Contaminant Mobility." *Greenhouse Gases : Science and Technology.*, vol. 11, no. 3, John Wiley & Sons, 2021, pp. 506–19, <https://doi.org/10.1002/ghg.2063>



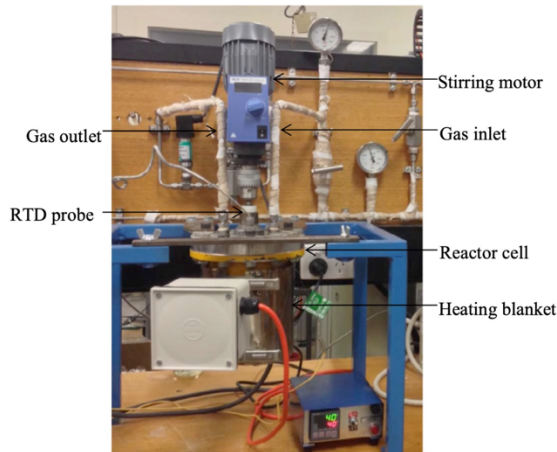
- Material: Fly ashes, Steel slags
- T= room
- P= 1 bar
- Time= 2h-168 h
- L/S ratio was varied from 0,1 to 0,5

Wang, Dongxing, et al. "Quantification and Micro-Mechanisms of CO2 Sequestration in Magnesia-Lime-Fly Ash/slag Solidified Soils." *International Journal of Greenhouse Gas Control.*, vol. 91, Elsevier Science,, 2019, <https://doi.org/10.1016/j.ijggc.2019.102827>.



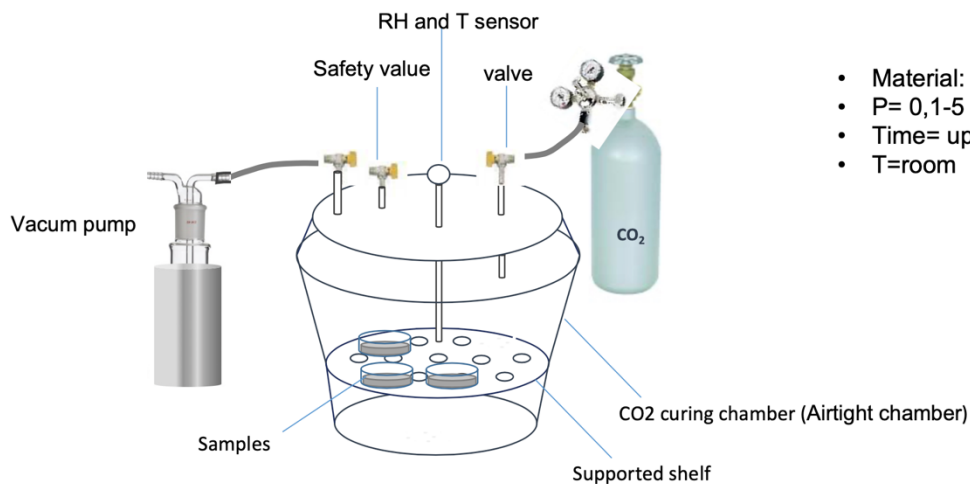
- Material: Fly ashes, Steel slags, clay
- T= room
- P= 150 to 300 kPa
- Time= 0.5, 1, 3, 6, 12 and 24 h

Ukwattage, NL, et al. "Steel-Making Slag for Mineral Sequestration of Carbon Dioxide by Accelerated Carbonation." *Measurement*, vol. 97, Published for IMEKO by the Institute of Measurement and Control, 2017, pp. 15–22, <https://doi.org/10.1016/j.measurement.2016.10.057>.



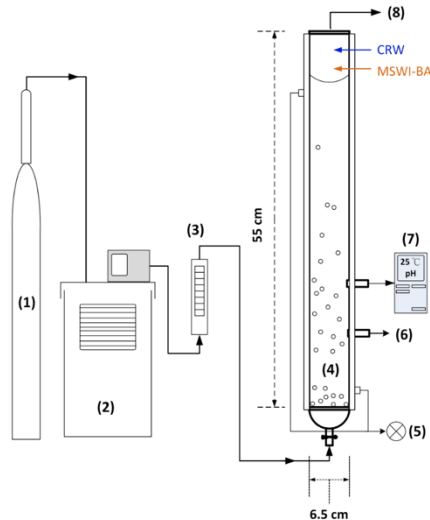
- Material: steel slags
- $P = 3\text{--}6\text{ MPa}$
- Time = 48h
- L/S ratio = 0,25-3
- $T = 40^\circ\text{C}$
- Rotation = 60 rpm

Xuan, Dongxing, et al. "Development of a New Generation of Eco-Friendly Concrete Blocks by Accelerated Mineral Carbonation." *Journal of Cleaner Production*, vol. 133, Elsevier Science Ltd, 2016, pp. 1235–41, <https://doi.org/10.1016/j.jclepro.2016.06.062>.



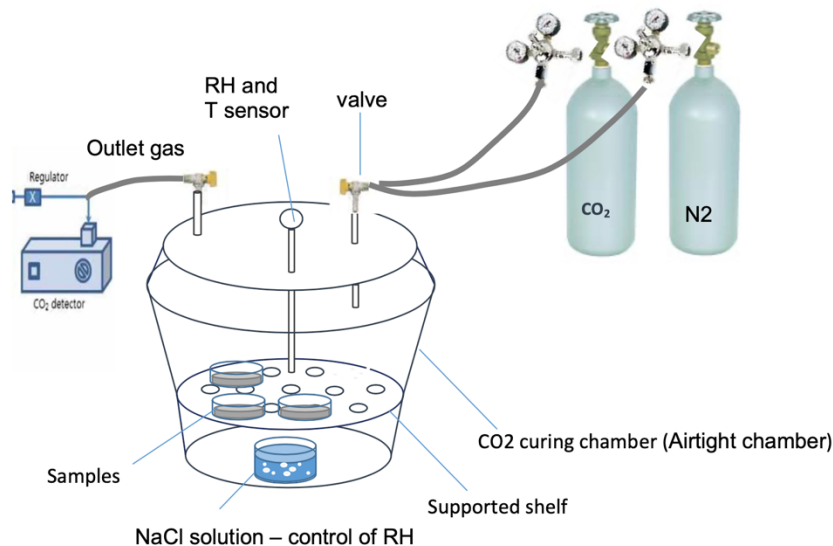
- Material: concrete
- $P = 0,1\text{--}5\text{ bar}$
- Time = up to 24h
- $T = \text{room}$

Chang, E. E., et al. "Accelerated Carbonation Using Municipal Solid Waste Incinerator Bottom Ash and Cold-Rolling Wastewater: Performance Evaluation and Reaction Kinetics." *Waste Management*, vol. 43, Pergamon Press,, 2015, pp. 283–92, <https://doi.org/10.1016/j.wasman.2015.05.001>.



- Material: Bottom ashes
- $P = 1$ atm
- Time = 120 min
- L/S ratio = 5, 10, 20, 40 mL/g
- $T = 25^\circ\text{C}$
- CO_2 flowrate = 0,2; 0,5; 1; 1,5 L/min

Nam, Seong-Young, et al. "Accelerated Carbonation of Municipal Solid Waste Incineration Bottom Ash for CO_2 Sequestration." *Geosystem Engineering*, vol. 15, no. 4, 2012, pp. 305–11, <https://doi.org/10.1080/12269328.2012.732319>.



- Material: Bottom ashes
- $P = 1$ atm; conc of CO_2 in N_2 10-30%
- L/S ratio = 0,1-0,3
- $T = 20-40-60^\circ\text{C}$
- $\text{RH} = 75\%$

ANNEX VIII: Calcimetry results and CO₂ content calculations

In the following tables, the calculations to get the CO₂ content of the samples tested through calcimetry is presented. Starting from the sample mass tested (ca. 5g) and the volume of CO₂ obtained with reaction with HCl (in mL) it is possible to calculate the moles of CO₂ which spilled from the reactor and so the gCO₂/kg dw. The formula utilized to pass from volume to moles of carbon dioxide was the ideal gas law. For detailed process and calculations, see paragraph 2.2.4 and 2.2.5. Letters A, B, C, stand for the different granulometries: namely 0/6mm, 6/16mm, 16/31mm. Numbers 00, 02, 03 refer to the L/S applied. t0, 02, 04, 08 are related to the carbonation treatment duration. a, b, c are the letters related to the replicate samples (of 200g each) which have been carbonated. a1, a2, a3 are the calcimetry tests (executed in triplicate per each sample of carbonated material). Outliers have been removed.

Calcimetry tests and results for 0/6 mm, L/S=0.0 L/kg, t=0 to 8h.

06-00	V(mL)	V (L)	n (mol)	m CO ₂ (g)	mass TQ (g)	TS %	dry mass (g)	gCO ₂ /g dw	CO ₂ [gCO ₂ kg ⁻¹ dw]
A/00/t0a1	34,37179863	0.034371799	0.001404915	0.061816256	5,5	86,08	4,734257625	0.013057223	13,06
A/00/t0a2	18,09042033	0.01809042	0.000739429	0.032534872	6,11	86,08	5,259329835	0.006186125	6,19
A/00/t0a3	48,84413489	0.048844135	0.001996458	0.087844153	5,73	86,08	4,932235671	0.017810121	17,81
A/00/t0b1	39,79892472	0.039798925	0.001626744	0.071576717	5,55	86,08	4,777296331	0.014982683	14,98
A/00/t0b2	61,50742912	0.061507429	0.002514058	0.110618563	5,7	86,08	4,906412448	0.022545712	22,55
A/00/t0b3	37,98988269	0.037989883	0.001552801	0.06832323	5,04	86,08	4,338301533	0.015748843	15,75
A/00/t0c1	54,27126099	0.054271261	0.002218287	0.097604615	5,09	86,08	4,381340239	0.022277342	22,28
A/00/t0c2	45,22605082	0.045226051	0.001848572	0.081337179	5,15	86,08	4,432986685	0.018348167	18,35
A/00/t0c3	28,94467253	0.028944673	0.001183086	0.052055794	5,42	86,08	4,665395696	0.011157852	11,16
A/00/02a1	72,36168132	0.072361681	0.002957716	0.130139486	5,59	86,08	4,811727295	0.027046314	27,05
A/00/02a2	57,88934505	0.057889345	0.002366172	0.104111589	5,82	86,08	5,009705342	0.020781979	20,78
A/00/02a3	54,27126099	0.054271261	0.002218287	0.097604615	5,51	86,08	4,742865366	0.020579251	20,58
A/00/02b1	79,59784945	0.079597849	0.003253487	0.143153435	5,6	86,08	4,820335037	0.029697818	29,70
A/00/02b2	65,12551318	0.065125513	0.002661944	0.117125538	5,3	86,08	4,562102802	0.025673586	25,67
A/00/02b3	83,21593351	0.083215934	0.003401373	0.149660409	5,68	86,08	4,889196966	0.030610427	30,61
A/00/02c1	72,36168132	0.072361681	0.002957716	0.130139486	5,5	86,08	4,734257625	0.02748889	27,49
A/00/02c2	61,50742912	0.061507429	0.002514058	0.110618563	5,05	86,08	4,346909274	0.025447636	25,45
A/00/02c3	32,56275659	0.032562757	0.001330972	0.058562769	5,02	86,08	4,321086051	0.013552789	13,55
A/00/04a1	70,55263928	0.070552639	0.002883773	0.126885999	5,33	86,08	4,587926026	0.027656505	27,66
a2	79,59784945	0.079597849	0.003253487	0.143153435	5,92	86,08	5,095782753	0.028092531	28,09
a3	79,59784945	0.079597849	0.003253487	0.143153435	5,37	86,08	4,62235699	0.030969792	30,97
A/00/04b1	79,59784945	0.079597849	0.003253487	0.143153435	5,92	86,08	5,095782753	0.028092531	28,09
b2	50,65317692	0.050653177	0.002070401	0.09109764	5,58	86,08	4,803119554	0.018966349	18,97
b3	72,36168132	0.072361681	0.002957716	0.130139486	5,41	86,08	4,656787955	0.027946191	27,95
A/00/04c1	39,79892472	0.039798925	0.001626744	0.071576717	6,03	86,08	5,190467905	0.013790032	13,79
c2	72,36168132	0.072361681	0.002957716	0.130139486	5,44	86,08	4,682611178	0.027792076	27,79
c3	72,36168132	0.072361681	0.002957716	0.130139486	5,01	86,08	4,312478309	0.030177424	30,18
A/00/08a1	79,59784945	0.079597849	0.003253487	0.143153435	5,49	86,08	4,725649884	0.030292857	30,29
a2	72,36168132	0.072361681	0.002957716	0.130139486	5,09	86,08	4,381340239	0.029703123	29,70
a3	86,83401758	0.086834018	0.003549259	0.156167383	5,65	86,08	4,863373742	0.032110916	32,11
A/00/08b1	104,9244379	0.104924438	0.004288688	0.188702255	6,07	86,08	5,22489887	0.036115963	36,12
b2	57,88934505	0.057889345	0.002366172	0.104111589	5,21	86,08	4,484633132	0.023215185	23,22
b3	86,83401758	0.086834018	0.003549259	0.156167383	5,57	86,08	4,794511813	0.032572114	32,57
A/00/08c1	72,36168132	0.072361681	0.002957716	0.130139486	5,99	86,08	5,156036941	0.025240216	25,24
c2	68,74359725	0.068743597	0.00280983	0.123632512	5,4	86,08	4,648180214	0.026598046	26,60
c3	81,40689148	0.081406891	0.00332743	0.146406922	5,39	86,08	4,639572473	0.031556124	31,56

Calcimetry tests and results for 0/6 mm, L/S=0.2 L/kg, t=0 to 8h.

