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TESI DI LAUREA

ACCELERATED CARBONATION: AN INNOVATIVE METHOD FOR CARBON DIOXIDE SEQUESTRATION FROM ALKALINE SOLID WASTES

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ABSTRACT

The scope of this thesis is the study of the quantity of CO_2 that different alkaline residues (Portland cement residues, foundry sands, clayey silts, and fly ashes) can absorb inside them through a particular reaction called accelerated carbonation. This evaluation has been carried out by analyzing different samples, directly in the laboratory, under controlled operative conditions.

Thus, the study has been divided into three parts: a first introduction that reports the general aspects of carbonation, starting from its advantageous and disadvantageous characteristics and finishing with the chemistry of the process and the description of the methods that can be used to measure the CO_2 absorption, a second part which is the section containing all the experimental activities performed, and, finally, a third fraction reporting all the graphs, tables, and data used in the thesis.

In particular, the second part of the work includes: a critical review realized by analyzing many papers based on accelerated carbonation, which have been selected from the literature to have good and complete knowledge about the process and, above all, of the results ($g_{CO2\_ABSORBED}/kg_{SORBENT}$) that can be found using different materials with different operative conditions, the experimental design used for the analysis, the procedures followed for the process, and all the chemical analyses performed at the end of the carbonation. In the end, the experimental results, in terms of carbon dioxide sequestration, have been explained and compared with the ones coming from the literature analysis.

PART I – INTRODUCTION

1. CLIMATE CHANGE

"Climate change means a change of climate which is attributed directly or indirectly to human activity that alters the composition of the global atmosphere and which is in addition to natural climate variability observed over comparable time periods" (United Nations Framework Convention on Climate Change –UNFCC, 1992, Article 1).

Thanks to this definition, it's possible to understand that human behavior is modifying the earth's climate, causing many adverse effects in every planet's region, and creating lots of problems for human life. Human activities, in fact, due to their greenhouse gas (GHG) emissions, mainly consisting of CO_2 and methane (CH_4), are the main cause of global warming. It has been assessed that the average global surface temperature measured during the last two decades (2001 – 2020) was 0.99 °C higher than 1950 – 1900. Moreover, it can be affirmed that since 1970 the temperature of the planet has increased at the highest rate compared to any other 50 – years period over the last 2000 years, and its raising since 1750 is caused by GHG emissions generated by anthropic activities such as fossil fuel combustions, industrial processes, agriculture operations, and transport usages. Another important aspect that allows us to better understand this situation is related to the CO_2 concentration that was equal to 410 parts per million (ppm), a value that has never been reached in any time over the past two million years, and that is 47 % higher than the one measured in 1750. In 2019 the global net quantity of anthropogenic GHG emissions was 59 $GtCO_2$ – eq and, considering the direct and indirect emissions by sector as presented in Figure 1, it's possible to say that the building category produced 16 % of the total value, transport one generated 15 % of it, agricultural one is responsible of the 22 % of it, and, finally, the emissions related to the industrial sector and the energy production process are respectively the 34 % and the 13 % of the total amount. (IPCC sixth assessment report, 2023).

Climate change has already caused and is still generating a lot of important damage, which could be irreversible, in terrestrial, freshwater, cryosphere, and ocean ecosystems. Indeed, due to global warming, mass mortality episodes both on land and in the ocean have been studied, and the retreat of glaciers causing hydrogeological changes rather than permafrost thaw processes have been observed. Moreover, other phenomena such as sea level rise, ocean acidification, land desertification, and exceptional meteorological episodes are caused by this situation.

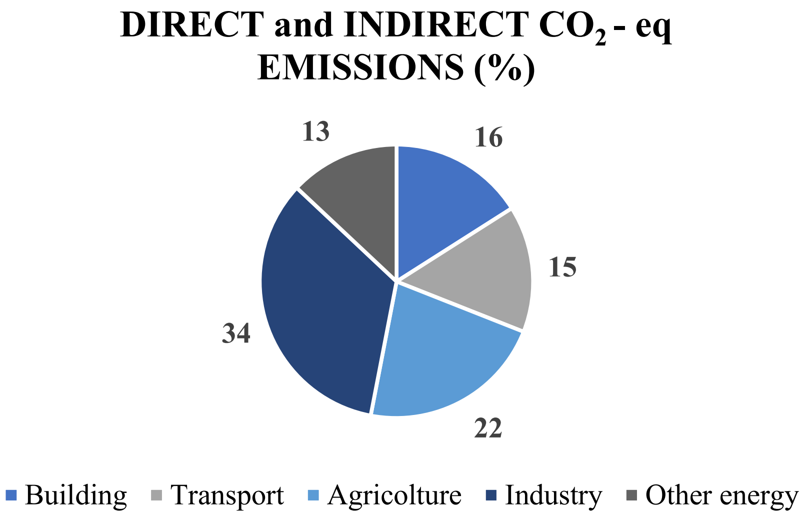


Figure 1: Direct and Indirect global CO_2 - eq emissions by sector (IPCC sixth assessment report, 2023).

For these reasons, it's necessary to implement new methods or improve the existing ones trying to reduce climate change or, at least, to control and reduce its negative effects; thus, to do this, lots of international environmental agreements have been established. They are necessary to increase people's awareness about the problem, trying to increase the efforts to reduce the emissions of CO_2 in all the activities sectors.