6-02										massa TQ (g)				
A/02/10a1	41,60796676	0,041607967	0,001700686					0,074830205		5,04	71,9197471	3,624755255	0,020644209	20,64
a2	37,98988269	0,037989883	0,001552801					0,06832323		5,16	71,9197471	3,711058952	0,01841071	18,41
a3	43,41700879	0,043417009	0,001774629					0,078083692		5,32	71,9197471	3,826130547	0,020408005	20,41
A/02/10b1	45,22605082	0,045226051	0,001848572					0,081337179		5,77	71,9197471	4,149769409	0,019600409	19,60
b2	39,79892472	0,039798925	0,001626744					0,071576717		5,94	71,9197471	4,27203298	0,01675472	16,75
b3	1,809042033	0,001809042	7,39429E-05					0,003253487		5,12	71,9197471	3,682291053	0,00088355	
A/02/10c1	45,22605082	0,045226051	0,001848572					0,081337179		5,27	71,9197471	3,790170674	0,021460031	21,46
c2	28,94467253	0,028944673	0,001183086					0,052055794		5,89	71,9197471	4,236073106	0,012288691	12,29
c3	50,65317692	0,050653177	0,002070401					0,09109764		5,57	71,9197471	4,005929915	0,022740697	22,74
A/02/02a1	56,08030302	0,056080303	0,00229223					0,100858102		5,39	71,9197471	3,87647437	0,026017998	26,02
a2	52,46221895	0,052462219	0,002144344					0,094351128		5,27	71,9197471	3,790170674	0,024893636	24,89
a3	54,27126099	0,054271261	0,002218287					0,097604615		6,06	71,9197471	4,358336676	0,022394923	22,39
A/02/02b1	52,46221895	0,052462219	0,002144344					0,094351128		5,36	71,9197471	3,854898446	0,024475645	24,48
b2	63,31647115	0,063316471	0,002588001					0,11387205		6,32	71,9197471	4,545328019	0,025052548	25,05
b3	57,88934505	0,057889345	0,002366172					0,104111589		6,36	71,9197471	4,574095918	0,022761129	22,76
A/02/02c1	14,47233626	0,014472336	0,000591543					0,026027897		5,94	71,9197471	4,27203298	0,006092626	
c2	68,74359725	0,068743597	0,00280983					0,123632512		5,71	71,9197471	4,106617561	0,030105679	30,11
c3	54,27126099	0,054271261	0,002218287					0,097604615		5,41	71,9197471	3,89085832	0,025085625	25,09
A/02/04a1	50,65317692	0,050653177	0,002070401					0,09109764		5,7	71,9197471	4,099425586	0,02222205	22,22
a2	47,03509286	0,047035093	0,001922515					0,084590666		6,01	71,9197471	4,322376803	0,019570405	19,57
a3	47,03509286	0,047035093	0,001922515					0,084590666		5	71,9197471	3,595987357	0,023523627	23,52
A/02/04b1	48,84413489	0,048844135	0,001996458					0,087844153		5,47	71,9197471	3,934010168	0,022329417	22,33
b2	50,65317692	0,050653177	0,002070401					0,09109764		6,5	71,9197471	4,674783564	0,019487028	19,49
b3	52,46221895	0,052462219	0,002144344					0,094351128		5,87	71,9197471	4,221689157	0,022349141	22,35
A/02/04c1	39,79892472	0,039798925	0,001626744					0,071576717		5,23	71,9197471	3,761402775	0,019029262	19,03
c2	41,60796676	0,041607967	0,001700686					0,074830205		5,52	71,9197471	3,969970042	0,01884906	18,85
c3	25,32658846	0,025326588	0,0010352					0,04554882		5,17	71,9197471	3,718250927	0,012250066	12,25
A/02/08a1	47,03509286	0,047035093	0,001922515					0,084590666		5,41	71,9197471	3,89085832	0,021740875	21,74
a2	37,98988269	0,037989883	0,001552801					0,06832323		5,55	71,9197471	3,991545966	0,017116984	17,12
a3	50,65317692	0,050653177	0,002070401					0,09109764		5,38	71,9197471	3,869282396	0,023543808	23,54
A/02/08b1	66,93455522	0,066934555	0,002735887					0,120379025		5,95	71,9197471	4,279224954	0,028131034	28,13
b2	50,65317692	0,050653177	0,002070401					0,09109764		6,13	71,9197471	4,406860499	0,020663244	20,66
b3	43,41700879	0,043417009	0,001774629					0,078083692		5,6	71,9197471	4,027505839	0,019387605	19,39
A/02/08c1	52,46221895	0,052462219	0,002144344					0,094351128		5,29	71,9197471	3,804554623	0,02479952	24,80
c2	52,46221895	0,052462219	0,002144344					0,094351128		5,16	71,9197471	3,711058952	0,025424314	25,42
c3	56,08030302	0,056080303	0,00229223					0,100858102		6,22	71,9197471	4,473408272	0,022546143	22,55

Calcimetry tests and results for 0/6 mm, L/S=0.3 L/kg, t=0 to 8h.

06-03				massa TQ (g)					
A/03/10a1	27,13563049	0,02713563	0,001109143	0,048802307	5,04	75,50	3,804954413	0,01282599	12,83
a2	34,37179863	0,034371799	0,001404915	0,061816256	5,7	75,50	4,303222253	0,014365109	14,37
a3	32,56275659	0,032562757	0,001330972	0,058562769	5,02	75,50	3,789855388	0,015452508	15,45
A/03/10b1	43,41700879	0,043417009	0,001774629	0,078083692	6,37	75,50	4,809039605	0,016236858	16,24
b2	41,60796676	0,041607967	0,001700686	0,074830205	5,51	75,50	4,159781511	0,017988975	17,99
b3	47,03509286	0,047035093	0,001922515	0,084590666	6,75	75,50	5,095921089	0,016599681	16,60
A/03/10c1	36,18084066	0,036180841	0,001478858	0,065069743	5,3	75,50	4,001241744	0,016262387	16,26
c2	27,13563049	0,02713563	0,001109143	0,048802307	5,7	75,50	4,30322253	0,011340875	11,34
c3	37,98988269	0,037989883	0,001552801	0,06832323	5,27	75,50	3,978593206	0,017172711	17,17
A/03/02a1	43,41700879	0,043417009	0,001774629	0,078083692	6,22	75,50	4,695796914	0,016628422	16,63
a2	47,03509286	0,047035093	0,001922515	0,084590666	6,09	75,50	4,597653249	0,018398662	18,40
a3	50,65317692	0,050653177	0,002070401	0,09109764	5,64	75,50	4,257925176	0,021394843	21,39
A/03/02b1	52,46221895	0,052462219	0,002144344	0,094351128	5,5	75,50	4,152231998	0,02272299	22,72
b2	48,84413489	0,048844135	0,001996458	0,087844153	6,23	75,50	4,703346427	0,018676947	18,68
b3	19,89946236	0,019899462	0,000813372	0,035788359	6,55	75,50	4,944930834	0,007237383	
A/03/02c1	39,79892472	0,039798925	0,001626744	0,071576717	5,89	75,50	4,446662995	0,016096726	16,10
c2	47,03509286	0,047035093	0,001922515	0,084590666	5,54	75,50	4,182430049	0,020225243	20,23
c3	48,84413489	0,048844135	0,001996458	0,087844153	5,52	75,50	4,167331024	0,021079236	21,08
A/03/04a1	52,46221895	0,052462219	0,002144344	0,094351128	6,56	75,50	4,952480347	0,019051288	19,05
a2	45,22605082	0,045226051	0,001848572	0,081337179	5,32	75,50	4,016340769	0,020251563	20,25
a3	45,22605082	0,045226051	0,001848572	0,081337179	6,36	75,50	4,801490093	0,016939987	16,94
A/03/04b1	43,41700879	0,043417009	0,001774629	0,078083692	5,32	75,50	4,016340769	0,019441501	19,44
b2	28,94467253	0,028944673	0,001183086	0,052055794	5,19	75,50	3,918197104	0,01328565	13,29
b3	39,79892472	0,039798925	0,001626744	0,071576717	6,1	75,50	4,605202762	0,015542577	15,54
A/03/04c1	56,08030302	0,056080303	0,00229223	0,100858102	7,04	75,50	5,314856958	0,018976635	18,98
c2	27,13563049	0,02713563	0,001109143	0,048802307	6,44	75,50	4,861886194	0,010037731	10,04
c3	39,79892472	0,039798925	0,001626744	0,071576717	5,28	75,50	3,986142718	0,017956386	17,96
A/03/08a1	36,18084066	0,036180841	0,001478858	0,065069743	5,19	75,50	3,918197104	0,016607062	16,61
a2	48,84413489	0,048844135	0,001996458	0,087844153	6,23	75,50	4,703346427	0,018676947	18,68
a3	61,50742912	0,061507429	0,002514058	0,110618563	6,12	75,50	4,620301787	0,023941848	23,94
A/03/08b1	48,84413489	0,048844135	0,001996458	0,087844153	5,58	75,50	4,2126281	0,020852577	20,85
b2	59,69838709	0,059698387	0,002440115	0,107365076	6,33	75,50	4,778841554	0,022466758	22,47
b3	48,84413489	0,048844135	0,001996458	0,087844153	5,86	75,50	4,424014456	0,019856208	19,86
A/03/08c1	43,41700879	0,043417009	0,001774629	0,078083692	5,84	75,50	4,408915431	0,017710408	17,71
c2	43,41700879	0,043417009	0,001774629	0,078083692	5,17	75,50	3,903098078	0,020005567	20,01
c3	37,98988269	0,037989883	0,001552801	0,06832323	5,73	75,50	4,325870791	0,015794099	15,79

Calcimetry tests and results for 6/16 mm, L/S=0.0 L/kg, t=0 to 8h.

6/16-00								massa TQ (g)			
B/00/00a1	28,94467253	0,028944673	0,001183086		0,052055794	5,19	98,63	5,118929932	0,010169273	10,17	
a2	41,60796676	0,041607967	0,001700686		0,074830205	6,14	98,63	6,05592096	0,012356536	12,36	
a3	48,84413489	0,048844135	0,001996458		0,087844153	5,37	98,63	5,296465074	0,01658543	16,59	
B/00/00b1	77,78880742	0,077788807	0,003179544		0,139899948	8,65	98,63	8,531549887	0,016397952	16,40	
b2	25,32658846	0,025326588	0,0010352		0,04554882	5,14	98,63	5,069614615	0,008984671	8,98	
b3	25,32658846	0,025326588	0,0010352		0,04554882	5,96	98,63	5,878385818	0,007748525	7,75	
B/00/00c1	65,12551318	0,065125513	0,002661944		0,117125538	6,06	98,63	5,977016453	0,019595987	19,60	
c2	5,427126099	0,005427126	0,000221829		0,009760461	5,05	98,63	4,980847044	0,001959599	1,96	
c3	50,65317692	0,050653177	0,002070401		0,09109764	6,5	98,63	6,410991244	0,014209603	14,21	
B/00/02a1	41,60796676	0,041607967	0,001700686		0,074830205	5,36	98,63	5,286602011	0,014154688	14,15	
a2	34,37179863	0,034371799	0,001404915		0,061816256	6,12	98,63	6,036194833	0,010240931	10,24	
2a3	32,56275659	0,032562757	0,001330972		0,058562769	5,79	98,63	5,710713739	0,010254895	10,25	
B/00/02b1	37,98988269	0,037989883	0,001552801		0,06832323	6,4	98,63	6,31236061	0,010823721	10,82	
b2	18,09042033	0,01809042	0,000739429		0,032534872	5,48	98,63	5,404958772	0,006019449	6,02	
b3	25,32658846	0,025326588	0,0010352		0,04554882	5,2	98,63	5,128792996	0,008881002	8,88	
B/00/02c1	34,37179863	0,034371799	0,001404915		0,061816256	6,88	98,63	6,785787656	0,009109666	9,11	
c2	25,32658846	0,025326588	0,0010352		0,04554882	5,36	98,63	5,286602011	0,008615897	8,62	
c3	23,51754643	0,023517546	0,000961258		0,042295333	5,41	98,63	5,335917328	0,007926535	7,93	
B/00/04a1	36,18084066	0,036180841	0,001478858		0,065069743	5,64	98,63	5,562767787	0,011697368	11,70	
a2	37,98988269	0,037989883	0,001552801		0,06832323	5,15	98,63	5,079477678	0,013450838	13,45	
a3	45,22605082	0,045226051	0,001848572		0,081337179	6,12	98,63	6,036194833	0,013474909	13,47	
B/00/04b1	34,37179863	0,034371799	0,001404915		0,061816256	5,34	98,63	5,266875884	0,011736798	11,74	
b2	54,27126099	0,054271261	0,002218287		0,097604615	5,55	98,63	5,474000216	0,017830583	17,83	
b3	32,56275659	0,032562757	0,001330972		0,058562769	5,3	98,63	5,22742363	0,011202989	11,20	
B/00/04c1	39,79892472	0,039798925	0,001626744		0,071576717	5,36	98,63	5,286602011	0,013539267	13,54	
c2	37,98988269	0,037989883	0,001552801		0,06832323	6,03	98,63	5,947427262	0,011487863	11,49	
c3	37,98988269	0,037989883	0,001552801		0,06832323	4,95	98,63	4,882216409	0,013994306	13,99	
B/00/08a1	52,46221895	0,052462219	0,002144344		0,094351128	6,62	98,63	6,529348006	0,014450314	14,45	
a2	36,18084066	0,036180841	0,001478858		0,065069743	5,68	98,63	5,602220041	0,011614992	11,61	
a3	34,37179863	0,034371799	0,001404915		0,061816256	5,42	98,63	5,345780392	0,011563561	11,56	
B/00/08b1	32,56275659	0,032562757	0,001330972		0,058562769	5,27	98,63	5,19783444	0,011266763	11,27	
b2	45,22605082	0,045226051	0,001848572		0,081337179	5,34	98,63	5,266875884	0,011544315	15,44	
b3	47,03509286	0,047035093	0,001922515		0,084590666	5,76	98,63	5,681124549	0,014889775	14,89	
B/00/08c1	54,27126099	0,054271261	0,002218287		0,097604615	5,85	98,63	5,76989212	0,016916194	16,92	
c2	41,60796676	0,041607967	0,001700686		0,074830205	5,56	98,63	5,48386328	0,013645527	13,65	
c3	41,60796676	0,041607967	0,001700686		0,074830205	5,32	98,63	5,247149757	0,014261115	14,26	

Calcimetry tests and results for 6/16 mm, L/S=0.2 L/kg, t=0 to 8h.

6/16-02								massa TQ (g)							
B/02/00a1	45,22605082	0,045226051	0,001848572				0,081337179	5,26	91,10	4,792010069	0,016973499	16,97			
a2	19,89946236	0,019899462	0,000813372				0,035788359	5,63	91,10	5,129090625	0,006977525	6,98			
a3	32,56275659	0,032562757	0,001330972				0,058562769	5,5	91,10	5,010656916	0,011687643	11,69			
B/02/00b1	19,89946236	0,019899462	0,000813372				0,035788359	5,43	91,10	4,946884919	0,007234524	7,23			
b2	19,89946236	0,019899462	0,000813372				0,035788359	5,51	91,10	5,019767201	0,007129486	7,13			
b3	21,70850439	0,021708504	0,000887315				0,039041846	5,77	91,10	5,256634619	0,007427156	7,43			
B/02/00c1	5,427126099	0,005427126	0,000221829				0,009760461	5,29	91,10	4,819340924	0,002025269	2,03			
c2	27,13563049	0,02713563	0,001109143				0,048802307	5,52	91,10	5,028877486	0,009704414	9,70			
c3	16,2813783	0,016281378	0,000665486				0,029281384	6,64	91,10	6,04922944	0,004840515	4,84			
B/02/02a1	25,32658846	0,025326588	0,0010352				0,04554882	5,32	91,10	4,84667178	0,009397958	9,40			
a2	18,09042033	0,01809042	0,000739429				0,032534872	5,16	91,10	4,700907216	0,006920977	6,92			
a3	28,94467253	0,028944673	0,001183086				0,052055794	5	91,10	4,555142651	0,011427918	11,43			
B/02/02b1	23,51754643	0,023517546	0,000961258				0,042295333	5,12	91,10	4,664466074	0,009067562	9,07			
b2	27,13563049	0,02713563	0,001109143				0,048802307	6,14	91,10	5,593715175	0,008724489	8,72			
b3	25,32658846	0,025326588	0,0010352				0,04554882	6,11	91,10	5,566384319	0,008182838	8,18			
B/02/02c1	21,70850439	0,021708504	0,000887315				0,039041846	6,31	91,10	5,748590025	0,006791552	6,79			
c2	28,94467253	0,028944673	0,001183086				0,052055794	5,74	91,10	5,229303763	0,009954632	9,95			
c3	32,56275659	0,032562757	0,001330972				0,058562769	6,05	91,10	5,511722607	0,01062513	10,63			
B/02/04a1	23,51754643	0,023517546	0,000961258				0,042295333	5,1	91,10	4,646245504	0,009103121	9,10			
a2	19,89946236	0,019899462	0,000813372				0,035788359	6,47	91,10	5,89435459	0,006071633	6,07			
a3	27,13563049	0,02713563	0,001109143				0,048802307	5,27	91,10	4,801120354	0,010164775	10,16			
B/02/04b1	41,60796676	0,041607967	0,001700686				0,074830205	5,37	91,10	4,892223207	0,015295746	15,30			
b2	43,41700879	0,043417009	0,001774629				0,078083692	6,92	91,10	6,304317429	0,012385749	12,39			
b3	16,2813783	0,016281378	0,000665486				0,029281384	5,43	91,10	4,946884919	0,005919156	5,92			
B/02/04c1	41,60796676	0,041607967	0,001700686				0,074830205	5,51	91,10	5,019767201	0,014907107	14,91			
c2	23,51754643	0,023517546	0,000961258				0,042295333	5,21	91,10	4,746458642	0,008910924	8,91			
c3	54,27126099	0,054271261	0,002218287				0,097604615	5,26	91,10	4,792010069	0,020368199	20,37			
B/02/08a1	27,13563049	0,02713563	0,001109143				0,048802307	5,5	91,10	5,010656916	0,009739702	9,74			
a2	39,79892472	0,039798925	0,001626744				0,071576717	5,75	91,10	5,238414048	0,013663814	13,66			
a3	34,37179863	0,034371799	0,001404915				0,061816256	5,87	91,10	5,347737472	0,011559329	11,56			
B/02/08b1	36,18084066	0,036180841	0,001478858				0,065069743	5,75	91,10	5,238414048	0,012421649	12,42			
b2	25,32658846	0,025326588	0,0010352				0,04554882	5,37	91,10	4,892223207	0,009310454	9,31			
b3	32,56275659	0,032562757	0,001330972				0,058562769	5,94	91,10	5,411509469	0,010821892	10,82			
B/02/08c1	25,32658846	0,025326588	0,0010352				0,04554882	6,09	91,10	5,548163749	0,008209711	8,21			
c2	32,56275659	0,032562757	0,001330972				0,058562769	5,95	91,10	5,420619754	0,010803704	10,80			
c3	32,56275659	0,032562757	0,001330972				0,058562769	5,34	91,10	4,864892351	0,012037834	12,04			

Calcimetry tests and results for 6/16 mm, L/S=0.3 L/kg, t=0 to 8h.