In particular, the Paris Agreement adopted in 2015 under the UNFCCC (United Nations Framework Convention on Climate Change) established different environmental targets at national and sub-national levels related to mitigation and adaptation actions against climate change that have to be reached. The main scope is to keep the global temperature increase well below 2 °C compared to the pre-industrial levels, but, at the same time, continuously work to don't exceed it over 1.5 °C to reduce the problems caused by climate change. However, now there is still a gap between the actual GHG emissions and the ones that have to be reached, so proceeding in this way, without any other strengthening of policies and implementation of new systems to reduce CO_2 emissions, emissions will rise, leading to global warming of 2.2 °C – 3.5 °C by 2100 with irreversible effects on the earth. In this situation, it's necessary to reach as soon as possible the condition of net zero or negative CO_2 emission that corresponds to the case in which the quantity of carbon dioxide production is equal to or lower than the amount removed from the atmosphere. This is the only way that allows us to limit climate change, but it requires rapid reductions of CO_2 emissions and, at the same time, a great improvement of the technologies to reduce the concentration of the gas in the atmosphere.

In this perspective, global net zero CO_2 has to be reached at the beginning of 2050s to maintain the global temperature increase below 1.5 °C and around 2070s to don't exceed the value of 2 °C of rising.

For the industrial sector, which, as previously said, is the main source of CO_2 emissions, there are different options, based on the circular economy concept, to reduce the emissions. Technics such as Carbon Capture and Storage (CCS), Carbon Capture and Utilisation (CCU), as well as direct air CO_2 capture can be used to strongly decrease the environmental impact of the industries.

The method that is object of this study is the accelerated carbonation reaction which is a CCS technique and, in particular, an approach that allows storing the CO_2 in inert material in a stabilized form. However, it can be considered part of the carbon capture and storage or utilization when the CO_2 is sequestered from anthropogenic emissions sources, whereas in the case of direct carbon dioxide absorption from the atmosphere, the reaction is included in the DACCS (Direct Air Carbon Capture and Storage) methodologies.

In the reports published in 2022 and 2023, this technique is not adequately commented on, and few references are reported in (IPCC, Climate change 2022 – mitigation of climate change), specifically in the sections called “Urban carbon cycle”, “New trends in emissions”, “CCS, CCU, Carbon Sources, Feedstocks, and Fuels”, and “Direct Air Carbon Capture and Storage (DACCS)”.

2. GENERAL CONCEPTS OF CARBONATION

The carbonation is a reaction that allows fixing the CO_2 into stable and inert solids. This process can naturally occur and is the cause of the formation of limestone and carbonic rocks such as the Dolomiti mountains. In particular, it happens when the carbon dioxide gets in touch with natural minerals containing metal oxides, causing the formation of carbonates where the CO_2 can permanently remain for geological scale periods. The great advantage of this process is that the final products of the carbonation are stable and can also generate positive effects on the environment. Unfortunately, this phenomenon is very slow to occur naturally, so the scope is to find a method with faster reaction kinetics, lower energy needs, and minimal energy and material losses. The scheme of a possible carbonation plant is shown in Figure 2.

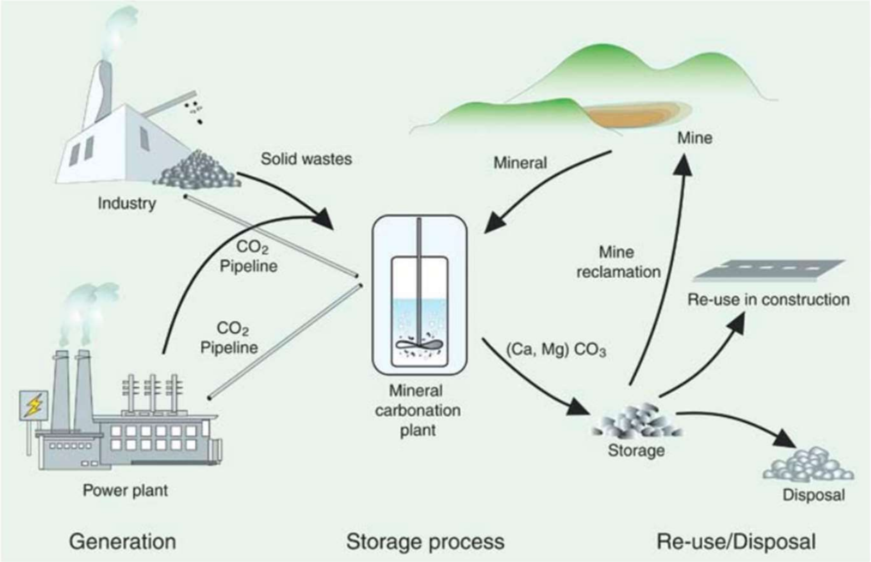


Figure 2: Possible scheme of a carbonation process (IPCC - carbon dioxide capture and Storage 2004)