6/16-03												massa TQ (g)					
B/03/00a1	47,03509286	0,047035093	0,001922515					0,084590666	6,04	93,97	5,675708377	0,014903984	14,90				
a2	43,41700879	0,043417009	0,001774629					0,078083692	5,38	93,97	5,055515077	0,01544525	15,45				
a3	9,045210165	0,00904521	0,000369714					0,016267436	5,81	93,97	5,459580409	0,002979613	2,98				
B/03/00b1	52,46221895	0,052462219	0,002144344					0,094351128	5,01	93,97	4,707830955	0,020041316	20,04				
b2	37,98988269	0,037989883	0,001552801					0,06832323	5,35	93,97	5,027324473	0,013590376	13,59				
b3	47,03509286	0,047035093	0,001922515					0,084590666	6,2	93,97	5,826058268	0,014519365	14,52				
B/03/00c1	39,79892472	0,039798925	0,001626744					0,071576717	6,4	93,97	6,013995631	0,011901691	11,90				
c2	34,37179863	0,034371799	0,001404915					0,061816256	5,68	93,97	5,337421123	0,011581671	11,58				
c3	28,94467253	0,028944673	0,001183086					0,052055794	5,19	93,97	4,876974582	0,010673788	10,67				
B/03/02a1	23,51754643	0,023517546	0,000961258					0,042295333	5,39	93,97	5,064911946	0,008350655	8,35				
a2	32,56275659	0,032562757	0,001330972					0,058562769	5,88	93,97	5,525358486	0,010598908	10,60				
a3	25,32658846	0,025326588	0,0010352					0,04554882	5,37	93,97	5,046118209	0,009026507	9,03				
B/03/02b1	19,89946236	0,019899462	0,000813372					0,035788359	5,55	93,97	5,215261836	0,006862236	6,86				
b2	65,12551318	0,065125513	0,002661944					0,117125538	5,29	93,97	4,970943264	0,023562035					
b3	19,89946236	0,019899462	0,000813372					0,035788359	5,21	93,97	4,895768318	0,00731006	7,31				
B/03/02c1	28,94467253	0,028944673	0,001183086					0,052055794	5,53	93,97	5,1964681	0,010017534	10,02				
c2	19,89946236	0,019899462	0,000813372					0,035788359	5,28	93,97	4,961546396	0,007213146	7,21				
c3	28,94467253	0,028944673	0,001183086					0,052055794	5,65	93,97	5,309230518	0,009804772	9,80				
B/03/04a1	25,32658846	0,025326588	0,0010352					0,04554882	5,77	93,97	5,421992936	0,008400752	8,40				
a2	10,8542522	0,010854252	0,000443657					0,019520923	5,26	93,97	4,942752659	0,003949403	3,95				
a3	12,66329423	0,012663294	0,0005176					0,02277441	5,38	93,97	5,055515077	0,004504864	4,50				
B/03/04b1	19,89946236	0,019899462	0,000813372					0,035788359	5,16	93,97	4,848783978	0,007380894	7,38				
b2	25,32658846	0,025326588	0,0010352					0,04554882	5,1	93,97	4,792402768	0,009504381	9,50				
b3	32,56275659	0,032562757	0,001330972					0,058562769	5,9	93,97	5,544152222	0,01056298	10,56				
B/03/04c1	21,70850439	0,021708504	0,000887315					0,039041846	5,09	93,97	4,7830059	0,008162617	8,16				
c2	36,18084066	0,036180841	0,001478858					0,065069743	5,6	93,97	5,262246177	0,012365393	12,37				
c3	21,70850439	0,021708504	0,000887315					0,039041846	5,51	93,97	5,177674364	0,007540421	7,54				
B/03/08a1	133,8691104	0,13386911	0,005471774					0,24075805	6,47	93,97	6,079773708	0,039599837					
a2	47,03509286	0,047035093	0,001922515					0,084590666	5,94	93,97	5,581739695	0,015154893	15,15				
a3	32,56275659	0,032562757	0,001330972					0,058562769	5,36	93,97	5,036721341	0,011627161	11,63				
B/03/08b1	25,32658846	0,025326588	0,0010352					0,04554882	5,59	93,97	5,252849309	0,00867126	8,67				
b2	18,09042033	0,01809042	0,000739429					0,032534872	5,43	93,97	5,102499418	0,006376262	6,38				
b3	32,56275659	0,032562757	0,001330972					0,058562769	5,79	93,97	5,440786672	0,010763658	10,76				
B/03/08c1	23,51754643	0,023517546	0,000961258					0,042295333	5,64	93,97	5,29983365	0,007980502	7,98				
c2	30,75371456	0,030753715	0,001257029					0,055309282	5,45	93,97	5,121293155	0,010799866	10,80				
c3	27,13563049	0,02713563	0,001109143					0,048802307	5,36	93,97	5,036721341	0,009689301	9,69				

Calcimetry tests and results for 16/31 mm, L/S=0.0 L/kg, t=0 to 8h.

16/31-00												massa TQ (g)							
C/00/00a1	66,93455522	0,066934555	0,002735887					0,120379025	5,36	97,97	5,25108238	0,022924612							
a2	10,8542522	0,010854252	0,000443657					0,019520923	5,29	97,97	5,182504811	0,003766697	3,77						
a3	3,618084066	0,003618084	0,000147886					0,006506974	5,75	97,97	5,633157404	0,00115512	1,16						
C/00/00b1	9,045210165	0,00904521	0,000369714					0,016267436	5,61	97,97	5,496002267	0,002959867	2,96						
b2	12,66329423	0,012663294	0,0005176					0,02277441	5,09	97,97	4,986568902	0,00456715	4,57						
b3	3,618084066	0,003618084	0,000147886					0,006506974	5,36	97,97	5,25108238	0,001239168	1,24						
C/00/00c1	3,618084066	0,003618084	0,000147886					0,006506974	5,5	97,97	5,388237516	0,001207626	1,21						
c2	16,2813783	0,016281378	0,000665486					0,029281384	6,32	97,97	6,191574746	0,004729231	4,73						
c3	5,427126099	0,005427126	0,000221829					0,009760461	5,3	97,97	5,192301607	0,001879795	1,88						
C/00/02a1	21,70850439	0,021708504	0,000887315					0,039041846	5,85	97,97	5,731125358	0,006812248	6,81						
2a2	21,70850439	0,021708504	0,000887315					0,039041846	5,37	97,97	5,260879175	0,007421164	7,42						
2a3	28,94467253	0,028944673	0,001183086					0,052055794	6,3	97,97	6,17981155	0,008434212	8,43						
C/00/02b1	16,2813783	0,016281378	0,000665486					0,029281384	6,36	97,97	6,230761928	0,004699487	4,70						
b2	10,8542522	0,010854252	0,000443657					0,019520923	5,37	97,97	5,260879175	0,003710582	3,71						
b3	3,618084066	0,003618084	0,000147886					0,006506974	5,47	97,97	5,35884713	0,001214249	1,21						
C/00/02c1	19,89946236	0,019899462	0,000813372					0,035788359	5,4	97,97	5,290269562	0,00676494	6,76						
c2	1,809042033	0,001809042	7,39429E-05					0,003253487	5,64	97,97	5,525392653	0,000588825	0,59						
c3	34,37179863	0,034371799	0,001404915					0,061816256	5,77	97,97	5,652750995	0,010935606	10,94						
C/00/04a1	9,045210165	0,00904521	0,000369714					0,016267436	5,38	97,97	5,270675971	0,003086404	3,09						
a2	18,09042033	0,01809042	0,000739429					0,032534872	5,89	97,97	5,77031254	0,005638321	5,64						
a3	7,236168132	0,007236168	0,000295772					0,013013949	5,07	97,97	4,966975311	0,002620095	2,62						
C/00/04b1	3,618084066	0,003618084	0,000147886					0,006506974	6,06	97,97	5,936858064	0,00109603	1,10						
b2	19,89946236	0,019899462	0,000813372					0,035788359	6,1	97,97	5,976045246	0,005988636	5,99						
b3	14,47233626	0,014472336	0,000591543					0,026027897	5,43	97,97	5,319659948	0,004892775	4,89						
C/00/04c1	10,8542522	0,010854252	0,000443657					0,019520923	5,21	97,97	5,104130447	0,003824534	3,82						
c2	14,47233626	0,014472336	0,000591543					0,026027897	5,76	97,97	5,642954199	0,004612459	4,61						
c3	10,8542522	0,010854252	0,000443657					0,019520923	5,36	97,97	5,25108238	0,003717505	3,72						
C/00/08a1	37,98988269	0,037989883	0,001552801					0,06832323	5,95	97,97	5,829093313	0,011721073	11,72						
a2	32,56275659	0,032562757	0,001330972					0,058562769	5,16	97,97	5,05514647	0,011584782	11,58						
a3	21,70850439	0,021708504	0,000887315					0,039041846	5,01	97,97	4,908194538	0,007954421	7,95						
C/00/08b1	28,94467253	0,028944673	0,001183086					0,052055794	5,15	97,97	5,045349675	0,010317579	10,32						
b2	32,56275659	0,032562757	0,001330972					0,058562769	5,21	97,97	5,104130447	0,011473603	11,47						
b3	27,13563049	0,02713563	0,001109143					0,048802307	5,22	97,97	5,113927243	0,009543019	9,54						
C/00/08c1	37,98988269	0,037989883	0,001552801					0,06832323	5,2	97,97	5,094333652	0,013411613	13,41						
c2	47,03509286	0,047035093	0,001922515					0,084590666	5,24	97,97	5,133520834	0,016478099	16,48						
c3	45,22605082	0,045226051	0,001848572					0,081337179	5,45	97,97	5,339253539	0,015233811	15,23						

Calcimetry tests and results for 16/31 mm, L/S=0.2 L/kg, t=0 to 8h.

16/31-02								massa TQ (g)							
C/02/0a1	65,12551318	0,065125513	0,002661944				0,117125538	5,29	92,09	4,8718168	0,02404145				
a2	34,37179863	0,034371799	0,001404915				0,061816256	6,44	92,09	5,930907408	0,010422732	10,42			
a3	0	0	0				0	5,1	92,09	4,696836612	0	0,00			
C/02/0b1	9,045210165	0,00904521	0,000369714				0,016267436	5,92	92,09	5,452014264	0,002983748	2,98			
b2	12,66329423	0,012663294	0,0005176				0,02277441	5,51	92,09	5,074425438	0,004488077	4,49			
b3	16,2813783	0,016281378	0,000665486				0,029281384	5,57	92,09	5,129682339	0,005708226	5,71			
C/02/0c1	12,66329423	0,012663294	0,0005176				0,02277441	5,29	92,09	4,8718168	0,004674726	4,67			
c2	7,236168132	0,007236168	0,000295772				0,013013949	6,2	92,09	5,709879803	0,002279198	2,28			
c3	10,8542522	0,010854252	0,000443657				0,019520923	6,01	92,09	5,534899616	0,003526879	3,53			
C/02/02a1	19,89946236	0,019899462	0,000813372				0,035788359	5,32	92,09	4,89944525	0,007304574	7,30			
a2	48,84413489	0,048844135	0,001996458				0,087844153	5,57	92,09	5,129682339	0,017124677	17,12			
a3	32,56275659	0,032562757	0,001330972				0,058562769	5,3	92,09	4,881026283	0,011998044	12,00			
C/02/02b1	25,32658846	0,025326588	0,0010352				0,04554882	5,78	92,09	5,323081494	0,008556852	8,56			
b2	43,41700879	0,043417009	0,001774629				0,078083692	5,54	92,09	5,102053889	0,015304364	15,30			
b3	19,89946236	0,019899462	0,000813372				0,035788359	5,84	92,09	5,378338395	0,006654166	6,65			
C/02/02c1	41,60796676	0,041607967	0,001700686				0,074830205	5,46	92,09	5,02837802	0,014881579	14,88			
c2	19,89946236	0,019899462	0,000813372				0,035788359	5,71	92,09	5,258615109	0,006805662	6,81			
c3	14,47233626	0,014472336	0,000591543				0,026027897	5,38	92,09	4,954702152	0,005253171	5,25			
C/02/04a1	36,18084066	0,036180841	0,001478858				0,065069743	5,6	92,09	5,15731079	0,012616991				
a2	9,045210165	0,00904521	0,000369714				0,016267436	5,38	92,09	4,954702152	0,003283232	3,28			
a3	10,8542522	0,010854252	0,000443657				0,019520923	5,05	92,09	4,650789194	0,004197336	4,20			
C/02/04b1	10,8542522	0,010854252	0,000443657				0,019520923	5,44	92,09	5,009959053	0,003896424	3,90			
b2	14,47233626	0,014472336	0,000591543				0,026027897	5,45	92,09	5,019168537	0,005185699	5,19			
b3	14,47233626	0,014472336	0,000591543				0,026027897	5,53	92,09	5,092844405	0,00511068	5,11			
C/02/04c1	12,66329423	0,012663294	0,0005176				0,02277441	5,9	92,09	5,433595296	0,004191407	4,19			
c2	9,045210165	0,00904521	0,000369714				0,016267436	5,16	92,09	4,752093514	0,003423215	3,42			
c3	10,8542522	0,010854252	0,000443657				0,019520923	5,6	92,09	5,15731079	0,003785097	3,79			
C/02/08a1	12,66329423	0,012663294	0,0005176				0,02277441	6,22	92,09	5,72829877	0,003975772	3,98			
a2	27,13563049	0,02713563	0,001109143				0,048802307	6,29	92,09	5,792765155	0,0084247	8,42			
a3	7,236168132	0,007236168	0,000295772				0,013013949	5,87	92,09	5,405966846	0,00240733	2,41			
C/02/08b1	19,89946236	0,019899462	0,000813372				0,035788359	5,63	92,09	5,184939241	0,006902368	6,90			
b2	25,32658846	0,025326588	0,0010352				0,04554882	5,37	92,09	4,945492668	0,009210168	9,21			
b3	21,70850439	0,021708504	0,000887315				0,039041846	5,69	92,09	5,240196142	0,007450455	7,45			
C/02/08c1	10,8542522	0,010854252	0,000443657				0,019520923	6,29	92,09	5,792765155	0,00336988	3,37			
c2	19,89946236	0,019899462	0,000813372				0,035788359	5,33	92,09	4,908654734	0,007290869	7,29			
c3	12,66329423	0,012663294	0,0005176				0,02277441	5,07	92,09	4,669208162	0,004877574	4,88			

Calcimetry tests and results for 16/31 mm, L/S=0.3 L/kg, t=0 to 8h.

16/31-03								massa TQ (g)				
C/03/0a1	16,2813783	0,016281378	0,000665486				0,029281384	5,49	91,20	5,006824104	0,005848295	5,85
a2	14,47233626	0,014472336	0,000591543				0,026027897	5,3	91,20	4,833546038	0,005384845	5,38
a3	18,09042033	0,01809042	0,000739429				0,032534872	5,78	91,20	5,271301151	0,006172076	6,17
C/03/0b1	14,47233626	0,014472336	0,000591543				0,026027897	5,27	91,20	4,806186344	0,005415499	5,42
b2	14,47233626	0,014472336	0,000591543				0,026027897	5,19	91,20	4,733227158	0,005498975	5,50
b3	19,89946236	0,019899462	0,000813372				0,035788359	5,26	91,20	4,797066445	0,007460468	7,46
C/03/0c1	10,8542522	0,010854252	0,000443657				0,019520923	5,28	91,20	4,815306242	0,004053932	4,05
c2	14,47233626	0,014472336	0,000591543				0,026027897	5,13	91,20	4,678507769	0,00556329	5,56
c3	9,045210165	0,00904521	0,000369714				0,016267436	5,33	91,20	4,860905733	0,003346585	3,35
C/03/02a1	68,74359725	0,068743597	0,00280983				0,123632512	5,6	91,20	5,107142984	0,024207764	24,21
a2	79,59784945	0,079597849	0,003253487				0,143153435	5,56	91,20	5,070663391	0,028231697	28,23
a3	65,12551318	0,065125513	0,002661944				0,117125538	5,4	91,20	4,92474502	0,023783066	23,78
C/03/02b1	50,65317692	0,050653177	0,002070401				0,09109764	5,32	91,20	4,851785835	0,018776105	18,78
b2	50,65317692	0,050653177	0,002070401				0,09109764	5,36	91,20	4,888265427	0,018635985	18,64
b3	61,50742912	0,061507429	0,002514058				0,110618563	5,41	91,20	4,933864918	0,022420266	22,42
C/03/02c1	86,83401758	0,086834018	0,003549259				0,156167383	5,01	91,20	4,569068991	0,034179257	34,18
c2	45,22605082	0,045226051	0,001848572				0,081337179	5,25	91,20	4,787946547	0,016987905	16,99
c3	32,56275659	0,032562757	0,001330972				0,058562769	5,01	91,20	4,569068991	0,012817221	12,82
C/03/04a1	12,66329423	0,012663294	0,0005176				0,02277441	5,02	91,20	4,578188889	0,004974546	4,97
a2	37,98988269	0,037989883	0,001552801				0,06832323	5,02	91,20	4,578188889	0,014923637	14,92
a3	21,70850439	0,021708504	0,000887315				0,039041846	5,35	91,20	4,879145529	0,008001779	8,59
C/03/04b1	28,94467253	0,028944673	0,001183086				0,052055794	5,95	91,20	5,42633942	0,00959317	9,59
b2	47,03509286	0,047035093	0,001922515				0,084590666	5,89	91,20	5,371620031	0,015747701	15,75
b3	54,27216099	0,054271261	0,002218287				0,097604615	6,75	91,20	6,155931275	0,015855378	15,86
C/03/04c1	19,89946236	0,019899462	0,000813372				0,035788359	5,11	91,20	4,660267973	0,007679464	7,68
c2	19,89946236	0,019899462	0,000813372				0,035788359	7,05	91,20	6,429528221	0,00556625	5,57
c3	25,32658846	0,025326588	0,0010352				0,04554882	5,51	91,20	5,0250639	0,009064327	9,06
C/03/08a1	12,66329423	0,012663294	0,0005176				0,02277441	5,04	91,20	4,596428685	0,004954805	4,95
a2	9,045210165	0,00904521	0,000369714				0,016267436	5,26	91,20	4,797066445	0,003391122	3,39
a3	9,045210165	0,00904521	0,000369714				0,016267436	5,16	91,20	4,705867464	0,003456841	3,46
C/03/08b1	10,8542522	0,010854252	0,000443657				0,019520923	5,45	91,20	4,970344511	0,003927479	3,93
b2	12,66329423	0,012663294	0,0005176				0,02277441	5,67	91,20	5,170982271	0,004404272	4,40
b3	12,66329423	0,012663294	0,0005176				0,02277441	5,21	91,20	4,751466955	0,004793132	4,79
C/03/08c1	9,045210165	0,00904521	0,000369714				0,016267436	5,82	91,20	5,307780744	0,003064828	3,06
c2	7,236168132	0,007236168	0,000295772				0,013013949	5,56	91,20	5,070663391	0,002566518	2,57
c3	9,045210165	0,00904521	0,000369714				0,016267436	5,18	91,20	4,72410726	0,003443494	3,44

Calcimetry tests and results for 0/6 mm, L/S=0.0 L/kg, t=8h. with different sample masses: 200, 100, 50, 25g.

A-50g-8h	90.45210165	0.090452102	0.003697144						0.162674358	5.33	86.08	4.587926026	0.035457058	35.46
	101.3063538	0.101306354	0.004140802						0.182195281	5.15	86.08	4.432986685	0.041099894	41.10
	99.49731181	0.099497312	0.004066859						0.178941794	5.47	86.08	4.708434402	0.038004521	38.00
	103.1153959	0.103115396	0.004214745						0.185448768	5.13	86.08	4.415771203	0.041996915	42.00
	139.2962365	0.139296237	0.005693603						0.250518511	5.39	86.08	4.639572473	0.053996034	54.00
	108.542522	0.108542522	0.004436573						0.195209229	5.16	86.08	4.441594427	0.04395026	43.95
	95.87922774	0.095879228	0.003918973						0.172434819	5.13	86.08	4.415771203	0.039049763	39.05
	92.26114368	0.092261144	0.003771087						0.165927845	5.01	86.08	4.312478309	0.038476216	38.48
	85.02497555	0.085024976	0.003475316						0.152913896	5.53	86.08	4.760080849	0.032124223	32.12
	0													
A-100g-8h	90.45210165	0.090452102	0.003697144						0.162674358	5.17	86.08	4.450202168	0.036554375	36.55
	95.87922774	0.095879228	0.003918973						0.172434819	5.99	86.08	5.156036941	0.033443286	33.44
	92.26114368	0.092261144	0.003771087						0.165927845	6.05	86.08	5.207683388	0.031862122	31.86
	97.68826978	0.09768827	0.003992916						0.175688306	5.45	86.08	4.691218919	0.03745046	37.45
	88.64305961	0.08864306	0.003623202						0.159420871	5.33	86.08	4.587926026	0.034747917	34.75
	97.68826978	0.09768827	0.003992916						0.175688306	5.19	86.08	4.46741765	0.039326591	39.33
	99.49731181	0.099497312	0.004066859						0.178941794	5.1	86.08	4.38994798	0.040761712	40.76
	86.83401758	0.086834018	0.003549259						0.156167383	5.48	86.08	4.717042143	0.033107057	33.11
	79.59784945	0.079597849	0.003253487						0.143153435	5.34	86.08	4.596533767	0.03114378	31.14
	0													
A-25g-8h	110.351564	0.110351564	0.004510516						0.198462716	5	86.08	4.303870568	0.046112613	46.11
	99.49731181	0.099497312	0.004066859						0.178941794	5.27	86.08	4.536279579	0.039446818	39.45
	103.1153959	0.103115396	0.004214745						0.185448768	5.18	86.08	4.458809909	0.041591539	41.59
	104.9244379	0.104924438	0.004286688						0.188702255	5.33	86.08	4.587926026	0.041130187	41.13
	112.160606	0.112160606	0.004584459						0.20716204	5.1	86.08	4.38994798	0.045949566	45.95
	110.351564	0.110351564	0.004510516						0.198462716	5.17	86.08	4.450202168	0.044596337	44.60
	101.3063538	0.101306354	0.004140802						0.182195281	5.44	86.08	4.682611178	0.038908907	38.91
	103.1153959	0.103115396	0.004214745						0.185448768	5.64	86.08	4.854766001	0.038199322	38.20
	94.07018571	0.094070186	0.00384503						0.169181332	5.32	86.08	4.579318285	0.036944655	36.94
	0													

ANNEX IX: Moisture content of the sample

	Moisture content calculations (campioni in stufa)					TS %
	Tare muffin heater (g)	Gross mass inlet heater (g)	Net mass inlet heater (g)	Gross mass outlet heater (g)	Net mass outlet heater (g)	
0/6-L/S0-t0	2,19		10,82	11,50	9,32	86,08
0/6-L/S0-t0A	2,16	18,05	15,89	15,62	13,46	84,73
0/6-L/S0-t0B	2,18	20,82	18,64	18,13	15,95	85,54
0/6-L/S0-t0C	2,19	18,49	16,30	16,16	13,96	85,68
0/6-L/S0-t8h	2,20	14,57	12,37	12,60	10,40	84,11
0/6-L/S0.2-t0	2,18	17,94	15,76	13,51	11,33	71,92
0/6-L/S0.2-t0A	2,19	24,55	22,35	18,59	16,40	73,35
0/6-L/S0.2-t0B	2,18	24,44	22,27	18,87	16,69	74,97
0/6-L/S0.2-t0C	2,17	30,37	28,20	22,61	20,43	72,46
0/6-L/S0.2-t8h	2,18	17,17	14,99	13,69	11,52	76,81
0/6-L/S0.3-t0	2,18	22,87	20,70	17,80	15,63	75,50
0/6-L/S0.3-t0A	2,20	29,42	27,21	21,72	19,52	71,71
0/6-L/S0.3-t0B	2,19	34,51	32,32	25,89	23,70	73,34
0/6-L/S0.3-t0C	2,21	28,35	26,14	21,28	19,07	72,96
0/6-L/S0.3-t8h	2,18	24,10	21,92	18,19	16,02	73,07
6/16-L/S0-t0	2,18		15,69	17,66	15,48	98,63
6/16-L/S0-t0A	2,19	32,70	30,51	31,93	29,74	97,49
6/16-L/S0-t0B	2,20	31,58	29,37	30,94	28,73	97,82
6/16-L/S0-t0C	2,18	34,66	32,48	33,85	31,66	97,48
6/16-L/S0.2-t0	2,18	29,04	26,86	26,65	24,47	91,10
6/16-L/S0.2-t0A	2,21	28,82	26,61	27,04	24,82	93,28
6/16-L/S0.2-t0B	2,21	25,49	23,28	23,96	21,75	93,44
6/16-L/S0.2-t0C	2,21	26,17	23,96	24,69	22,48	93,83
6/16-L/S0.3-t0	2,20	28,84	26,64	27,23	25,04	93,97
6/16-L/S0.3-t0A	2,20	29,26	27,07	26,84	24,64	91,04
6/16-L/S0.3-t0B	2,18	40,44	38,26	37,43	35,25	92,13
6/16-L/S0.3-t0C	2,20	30,67	28,47	28,73	26,53	93,20
16/31-L/S0-t0	2,17		30,97	32,51	30,34	97,97
16/31-L/S0-t0A	2,18	38,51	36,33	36,72	34,54	95,06
16/31-L/S0-t0B	2,20	44,64	42,45	42,09	39,89	93,99
16/31-L/S0-t0C	2,20	42,25	40,06	40,99	38,79	96,83
			0,00			
16/31-L/S0.2-t0	2,20	30,94	28,74	28,67	26,47	92,09
16/31-L/S0.2-t0A	2,18	39,31	37,13	37,59	35,41	95,36
16/31-L/S0.2-t0B	2,20	38,88	36,68	36,52	34,32	93,56
16/31-L/S0.2-t0C	2,20	41,53	39,33	39,51	37,31	94,87
16/31-L/S0.3-t0	2,17	30,38	28,21	27,89	25,73	91,20
16/31-L/S0.3-t0A	2,2073	36,2141	34,01	34,12	31,91	93,83
16/31-L/S0.3-t0B	2,199	47,4406	45,24	45,38	43,18	95,44
16/31-L/S0.3-t0C	2,20	44,23	42,03	42,04	39,83	94,77

The following table collects the mean values of the conducted tests when samples haven't already carbonated:

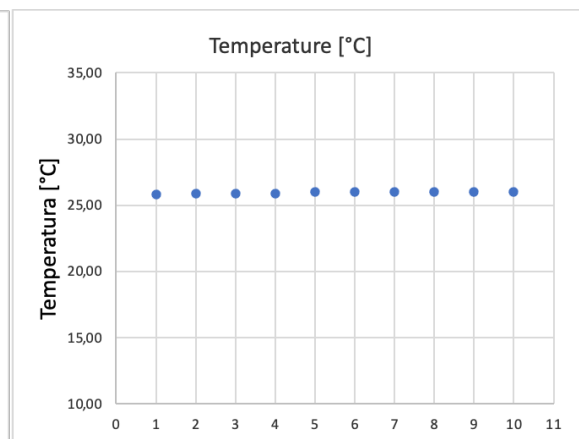
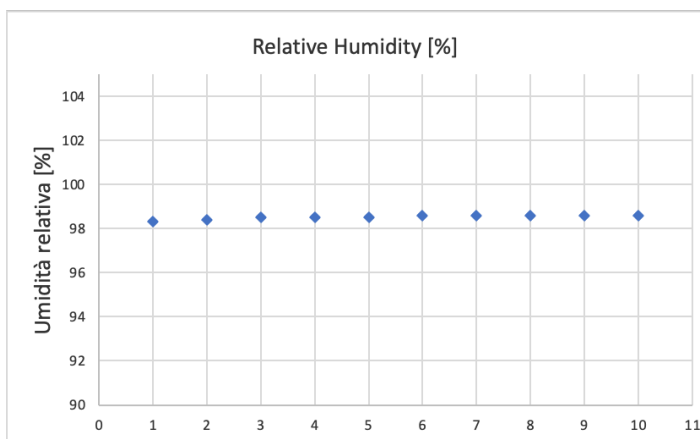
Sample	TS %
0/6-L/S0-t0	85,51
0/6-L/S0.2-t0	73,17
0/6-L/S0.3-t0	72,67
6/16-L/S0-t0	97,86
6/16-L/S0.2-t0	92,91
6/16-L/S0.3-t0	92,58
16/31-L/S0-t0	95,96
16/31-L/S0.2-t0	93,97
16/31-L/S0.3-t0	93,81

ANNEX X: Temperature, pressure and relative humidity of the CO₂ incubator

Temperature, pressure and relative humidity have been measured 10 times every 6 min, for a total test duration of 1h. It can be seen that all the parameters keep constant values.

Date	Relative Humidity [%RH]	Temp[°C]	Pressione [mbar]
22/09/22	98,3	25,8	1022,7
22/09/22	98,4	25,9	1022,8
22/09/22	98,5	25,9	1022,8
22/09/22	98,5	25,9	1022,7
22/09/22	98,5	26	1022,6
22/09/22	98,6	26	1022,7
22/09/22	98,6	26	1022,6
22/09/22	98,6	26	1022,5
22/09/22	98,6	26	1022,5
22/09/22	98,6	26	1022,4

MEAN		
Relative Humidity [%RH]	Temp[°C]	Pressione [mbar]
98,52	26,00	1022,70



ANNEX XI: Hardness test of leachate water

Leachate “hardness” tests have been carried out according to APAT CNR IRSA 2040 Met.B Man 29 2003 on the liquid filtered by paper filter to assess the quantity of carbonates that have been washed when demineralized water was added and then removed from the samples.

	mass (g)	H ₂ O (mL)	Leachate (mL)	Leachate/H ₂ O (%)
0/6-02	627,83	125,57	20	15,93
0/6-03	610,48	183,14	50	27,30
6/16-02	638,19	127,64	70	54,84
6/16-03	553,22	165,966	130	78,33
16/31-02	617,13	123,43	90	72,92
16/31-03	560,34	168,102	155	92,21

Diluted samples 1-10	Num. drops fenolftaleina	Num. drops Bromocresolo	tot drops	x20	mgCaCO ₃ /L
0/6-02	1	5	6	120	1200
0/6-03	4	6	10	200	2000
6/16-02	0	4	4	80	800
6/16-03	2	14	16	320	3200
16/31-02	4	3	7	140	1400
16/31-03	2	4	6	120	1200

Sample	mgCaCO ₃ /L
0/6-02	1200
0/6-03	2000
6/16-02	800
6/16-03	3200
16/31-02	1400
16/31-03	1200

ANNEX XII: Leachate tests analyses

UNI EN 12457-2 2004 leaching test was performed on 0/6mm fraction sample, at t=0h on the untreated material and after 8h carbonation tests as reported in Part II, paragraphs 2.2.2; 2.2.4; 2.3.3 of the main text. Tests were carried out in triplicate.