Suitable materials that can be used for the process are those containing alkaline metals such as sodium (Na), Potassium (K), Calcium (Ca), and Magnesium (Mg), which are present in lots of natural rocks but also many alkaline wastes. However, Sodium carbonates and Potassium are very soluble and can be used only if the storage is done in dry and watertight underground places, so Calcium oxides and Magnesium are the most preferred elements for carbonation. However, Calcium and Magnesium oxides are not present as pure oxides in nature, while it's possible to find them combined with Silicon (Si) and other elements in a silicate form. Thus, the Magnesium oxides can be found in the igneous rocks where the carbonate presence is quite null and, in particular, the main supplier of them are the olivine minerals ($(Mg, Fe) SiO_4$), serpentine minerals ($Mg_3(Si_2O_5)(OH)_4$), and talcum ($Mg_3Si_4O_{10}(OH)_2$). On the other hand, Calcium oxides are mostly present in the wollastonite ($CaSiO_3$), which is much rarer than the previous materials. Moreover, it's possible to affirm that the quantity of Magnesium oxide necessary to absorb carbon dioxide (3,3 kg_{MgO}/kg_{CO2}) is lower than the one required for Calcium oxide usage (4,7 kg_{CaO}/kg_{CO2}), and so it's notable that the farmer also permits a lower cost than the latter. But, even if the quantity of natural rocks on the earth's surface is higher than that one of the alkaline wastes, these last materials are readily available and reactive, so they are easier to use than the others. Furthermore, the carbonation of the alkaline wastes is not only a method to store CO_2 and so does decrease its emission from industries and energy supplier activities, but it also represents a possibility to reuse and recycle waste materials instead of putting them in a landfill. So, carbonation is a method in agreement with the circular economy concept.

Waste streams that can be used for the carbonation reaction include fly ashes from MSW incinerators, bottom ashes, slags coming from BOFs (Basic Oxygen Furnaces), EAFs (Electric Arc Furnaces), and BFs (Blast Furnaces), and cement wastes.

The amount of Calcium oxide, whose use allows a faster process instead of use Magnesium oxide, in these materials is about 30 % by weight of material for the fly ashes, 20 % for the bottom ashes, and 65 % for the slags. Nevertheless, one or more pre-treatment actions have to be performed on the material to increase the efficiency and the velocity of the carbonation. Initially, the sorbent can be processed through a granulometric reduction to have smaller grains and, so, a bigger specific area for better contact between liquid and solid parts and better material reactivity. Moreover, sometimes the removal of the present inert substances can be important in increasing the concentration of useful material. In this case, some different methods could be applied: magnetic, gravimetric, and electrostatic separation.

The carbonation process can be implemented in different methodologies. First of all, it can be done in situ or ex-situ: the former method is performed in the place where the source of CO_2 is located, whereas the latter is done in another location. Thus, in the case of in situ carbonation, the carbon dioxide produced by the combustion process is kept before going into the atmosphere and injected inside the materials (natural rocks or alkaline wastes); in such situations, the operative costs can be abated, but, as same as in ex-situ reaction, it's always necessary to have the sorbent materials already available and treated. For this reason, the possibility of using this methodology in a place where combustion plants produce wastes like fly or bottom ashes is very interesting and is the object of many investigations.

These types of sorbents are generally fine and thus don't need pre-treatments differently from other materials, and their availability in the same place where the combustion occurs makes the cost of transport null.

An aspect that is fundamental for this reaction is the presence of water that permits it to reach a good efficiency in the process and absorb a great quantity of CO_2. A liquid film ensures to have a faster reaction even if a humid process occurs at a lower temperature than the ones of a dry approach, so the quantity of heat and so of energy that can be recovered with this method is less than the second one. On the other hand, a dry approach has to be performed at higher temperatures allowing a greater heat recovery, but its kinetic is very slow. Furthermore, when a humid technique is carried out, it must be considered that the wastewater exiting the carbonation chamber after the reaction occurred has to be properly treated before discharge.

In addition, the carbonation processes can also be divided into two categories: the direct method and the indirect one. The first approach, which can be performed with or without the water, is related to the case in which the CO_2 directly contacts the material, while in the second situation, the reactive elements (Ca^{2+}, Mg^{2+}) are firstly extracted from the sorbent using acid (HCl) or other solvents, and then contacted the carbon dioxide to increase the efficiency of the reaction even if the operative cost increases.

Finally, it's possible to affirm that this CCS technique is an advantageous method because it firstly allows for a decrease in the quantity of CO_2 emitted by the industries and, so, also its concentration in the atmosphere by storing it in a stable form that can remain like this for a geological period without creating any problems. The final products are stable and inert because the carbonation also improves the environmental quality of the sorbent used, reducing the possible releases of heavy metals from it and improving the mechanical properties of the cement aggregates by making them suitable for other construction activities.

3. PRESENT SITUATION AND FUTURE PLANS

Lots of studies have been performed to determine the best condition to operate to reach the maximum efficiency and to absorb as much quantity of CO_2 as possible in the chosen sorbents. Nevertheless, these analyses have always been performed at a lab scale, and only in a few cases pilot plants for accelerated carbonation have been built, so the problem is that there isn't an industrial plant yet, and the results are all associated with experimental research. The difficulties consist in quantifying the flux of materials entering and exiting the carbonation chamber and minimizing the amount of energy required by the process. The most preferred option is to implement the accelerated carbonation technic in places where both carbon dioxide emissions and alkaline waste production are present, thus near an incinerator plant. In this case, the operative costs could be abated because the transport of sorbent or CO_2 from one location to another is not needed even if some pre-treatments, such as the granulometric reduction, have to be done.

Regarding the estimation of the operative costs of a plant like this, there aren't real data, and the only ones existing are extrapolated from the lab test and scaled up to an industrial one. Some hypotheses confirm that the price of sequestered CO_2 could be 70 €/t, a value that could be decreased to 20 €/t – 30 €/t if the procedure is efficiently done and the thermic energy is recovered from the reaction, a realistic possibility that has still to be confirmed (Ciccu et al. 2011).

Carbonation is an exothermic reaction, so the recovery of energy is a real possible option to abate the costs, which can also be decreased by thinking about the generated by-products that could be reused in other projects instead of discharged into landfills.

Thus, the plan is to increase the efficiency of this methodology, decrease the cost, the energy and material losses, and, finally, build a plant on an industrial scale.

3.2 Existing plants

3.2.1 Carbon8 System Ltd

As said in the previous chapters, accelerated carbonation technology (ACT) is a reaction taking place when carbon dioxide combines with Calcium or Magnesium oxides that can be present in lots of alkaline industrial wastes to produce carbonates. A real application of this technique was performed by the Carbon8 System Ltd., a spinout- company of Greenwich University, formed in 2006, that applied accelerated carbonation to hazardous wastes, producing non-hazardous construction products. From 2009 to 2012, lots of pilot and full-scale plants were studied to demonstrate their efficiency and, having reached the commercial license for the use of accelerated carbonation to treat air pollution residues (APCs) in the UK in 2010, in 2012 the company implemented a commercial plant at Brandon in Suffolk, UK which represented the first plant of its kind ever built in the world and which is still in operation with a capacity to treat 30’000 tons of APCs per year. Then, in 2016 a second commercial plant was commissioned, soon followed by a third one, making the Carbon8 System Ltd a world leader in this field. Furthermore, in 2018 it created the CO_2ntainer: a device, presented in Figure 3, representing a compact and mobile CCUS solution to implement the accelerated carbonation process.

The CO_2ntainer permits the capture of carbon dioxide directly from the flue stack, without any pre-treatment, and induces its reaction with the industrial residues destinated for landfill. In this way, it’s possible to capture 1’500 – 4’000 t_{CO2}/year by treating 12’000 t_{wastes}/year. Inside the CO_2ntainer, accelerated carbonation occurs, permanently storing CO_2 within 20 minutes, and producing carbonate products useful for the construction industry in a variety of applications such as cement blocks, road fillers, and green roofing substrates.

Figure 3: Representation of the CO_2ntainer created in 2018 (Carbon8 Systems Ltd).

3.2.2 Carbfix project

Another example of a carbonation process executed on an industrial scale is represented by the Carbfix CCS Facility, a project implemented in Iceland based on CO_2 storage activity. The project started in 2007 with the idea of accelerating a natural process of carbon dioxide fixation into natural underground rocks, forming carbonate minerals that can be stable for hundreds of years. After a first preparation phase necessary to optimize the process through lab experiments, in 2012 the project was scaled up to a pilot scale plant located 3 km far from the Hellisheidi power plant in South-West Iceland. The results obtained by dissolving firstly 175 tons (100 $\%_{CO_2}$) and secondly 73 tons (75 $\%_{CO_2}$ and 25 $\%_{H_2S}$) confirmed that 95 % of the total carbon dioxide was mineralized to carbonate minerals in less than two years, validating the great velocity of the process (Matter et al. 2016).

Thanks to these good results, the project was implemented on an Industrial scale in 2014, and it's still continuously running without any incident ever since, injecting 12'000 tons/year of CO_2 into shallow basalts (400 – 800m depth). The scheme and the top view of the plant are presented in Figure 4.

Furthermore, in 2017 Carbfix also adopted a direct air capture (DAC) technology to remove CO_2 directly from the atmosphere, and now it's still growing in developing a method to use seawater as a source of H_2O to dissolve the carbon dioxide before its injection inside the rocks to possibly expand the applicability of this technology also in the water-scarce regions.

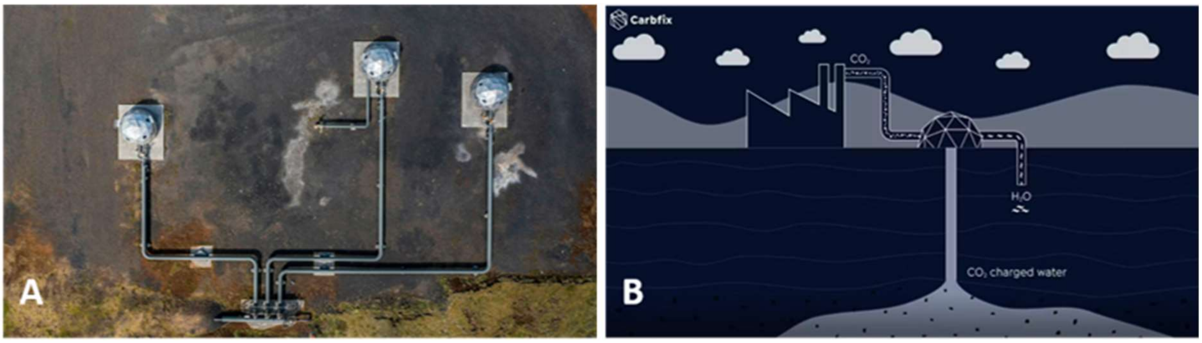


Figure 4: Carbfix CCS facility: (A) Top view of the plant; (B) scheme of the plant.

Taking into consideration the basaltic rocks present worldwide, a storage potential of mineral carbonation equal to 100'000 Gt_{CO2} - 250'000 Gt_{CO2} has been assessed. But, even if this amount is very significant, other industrial-scale plants like Carbfix are needed to reach it.