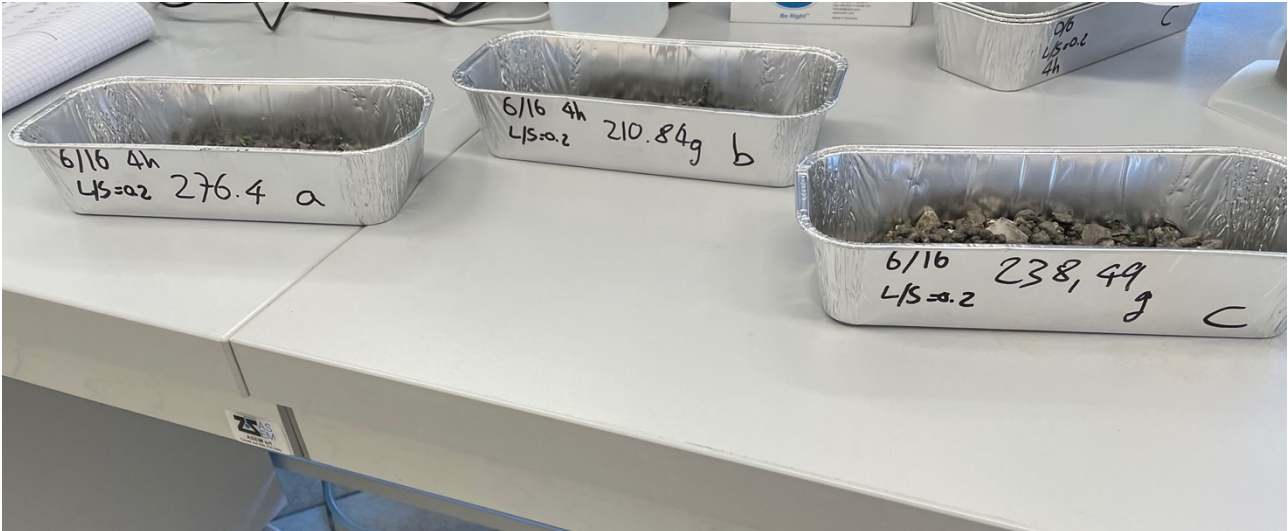
Sample	pH	Conductivity [mS/cm]
t0/00/a	11,82	3,55
t0/00/b	11,96	3,58
t0/00/c	11,90	3,76
t8h/00/a	9,26	3,76
t8h/00/b	9,45	3,68
t8h/00/c	9,58	3,72
t8h/02/a	11,32	2,90
t8h/02/b	11,25	3,30
t8h/02/c	11,28	3,22
t8h/03/a	11,53	2,55
t8h/03/b	11,5	2,66
t8h/03/c	11,56	2,58

UNI EN 12457-2		0/6-00-t0		0/6-00-t8		0/6-02-t8		0/6-03-t8	
Chemical	Unit	M	σ	M	σ	M	σ	M	σ
pH	/	11,89	0,07	9,43	0,16	11,28	0,04	11,53	0,03
Barium	mg/l Ba	0,09	0,01	0,05	0,00	0,04	0,00	0,06	0,01
Cadmium	µg/l Cd	<1	0,00	<1	0,00	<1	0,00	<1	0,00
Chromium	µg/l Cr	54,00	15,59	203,00	9,64	115,33	4,73	34,67	6,66
Molybdenum	mg/l Mo	0,03	0,03	0,11	0,00	0,03	0,01	<0.013	0,01
Nickel	µg/l Ni	14,33	0,58	8,43	0,38	12,00	1,00	9,40	0,60
Lead	µg/l Pb	21,67	7,09	<5	0,00	<5	>0	11,37	7,70
Copper	mg/l Cu	0,89	0,03	0,77	0,02	0,82	0,07	0,63	0,03
Antimony	mg/l Sb	0,03	0,01	0,08	0,01	0,06	0,01	0,04	0,00
Zinc	mg/l Zn	0,11	0,08	0,00	0,00	0,02	0,00	0,04	0,00
Magnesium	mg/l	0,02	0,01	5,80	3,55	0,15	0,10	0,01	0,00
Calcium	mg/l	52,67	2,52	224,33	5,51	81,67	5,03	59,67	1,53
Sulfates	mg/L SO4=	115,33	21,57	828,67	39,72	416,33	61,01	160,00	11,27
Chlorides	mg/L Cl-	462,33	17,90	528,00	19,67	375,67	104,71	345,33	14,74

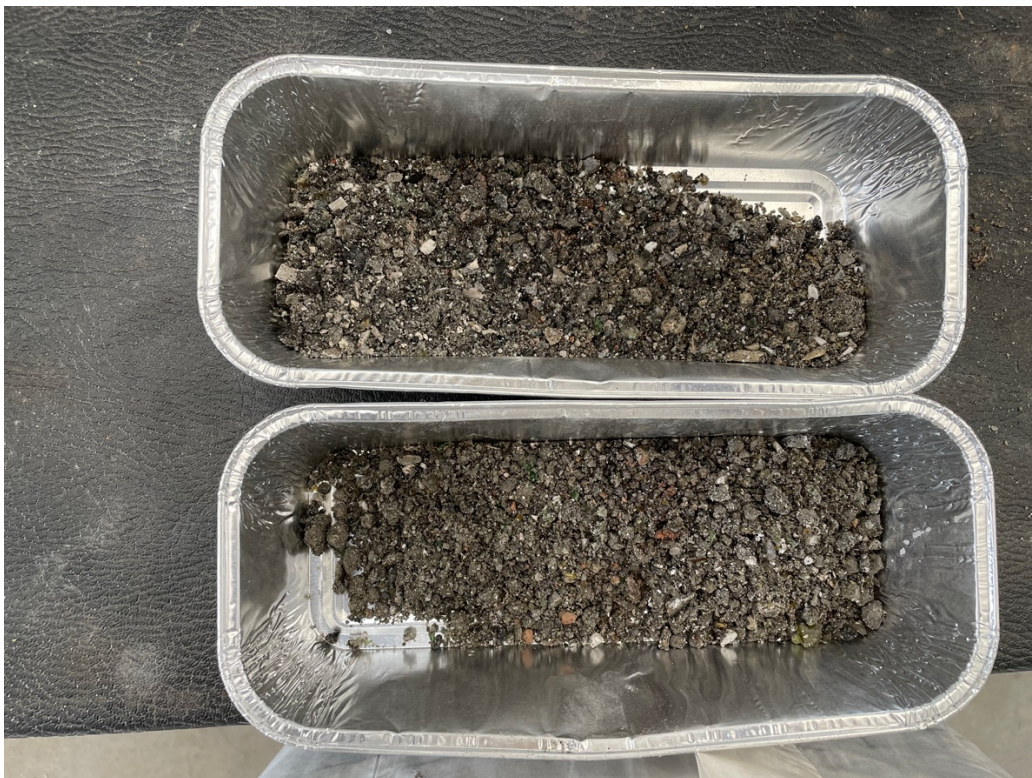
		0/6-00-10			0/6-00-18			0/6-02-18			0/6-03-18		
pH	/	11,82	11,96	11,9	9,26	9,45	9,58	11,32	11,25	11,28	11,53	11,5	11,56
Arsenico	µg/l As	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20
Bario	mg/l Ba	0,08	0,09	0,09	0,05	0,05	0,05	0,04	0,04	0,04	0,06	0,05	0,06
Berillio	µg/l Be	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Cadmio	µg/l Cd	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Cobalto	µg/l Co	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Cromo	µg/l Cr	72	45	45	214	196	199	110	119	117	29	42	33
Mercurio	µg/l Hg	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5
Molibdeno	mg/l Mo	0,06	0,01	0,02	0,11	0,11	0,11	<0,01	0,03	0,02	0,02	<0,01	<0,01
Nichel	µg/l Ni	15	14	14	8,6	8,7	8	11	13	12	8,8	10	9,4
Piombo	µg/l Pb	14	23	28	<5	<5	<5	<5	<5	5	8,9	5,2	20
Rame	mg/l Cu	0,89	0,87	0,92	0,79	0,77	0,76	0,75	0,89	0,83	0,6	0,65	0,63
Antimonio	mg/l Sb	0,03	0,03	0,02	0,09	0,07	0,07	0,05	0,06	0,06	0,04	0,04	0,04
Selenio	µg/l Se	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Vanadio	µg/l V	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Zinco	mg/l Zn	0,046	0,079	0,203	0,003	0,002	0,004	0,023	0,02	0,02	0,038	0,034	0,043
Sodio	mg/l	205	216	224	230	221	193	220	222	210	149	163	156
Potassio	mg/l	301	319	328	285	273	280	273	313	299	220	238	230
Magnesio	mg/l	0,02	0,01	<0,01	9,9	3,8	3,7	0,22	0,2	0,03	0,01	0,01	0,01
Calcio	mg/l	55	53	50	218	227	228	77	87	81	58	60	61
Fluoruri	mg/l F-	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Solfati	mg/L SO4=	140	100	106	860	842	784	415	356	478	153	173	154
Nitrati	mg/L NO3-	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25	<0,25
Cloruri	mg/L Cl-	458	447	482	546	531	507	295	494	338	334	362	340

ANNEX XIII: Experimental setup photos

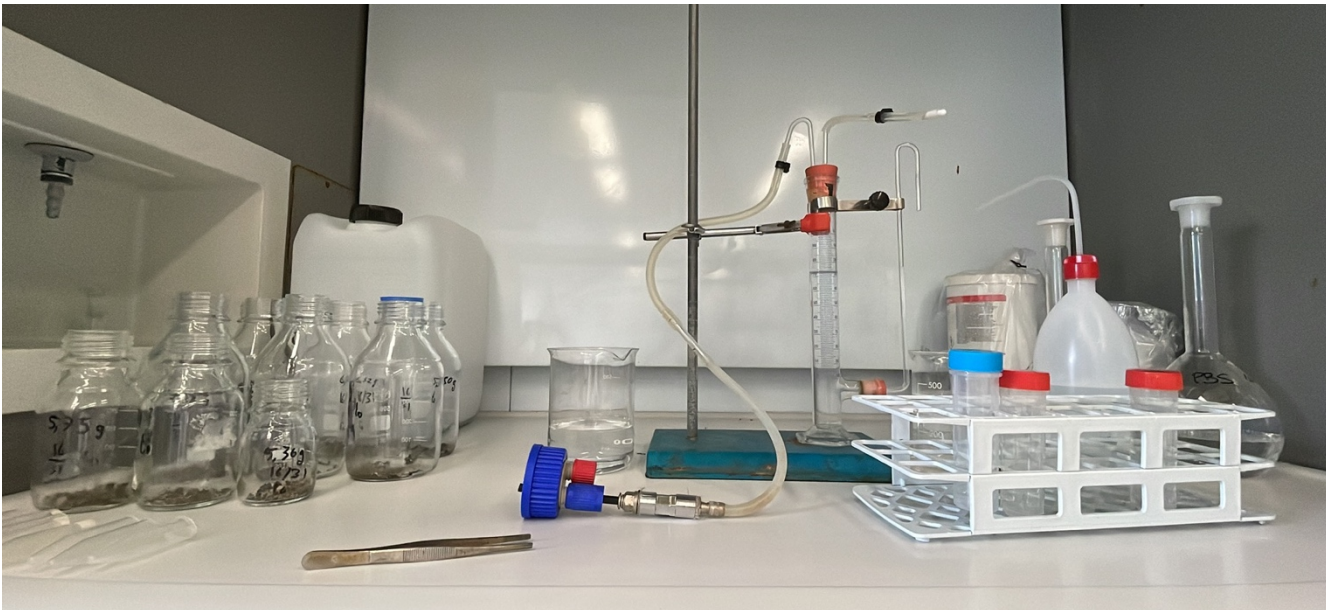
Aluminum vessels containing 6/16 fraction sample with applied L/S=0.2 L/kg ready to be incubated for 4h in the static chamber for accelerated carbonation treatment.



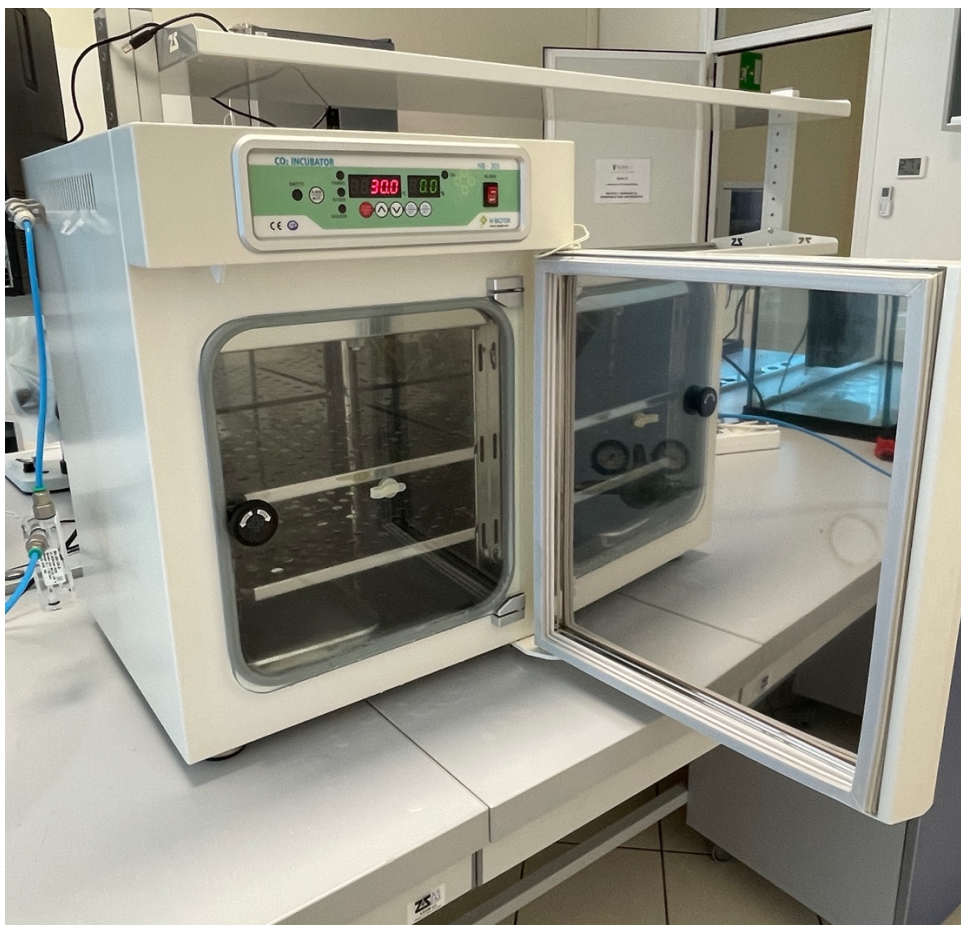
16/31 mm fraction samples have been crushed before to carry out calcimetry test



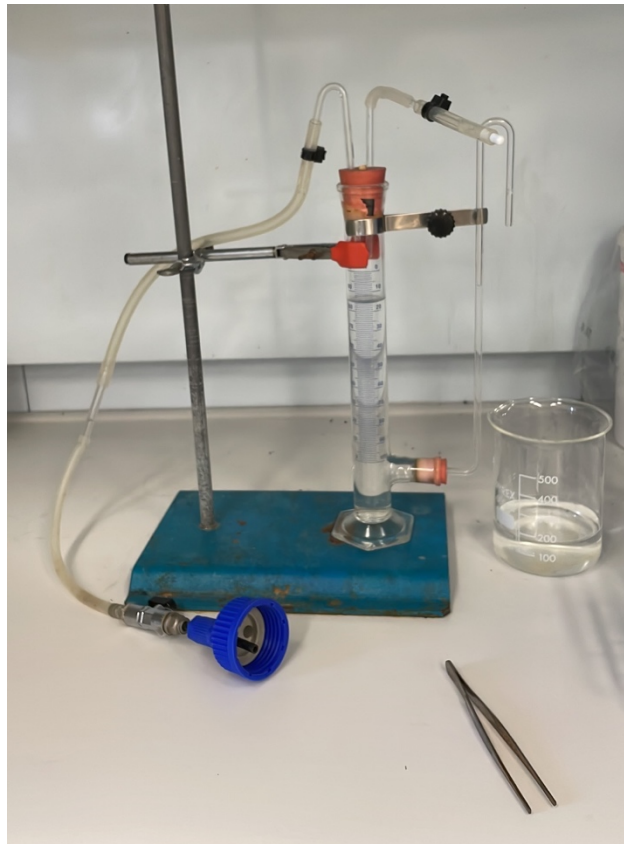
Experimental setup to carry out calcimetry analyses as described in DM 13 09 1999 Met. V.1. On the left bottles containing 5g of sample. In the center the Dietrich-Fruehling calcymeter. On the right Falcon test tubes containing HCl.