4. CHEMISTRY OF THE REACTION

4.1 General chemical aspects of carbonation

As already said in the previous chapter, carbonation is a reaction that occurs when the CO_2 reacts with metal oxides such as Magnesium (MgO) and Calcium oxides (CaO), generates carbonates as final products, and releases heat according to the following reaction:

$$MO + CO_2 \rightarrow MCO_3 + heat$$

Where M is the divalent metal (Calcium, Magnesium), so MO is the metal oxide, and MCO_3 is the carbonate produced at the end of the process.

Regarding the heat, its production depends on the specific metal used as a source of oxides, and its quantity is generally a large portion of the total amount generated by the upstream combustion activities that produce carbon dioxide (393,8 kJ/mol_{CO2} for combustion of elemental carbon). In the case of Calcium oxide, the reaction becomes:

$$CaO + CO_2 \rightarrow CaCO_3 + heat$$

The heat generated in this process is 46 % of the previous total quantity, thus a value equal to 181,15 kJ/ mol_{CO2} that corresponds to a negative enthalpy ($\Delta H = -181,15$ kJ/ mol_{CO2}), indicating the presence of an exothermic reaction.

While considering the method based on the use of Magnesium oxides (MgO) the carbonation became:

$$MgO + CO_2 \rightarrow MgCO_3 + 118\ kJ/mol_{CO_2}$$

Regarding the natural silicates such as the Olivine, Serpentine, and Wollastonite, already cited before, the exothermic reactions of carbonation are the following:

Olivine:

$$Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2 + 89\ kJ/mol_{CO_2}$$

Serpentine:

$$Mg_2Si_2O_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + 2SiO_2 + 2H_2O + 64\ kJ/mol_{CO_2}$$

Wollastonite:

$$CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2 + 90\ kJ/mol_{CO_2}$$

Having said that the process is exothermic, it's possible to affirm that, from a thermodynamic point of view (Le Châtelier principle), the production of carbonates can happen when the temperature is quite low, whereas if its value is much high ($T > 900$ °C for the CaO, and $T > 300$ °C for the MgO) the reverse reaction, called calcination, could occur.

In Chapter 2, it was said that the presence of water in the materials used as sorbents in the reaction is the necessary condition to speed up the carbonation. Considering the aqueous method, the metal oxides (CaO or MgO) present in the sample react with water generating the relative hydroxides ($Ca(OH)_2$ or $Mg(OH)_2$), and the operations, in the presence of Ca and Mg respectively) can be schematized in this way:

Dissolution of carbon dioxide in water:

$$CO_2 + H_2O \rightarrow H_2CO_3 \rightarrow H^+ + HCO_3^- \rightarrow 2H^+ + CO_3^{2-}$$

Dissolution of Calcium oxide in water:

$$CaO + H_2O \rightarrow Ca(OH)_2$$

$$Ca(OH)_2 \rightarrow Ca^{2+} + 2OH^-$$

Calcium carbonate precipitation:

$$CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3$$

Dissolution of Magnesium oxide in water:

$$MgO + \ H_2O \rightarrow Ca(OH)_2$$

$$Mg(OH)_2 \rightarrow Mg^{2+} + 2OH^-$$

Magnesium carbonate precipitation:

$$CO_3^{2-} + Mg^{2+} \rightarrow MgCO_3$$

Thank this schematization, it's possible to understand that the whole carbonation process can be divided into three steps: dissolution of CO_2 into water, dissolution of metal oxides into the water, and carbonate precipitation.

4.1.1 Carbon dioxide dissolution in water

The first step of carbonation consists of the dissolution of CO_2 in water. To study this phenomenon is necessary to know the characteristics of carbon dioxide and how it really can diffuse into the solution. Carbon dioxide can be present as solid, liquid, gas, or in a particular situation of equilibrium, called triple point, in which all three phases can coexist. Thanks to Figure 5, which presents the P/T graph of CO_2, it's possible to note that its triple point takes place when the temperature is –56,6°C and pressure corresponds to 5,2 bar, while its critical point occurs when T = 31 °C and P = 73,8 bar, indicating that over these values the substance cannot exist as a gas-liquid mixture.

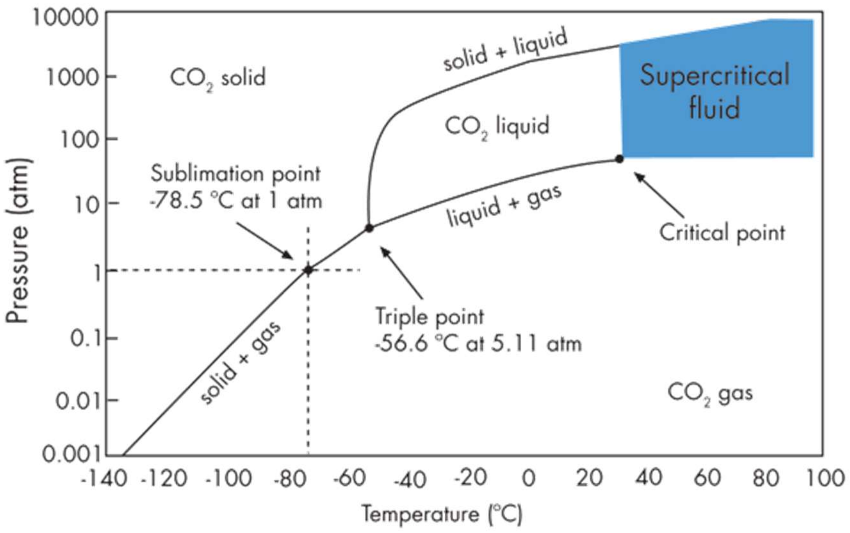


Figure 5: P/T diagram for carbon dioxide (CO_2).