Empty CO₂ incubator utilized for carry out the accelerated carbonation tests.



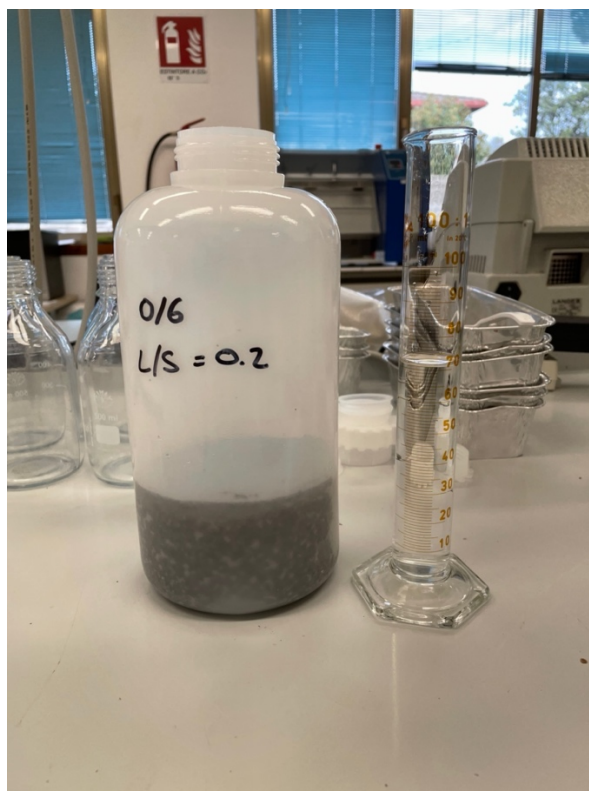
Dietrich-Fruehling apparatus utilized.



CO₂ incubator filled with aluminum vessels containing samples. Temperature (28.6°C) and CO₂ concentration (10%) can be read on the control panel.



2L plastic bottle filled with 600g of 0/6 mm fraction sample and water for an amount of $L/S=0.2$ L/kg. the bottle is then inserted in the agitator for 30 min at 8 rpm.



CO₂ cylinder connected to the incubator.



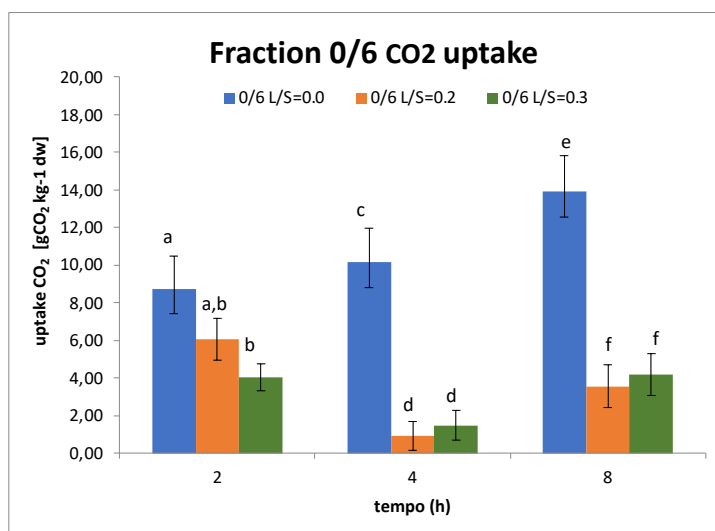
ANNEX XIV: Experimental Results

Tables and graphs with the experimental results expressed in gCO₂/kg dw **uptake**. A, B, C letters stand for the granulometry, namely 0/6, 6/16 and 16/31mm.

The following 3 graphs and table shows the uptake on the same fraction (fraction fixed) with different times and L/S ratio.

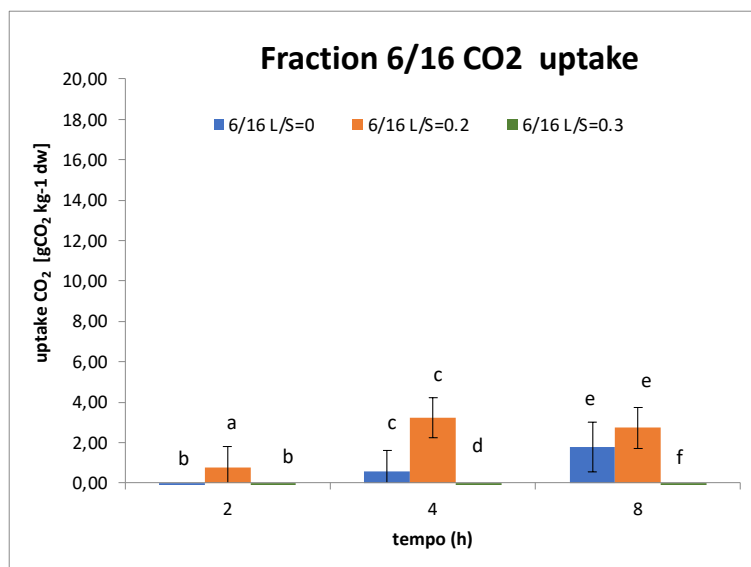
Uptake CO₂ [gCO₂ kg⁻¹ dw] fraction 0/6

t (h)	A/00/	SE	A/02/	SE	A/03/	SE
0	0,00 ±	1,75	0 ±	1,09	0 ±	0,72
2	8,75 ±	1,79	6,059964 ±	0,79	4,04 ±	0,7831682
4	10,15 ±	1,90	0,9182389 ±	1,12	1,47 ±	1,1161989
8	13,92 ±	1,35	3,5564022 ±	1,11	4,19 ±	0,8874718



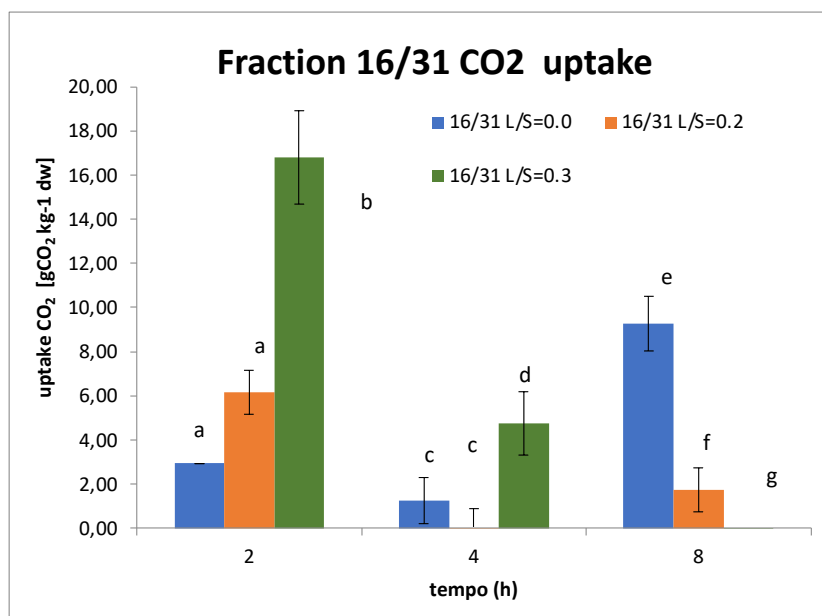
uptake CO₂ [gCO₂ kg⁻¹ dw] fraction 6/16

t (h)	B/00/	SE	B/02/	SE	B/03/	SE
0	0,00	±1,80	0,00	±1,42	0,00	±1,54
2	-2,44	±0,75	0,79	±0,52	-4,20	±0,48
4	0,57	±0,38	3,24	±1,58	-4,81	±0,89
8	1,78	±0,65	2,73	±0,56	-2,72	±0,89



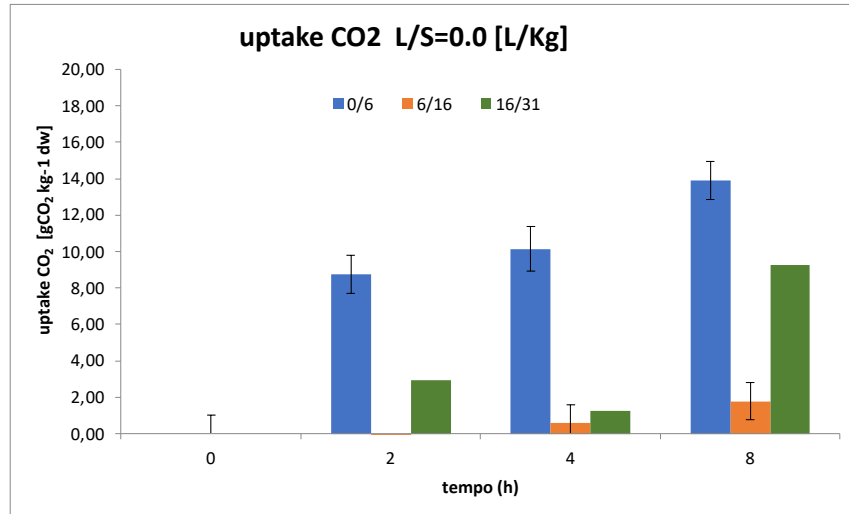
uptake CO₂ [gCO₂ kg⁻¹ dw] fraction 16/31

t (h)	C/00/	SE	C/02/	SE	C/03/	SE
0	0,00	±0,51	0,00	±1,01	0,00	±0,39
2	2,93	±1,13	6,17	±1,48	16,81	±2,13
4	1,25	±0,51	-0,13	±0,23	4,74	±1,43
8	9,28	±0,90	1,73	±0,80	-1,64	±0,27

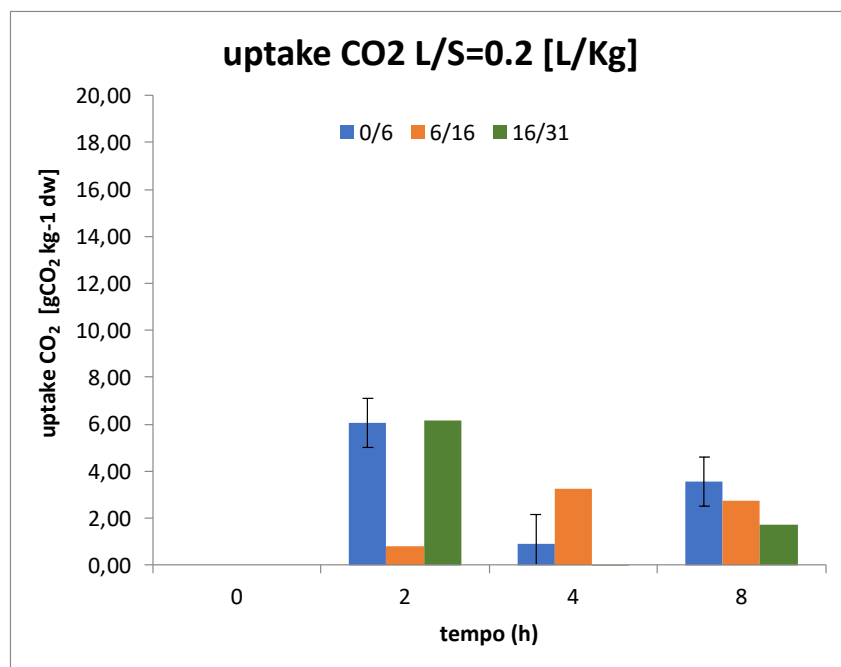


The following 3 graphs and table shows the uptake on the different fractions when the L/S is fixed.

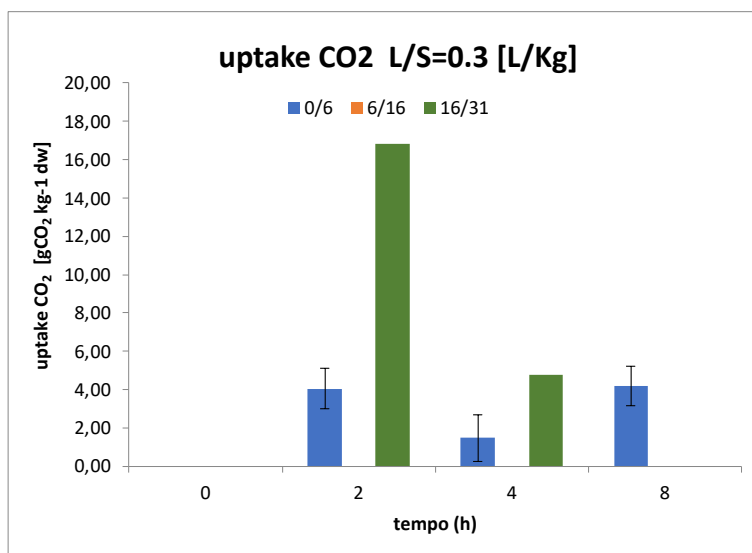
uptake CO ₂ [gCO ₂ kg ⁻¹ dw] L/S=0.0 [L/Kg]			
t (h)	A/00/	B/00/	C/00/
0	0,00	0,00	0,00
2	8,75	-2,44	2,93
4	10,15	0,57	1,25
8	13,92	1,78	9,28



uptake CO ₂ [gCO ₂ kg ⁻¹ dw] L/S=0.2 [L/Kg]			
t (h)	A/02/	B/02/	C/02/
0	0,00	0	0
2	6,06	0,79	6,17
4	0,92	3,24	-0,13
8	3,56	2,73	1,73