Regarding carbon dioxide solubility, it depends on the temperature according to Henry's law described below:

$$C_a = H \cdot p$$

Where C_a is the concentration of the specie in the solution, p is the partial pressure that this specie exerts in the gas phase under equilibrium conditions, and, finally, H is Henry's constant.

H can be expressed as a function of the temperature thanks to the Van't Hoff equation:

$$\frac{d \ln H}{d\left(\frac{1}{T}\right)} = \frac{-\Delta_{sol}H}{R}$$

Where R ($m^3 \cdot Pa\ mol^{-1}\ K\text{-}^1$) is the universal gas constant, and $\Delta_{sol}H$ is the enthalpy. This formulation is valid only in the temperature ranges in which the enthalpy remains constant and, for this reason, with the Krichevsky – Illinskaya, described below, equation H is adjusted to high-pressure values.

$$lnH = lnH^* - \frac{A}{RT}\left(x_j^2 - 1\right) - \frac{v_m^\infty (p - p^*)}{RT}$$

Where H and H* are, respectively, Henry's constants referred to reference pressure p* and system pressure p, A represents the Margules constant, x_j is the molar fraction of water, and, finally, v_m^∞ describes the molar volume of solute at infinite dilution.

Another possible equation that expresses Henry's constant as a function of the temperature is the Morel and Hering equation (1993):

$$H_T = H_{298K} \cdot \exp\,[C \cdot (\frac{1}{T} - \frac{1}{298})$$

In this case, H_T is Henry's parameter function of the temperature, whereas H_{298K} is the one referred to as a temperature value of 298 K (25 °C). Finally, C is the constant 2400 K for CO_2, and T is the temperature expressed in Kelvin (K).

However, thanks to Henry's low, it's possible to understand that at a constant temperature, the solubility of a specie, described as its ability to diffuse in an aqueous solution, is directly proportional to the pressure of the species, in the gaseous phase, over the solution. Greater the H of a substance, the better its solubility. In the case of CO_2, at T = 25 °C, the H is equal to 3,1 M/atm (mol/L·atm). According to the low, it's possible to have a better diffusion of CO_2 in the water by increasing the partial pressure.

When the gaseous form of carbon dioxide dissolves into the H_2O, it generates the carbonic acid (H_2CO_3) which dissociates, producing hydrons (H^+) and hydrogen carbonate ion (HCO_3^-), with the final formation of the carbonate ion (CO_3^{2-}). This system causes a decrease in the pH of the solution from the neutral value of 7 to the acid one of 5,5.

The occurring reactions are the following:

$$CO_{2(g)} + H_2O \rightarrow H_2CO_{3(aq)}$$

$$H_2CO_3 \rightarrow H^+ + HCO_3^- \rightarrow 2H^+ + CO_3^{2-}$$

So, the effective equilibrium reaction occurring in the solution is found:

$$CO_{2(g)} + H_2O \rightarrow 2H^+ + CO_3^{2-}$$

4.1.2 Dissolution of metal oxides into the water

The second step of the carbonation happens when the reactive elements contained inside the sorbent materials, that is the metal oxides, enter in contact with the water. Considering Calcium oxide, which guarantees faster carbonation occurring, the reaction that takes place is the following:

$$CaO + H_2O \rightarrow Ca(OH)_2 \rightarrow Ca^{2+} + 2OH^-$$

In this way, Calcium hydroxides ($Ca(OH)_2$) are generated. Then these products dissociate, causing the release of Calcium ions (Ca^{2+}), necessary for the third phase of the process. The efficiency of this second stage can be increased by treating the material before exposing it to carbonation. The pre-treatments that can be implemented are the granulometric reduction and separation of inert and non-reactive elements. In the first case, smaller grain sizes allow having a bigger surface area, so a better contact between carbon dioxide and sorbent while the removal of the elements that are non-useful to the process permits to increase in the concentration of the reactive components.

4.1.3 Precipitation of carbonates

The last part of accelerated carbonation involves the formation of carbonates as end products. Calcium ions (Ca^{2+}), released from the dissolution of Calcium oxides in water, react with carbonate ions (CO_3^{2-}), generated by the diffusion of CO_2 in H_2O. The reaction is:

$$CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3$$

Thus, now it's possible to understand how the carbonation reaction takes place and how CO2 can be converted into carbonates and fixed in alkaline wastes or natural rocks, making possible its concentration reduction into the atmosphere.

5. OPERATIVE METHOD OF PROCESS

As said in the previous chapter, some pre-treatments have to be done on the material before exposing it to an accelerated carbonation process. The possible actions that can be implemented to prepare the sample include granulometric reduction, remotion of inert and non-reactive elements, and thermal treatments. All of these allow us to reach a higher efficiency in terms of CO_2 absorbed ($g_{CO2\_ABSORBED}/kg_{SORBENT}$) by decreasing the grains size to improve the contact between gas and a solid fraction, increasing the reactive elements concentration in the samples and also eliminating the crystallization water from the material to increase the porous system allowing a better diffusion of CO_2 inside it.

Once the samples have been prepared, the choice of the quantity of water to add is needed to reach the wanted L/S ratio to analyze before exposing the sorbents to the carbonation reaction. If an indirect method is implemented, also the extraction of the reactive base from the mineral has to be carried out by using acids or other solvents, whereas, in the case of direct procedure, the samples can be directly put in contact with the gaseous carbon dioxide.

A general scheme of the carbonation procedure can be seen in Figure 6.

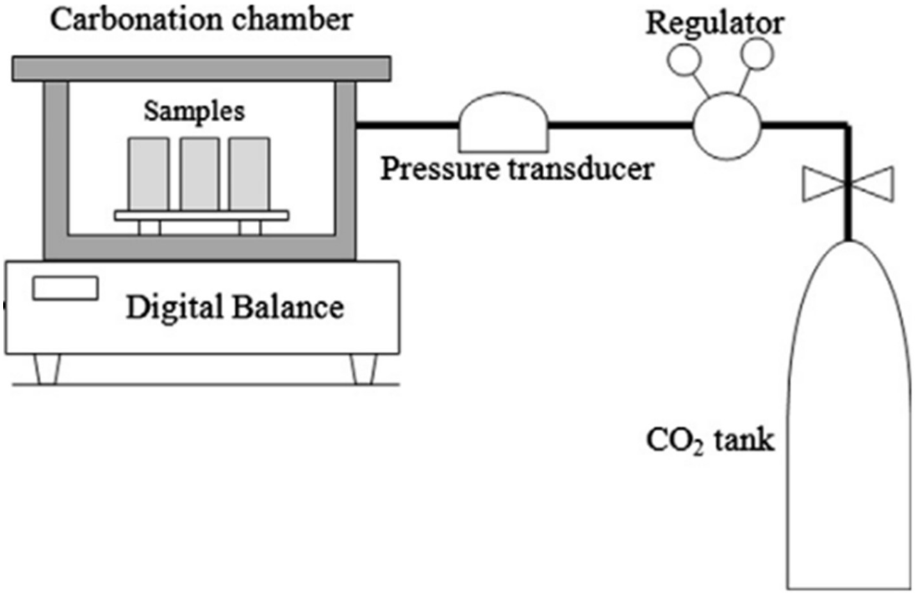


Figure 6: General scheme of the accelerated carbonation process (Rostami et al. 2011).

The samples are put inside the carbonation chamber, an incubator in which the reaction takes place for a time chosen as the operative condition at the beginning of the analysis. Generally, the incubators can be opened or closed; in the first case, represented by the previous figure, the reactor is connected to the CO_2 tank by a small tube in which a regulator is present, allowing the adjustment of the concentration and flow of the gas, while in the second one, the gas exchange in the chamber is avoided and the gas parameters have to be chosen before closing the valves. However, both the set-up configurations are characterized by pressure and temperature sensors that permit variation in their value inside the camber, according to the decisions made and the scope of the experiment.

A particular case of a carbonation chamber is shown in Figure 7, in which a rotating reactor is presented. In this situation, the process is the same as the previous one. Hence there are a gas cylinder and a vacuum pump linked to the incubator through the device that makes the setting of the operative parameters possible. But, differently from the first process, this one also exhibits a rotation movement of the samples during the carbonation reaction. In the figure, this is explained through the two different views of the reactor: the front view (c) indicates the circular movement of the grains inside the chamber, while the lateral one (b) explains that this operation is induced by an electric motor placed behind the reactor.

This is a method to enhance the sequestration of carbon dioxide because the continued rotation of the material grains during the reaction duration helps to a better diffusion of the CO_2 inside the sample and so allows to have higher final results in terms of CO_2 absorption.

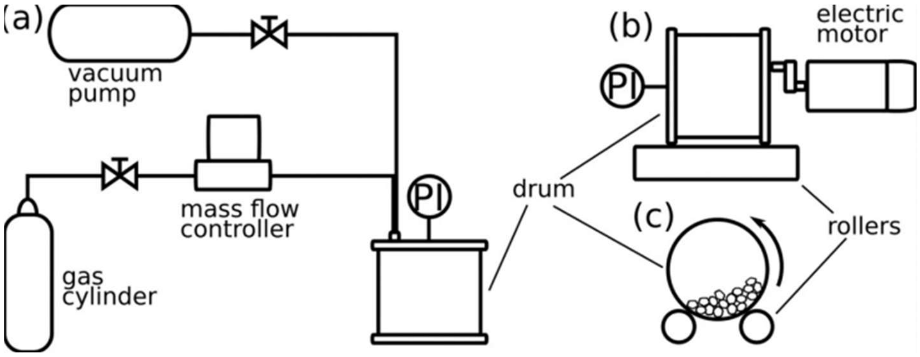


Figure 7: Use of a rotational carbonation chamber in the accelerated carbonation process (Dos Reis et al., 2020).

6. CO_2 ABSORPTION CALCULATION METHODS

The quantity of carbon dioxide that can be absorbed by a material through the carbonation reaction can be determined by many different theoretical models, while experimental analyses are used to find the real obtained results and to compare them with the theoretical ones to know if the expected values are higher or lower than the measured ones.