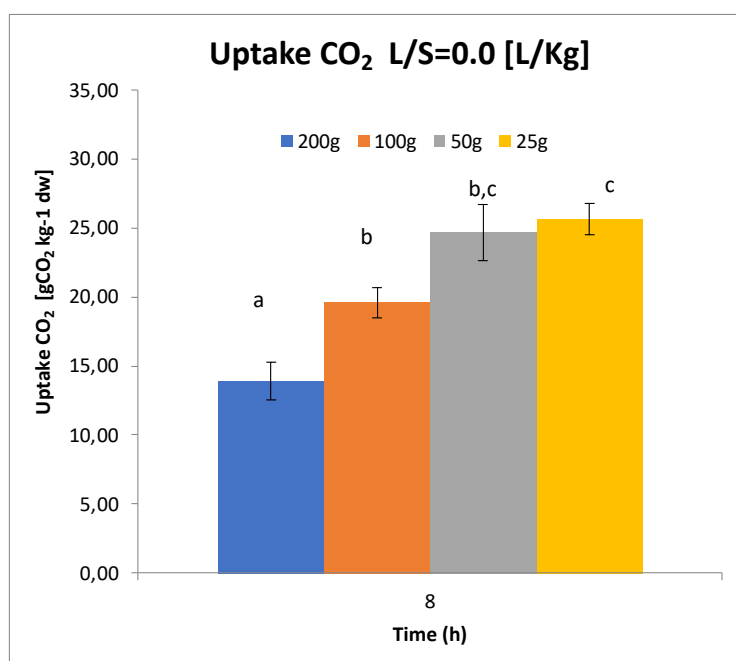


uptake CO2 [gCO2 kg-1 dw] L/S=0.3 [L/Kg]			
t (h)	A/03/	B/03/	C/03/
0	0,00	0	0
2	4,04	-4,20	16,81
4	1,47	-4,81	4,74
8	4,19	-2,72	-1,64



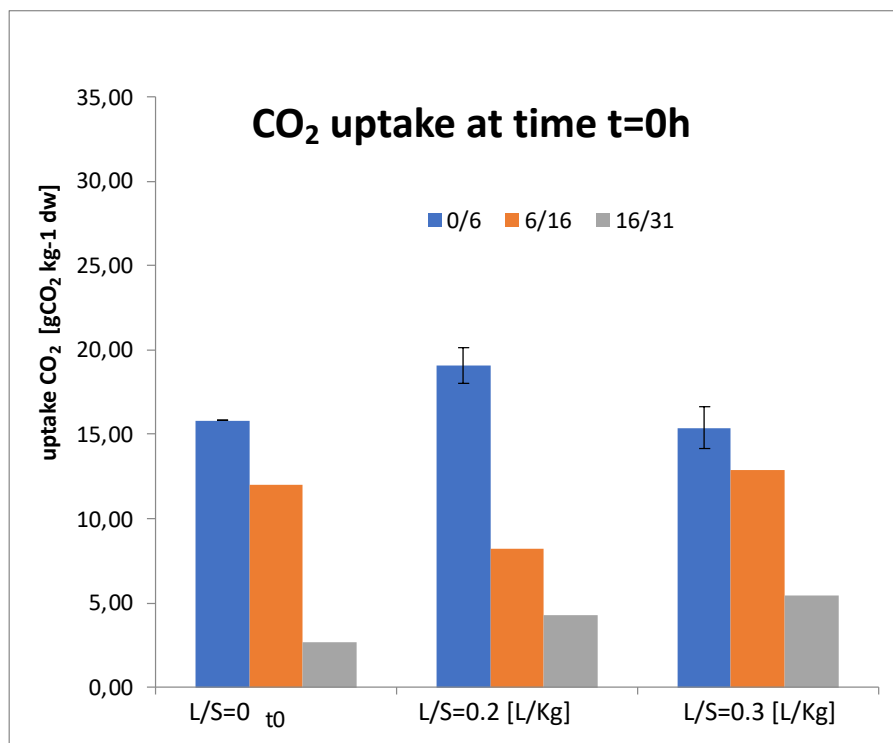
Uptake of 0/6mm fraction with L/S =0.0 L/kg and different masses: i.e. 200, 100, 50, 25g.

uptake CO2 [gCO2 kg-1 dw] L/S=0.0 [L/Kg]								
t (h)	A/00/200g		A/00/100g		A/00/50g		A/00/25g	
		SE		SE	g	SE	g	SE
8	13,92	1,35	19,59	±1,12	24,67	2,06	25,64	±1,14



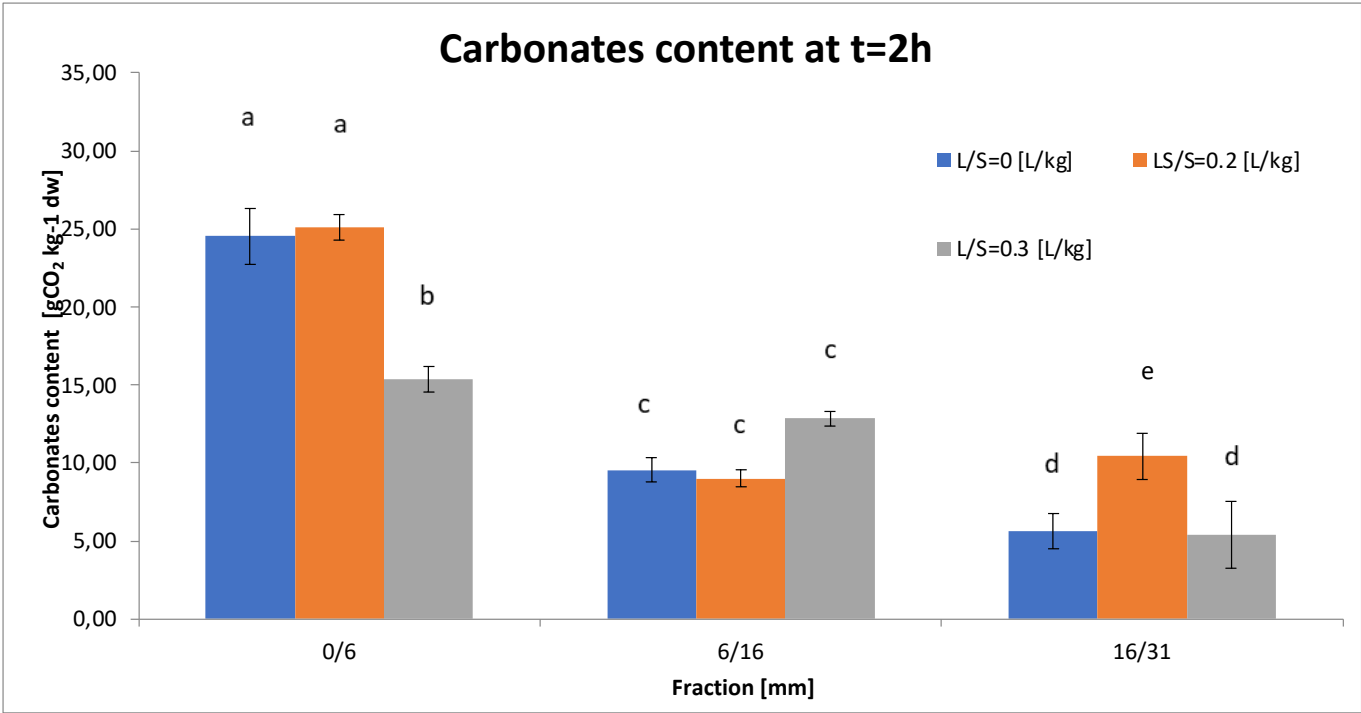
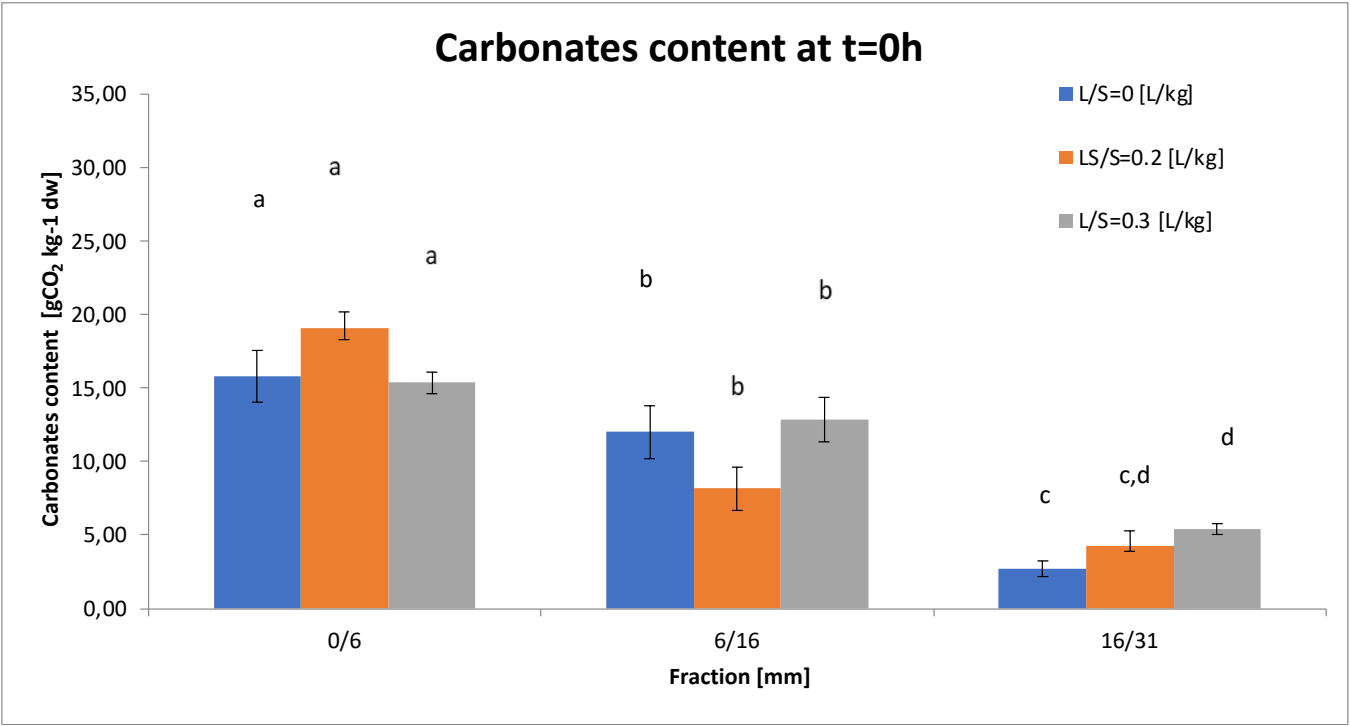
In the following table the carbonates content of the different fraction with different L/S ratio are displayed

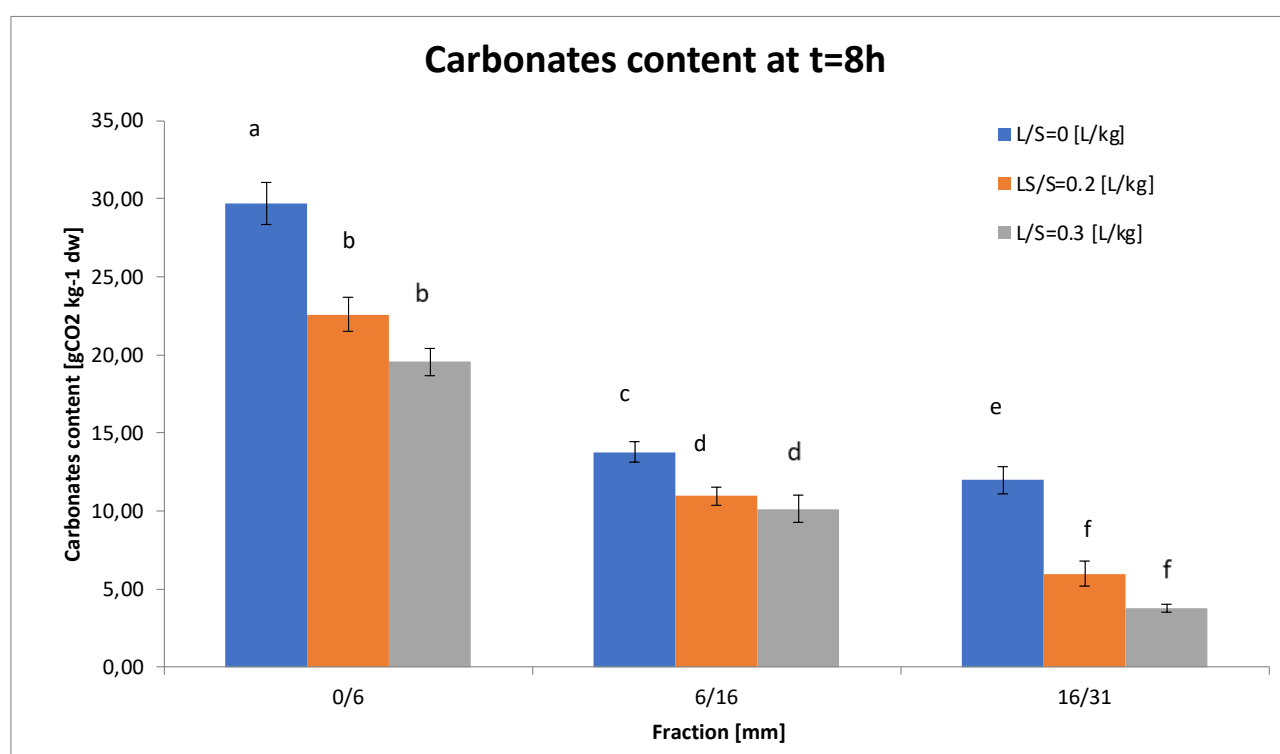
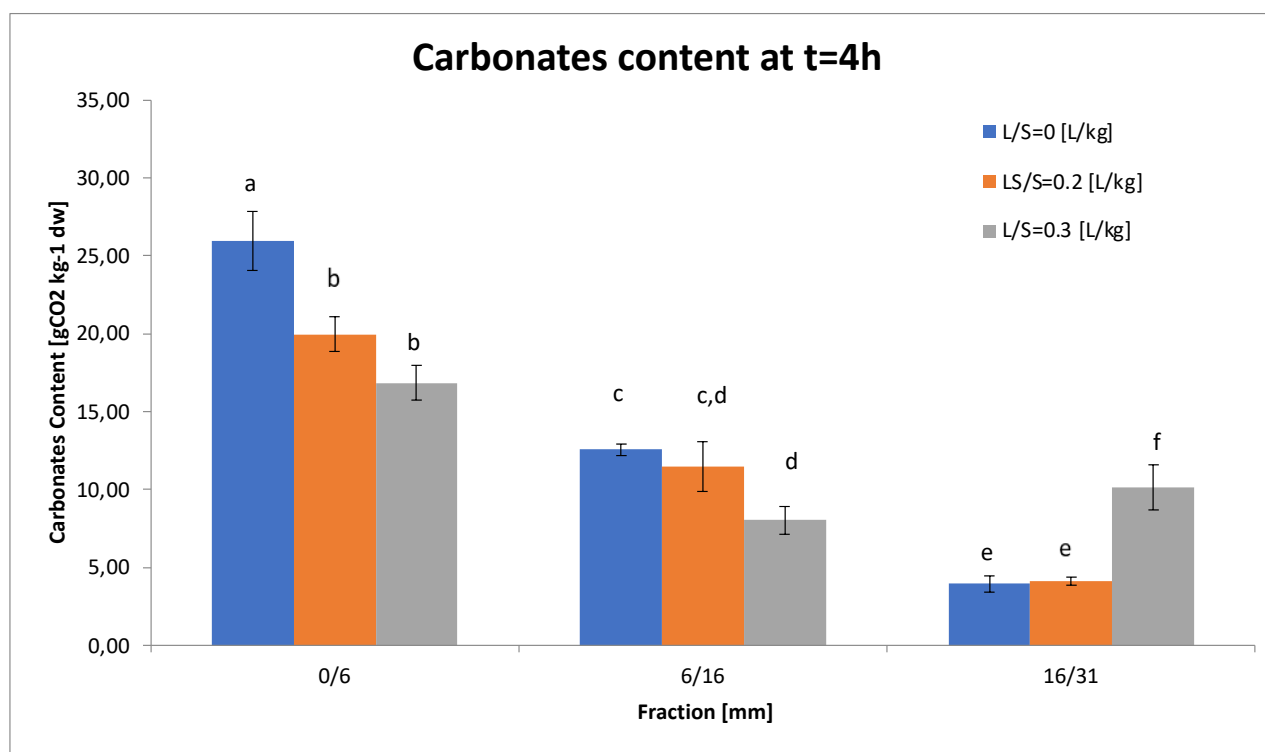
Carbonates content t=0h	
Fraction	gCO ₂ /kg dw
A/00	15,79
A/02	19,04
A/03	15,36
B/00	12,00
B/02	8,22
B/03	12,85
C/00	2,69
C/02	4,26
C/03	5,42



In the following table and graphs the carbonates content are reported in terms of gCO₂/kgdw.

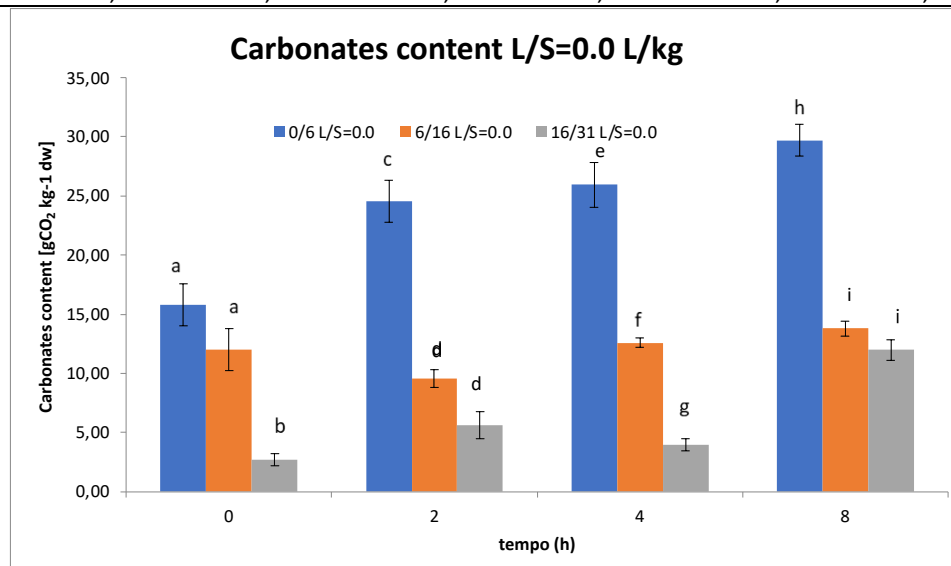
CARBONATES CONTENT									
Fraction	L/S ratio	0h	SE	2h	SE	4h	SE	8h	SE
0/6	L/S=0 [L/kg]	15,79	±1,75	24,54	±1,79	25,94	±1,90	29,71	±1,35
6/16		12,00	±1,80	9,56	±0,75	12,57	±0,38	13,78	±0,65
16/31		2,69	±0,51	5,62	±1,13	3,94	±0,51	11,97	±0,90
0/6	LS/S=0.2 [L/kg]	19,04	±1,09	25,10	±0,79	19,96	±1,12	22,59	±1,11
6/16		8,22	±1,42	9,01	±0,52	11,46	±1,58	10,95	±0,56
16/31		4,26	±1,01	10,43	±1,48	4,13	±0,23	5,99	±0,80
0/6	L/S=0.3 [L/kg]	15,36	±0,72	19,40	±0,78	16,83	±1,12	19,55	±0,89
6/16		12,85	±1,54	8,65	±0,48	8,04	±0,89	10,13	±0,89
16/31		5,42	±0,39	22,23	±2,13	10,16	±1,43	3,78	±0,27



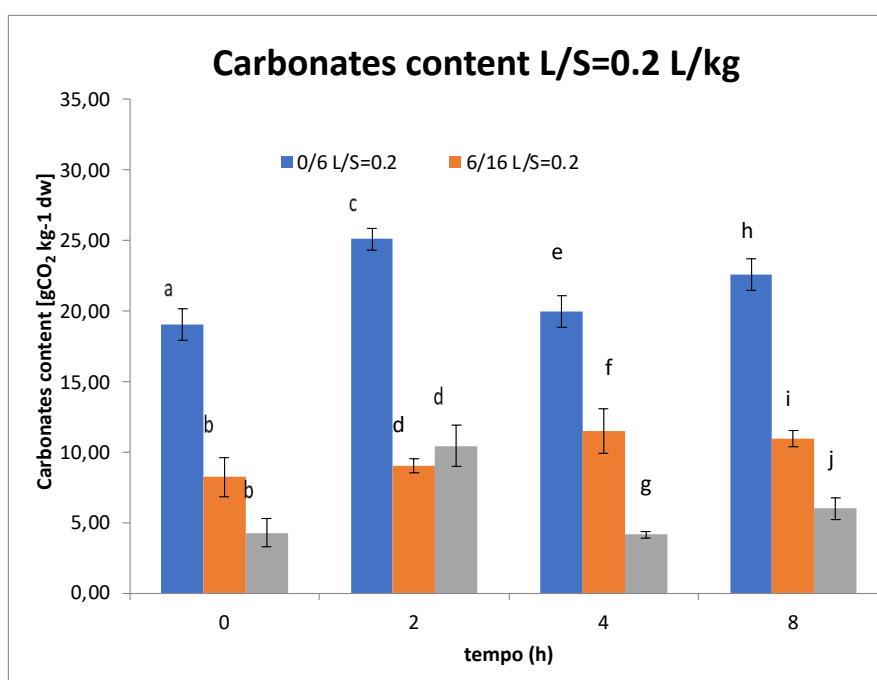


L/S is fixed, and carbonates content is displayed related to time and granulometry.

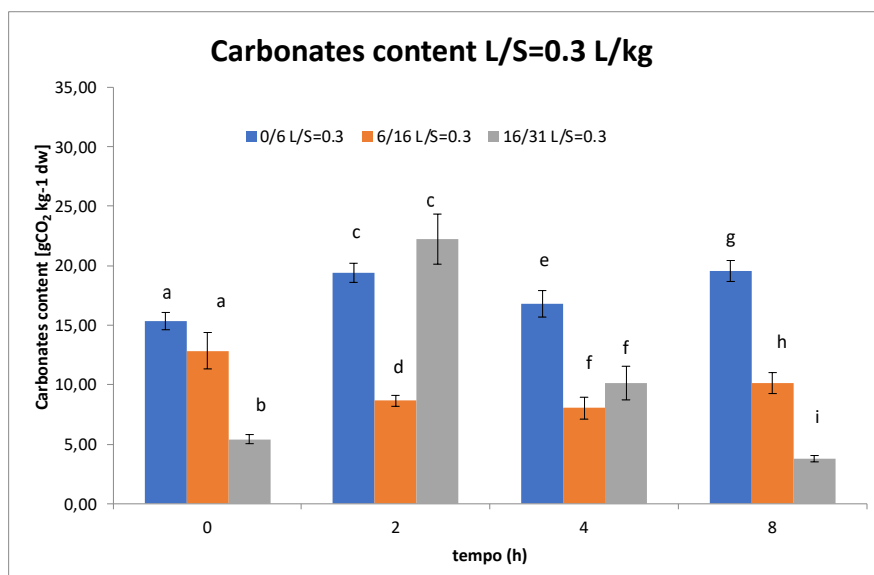
CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] L/S=0.0 [L/Kg]						
t (h)	A/00/	SE	B/00/	SE	C/00/	SE
0	15,79	±1,75	12,00	±1,80	2,69	±0,51
2	24,54	±1,79	9,56	±0,75	5,62	±1,13
4	25,94	±1,90	12,57	±0,38	3,94	±0,51
8	29,71	±1,35	13,78	±0,65	11,97	±0,90



CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] L/S=0.2 [L/Kg]						
t (h)	A/02/	SE	B/02/	SE	C/02/	SE
0	19,04	±1,09	8,22	±1,42	4,26	±1,01
2	25,10	±0,79	9,01	±0,52	10,43	±1,48
4	19,96	±1,12	11,46	±1,58	4,13	±0,23
8	22,59	±1,11	10,95	±0,56	5,99	±0,80

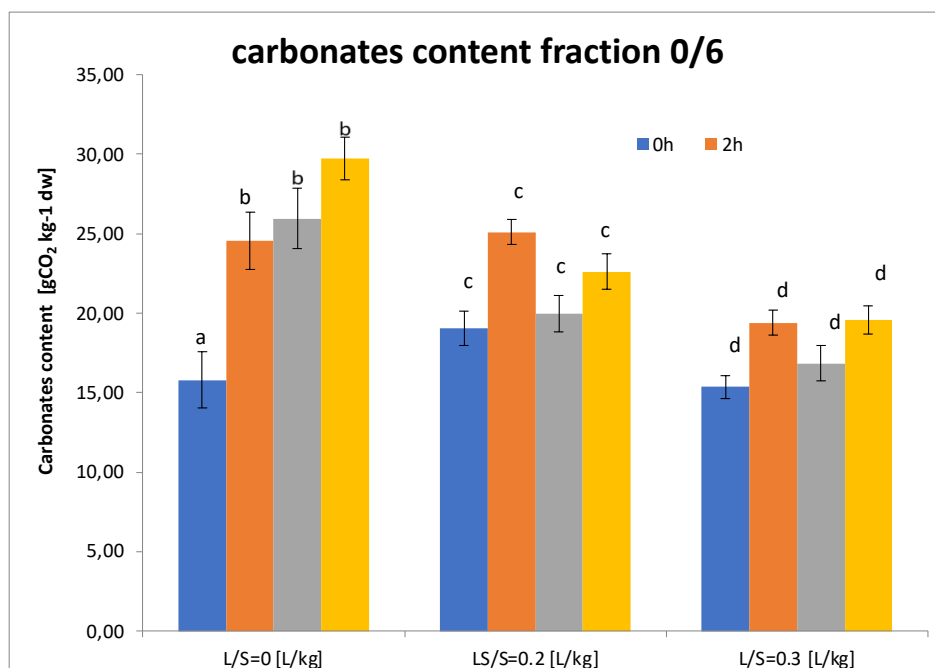


CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] L/S=0.3 [L/kg]						
t (h)	A/03/	SE	B/03/	SE	C/03/	SE
0	15,36	±0,72	12,85	±1,54	5,42	±0,39
2	19,40	±0,78	8,65	±0,48	22,23	±2,13
4	16,83	±1,12	8,04	±0,89	10,16	±1,43
8	19,55	±0,89	10,13	±0,89	3,78	±0,27

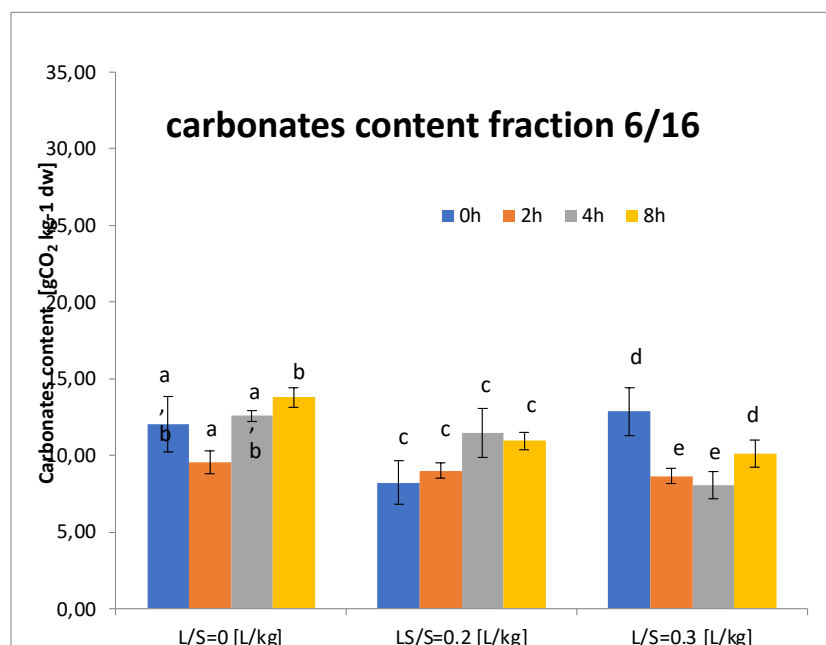


The following graphs and tables display the carbonates content when the fraction is fixed.

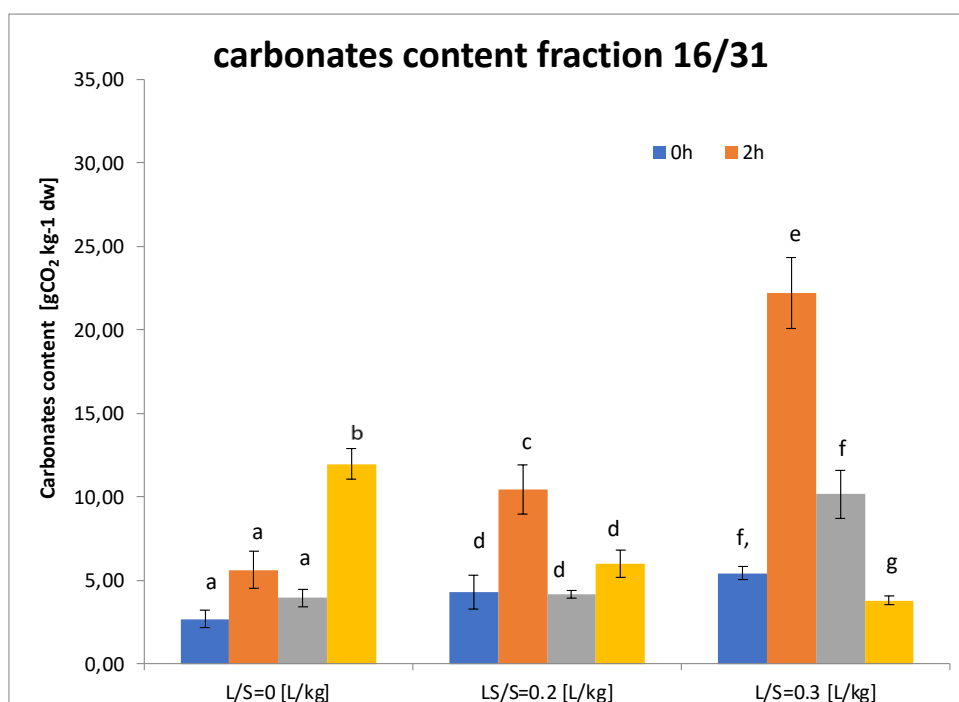
CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] FRACTION 0/6								
0/6	0h	SE	2h	SE	4h	SE	8h	SE
L/S=0	15,79	1,75	24,54	1,79	25,94	1,90	29,71	1,35
L/S=0.2	19,04	1,09	25,10	0,79	19,96	1,12	22,59	1,11
L/S=0.3	15,36	0,72	19,40	0,78	16,83	1,12	19,55	0,89



CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] FRACTION 6/16								
6/16	0h	SE	2h	SE	4h	SE	8h	SE
L/S=0	12,00	1,80	9,56	0,75	12,57	0,38	13,78	0,65
L/S=0.2	8,22	1,42	9,01	0,52	11,46	1,58	10,95	0,56
L/S=0.3	12,85	1,54	8,65	0,48	8,04	0,89	10,13	0,89



CARBONATES CONTENT [gCO ₂ kg ⁻¹ dw] FRACTION 16/31								
16/31	0h	SE	2h	SE	4h	SE	8h	SE
L/S=0	2,69	0,51	5,62	1,13	3,94	0,51	11,97	0,90
L/S=0.2	4,26	1,01	10,43	1,48	4,13	0,23	5,99	0,80
L/S=0.3	5,42	0,39	22,23	2,13	10,16	1,43	3,78	0,27



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