6.1 Theoretical Absorption

6.1.1 Steinour model

In theory, the CO_2 uptake can be determined based on the chemical compositions of the material used as a sorbent in the carbonation. Generally, in literature, the most applied theoretical model is based on the Steinour equation (Steinour, 1959) described below:

$$CO_2(\%, max) = 0{,}785 \cdot (CaO - 0{,}7SO_3) + 1{,}091MgO + 1{,}420Na_2O + 0{,}935K_2O$$

CO_2 (%, max) represents the ideal maximum quantity, reported as a percentage value, of carbon dioxide that can be absorbed by a material, according to its chemical composition and, so, to its oxides content (CaO, SO_3, MgO, Na_2O, and K_2O) included in the formula with their respective mass fractions. This formula was determined to measure the CO_2 absorption in cement-based materials, but it's also used for other materials, such as alkaline wastes characterized by a great number of oxides inside them, as explained in many papers. In particular, it has been assessed that the Steinour approach can also be used for fly ashes (Yuan et al., 2022), municipal solid waste incinerator bottom ashes (MSWI-BA) (Chang et al., 2015) (Nam et al., 2015), petroleum coke fly ashes (Pei et al., 2018), biomass bottom ashes, biomass fly ashes, refuse-derived fuel fly ashes, and stainless-steel slags (Schnabel et al., 2021).

Many papers use the original Steinour equation (Nam et al., 2015) (Pei et al., 2018) (Yi et al., 2020) (Xuan et al., 2016), while in other studies, the theoretical absorption of CO_2 is measured by using modified versions of the Steinour formula to have simulated results more comparable with the ones experimentally obtained.

In particular, (Xuan et al., 2019) calculated the carbon dioxide uptake using RCAs (Recycled Concrete Aggregates) as sorbent through the Steinour equation modified by Huntzinger (Huntzinger et al., 2019):

$$\begin{aligned}\%Theory_{CO_2} = 0{,}785 \cdot \left(X_{CaO} - 0{,}56X_{CaCO_3} - 0{,}70X_{SO_3}\right) + 1{,}09X_{MgO} + 0{,}71X_{Na_2O} \\ + 0{,}47X_{K_2O}\end{aligned}$$

Where X, in this case, represents the mass ratio of the relative individual oxides. Here, differently from the original formulation, the factor multiplied by Na_2O and K_2O is halved, and the presence of $CaCO_3$ has been added.

The same formula (Huntzinger et al., 2009; Steinour, 1959) has been used by (Chang et al., 2015) for MSWI-Bas, even if referred only to Calcium oxides (CaO), Calcium carbonates ($CaCO_3$), and Sulfur trioxide (SO_3):

$$\%Theory_{CO_2} = \frac{44}{56}(CaO - \frac{56}{100}CaCO_3 - \frac{56}{80}SO_3)$$

Where CaO and SO_3 are the weight fractions of the respective substances measured in the sorbent by XRF (X-Ray Fluorescence) analysis, and CaCO3 is the weight fraction of the Calcium carbonates analyzed with TGA (Thermogravimetric Analysis).

A similar equation has been used by (Yuan et al., 2022) for the calculation of the CO_2 sequestrated by fly ashes, but in this case, some parameters have been changed, and Magnesium oxides (MgO) have been considered instead of Calcium carbonates. The equation is the following:

$$\%Theory_{CO_2} = [\frac{44}{56}\left(CaO - \frac{56}{100}CaCO_3\right) + \frac{44}{40}MgO] \cdot 1000$$

Also (Schnabel et al., 2021) considered the Steinour equation modified by Huntzinger et al., considering that the hydroxides of Ca, Mg, Na, and K are reactive elements for the carbonation while the corresponding carbonates, Sulfur, and chlorine compounds hinder the process. The formula used in the study is based on the elemental composition and carbonate content found thanks to XRF and TGA and looks like this:

$$\%Theory_{CO_2} = 0{,}785 \cdot \left(X_{Cao} - 0{,}56X_{CaCo_3} - 0{,}70X_{SO_3}\right) + 1{,}09X_{Mgo} + 0{,}71X_{Na_2O} + 0{,}468 \cdot (X_{K_2O} - 0{,}632XKCl)$$

The same equation has also been implemented by (Soares et al., 2022) for the measure of carbon dioxide sequestrated by bottom ashes as sorbents in the reaction of accelerated carbonation.

6.1.2 Response surface methodology (RSM)

This is a semi-empirical method implemented by (Kaliyavaradhan et al., 2020) to optimize the theoretical CO_2 absorption evaluation, considering, differently from the previous approach, many different operative parameters (liquid-solid ratio, duration of the process) and their interactions. The relationship between these variables has been evaluated through a regression procedure based on the following 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 is the predicted response, β_0 the intercept, β_i and β_j Represent the linear effect coefficients, β_{ii} and β_{jj} the quadratic effect coefficients, β_{ij} the interaction effect coefficient, X_i and X_j the independent variables, and, finally, ε is the residual.

RSM has been used to determine the theoretical CO_2 absorbed by CSW (Concrete Slurry Waste), taking into consideration the decided ranges used for the operative parameters. In this particular case, the maximum carbon dioxide uptake has been found using the water-solid ratio (w/s) in a range between 0,1 and 0,7, and the duration time (t) of the reaction between 1 h and 168 h.

The equation used to practically predict the CO_2 uptake is this:

$$\begin{aligned}\%Theory_{CO_2} = {} & 5{,}607 + 90{,}906\left(\frac{w}{s}\right) + 0{,}193(t) - 0{,}0687\left(\frac{w}{s}\cdot t\right) - 257{,}917\left(\frac{w}{s}\right)^2 \\ & - 0{,}00206(t)^2 - 0{,}00172\left(\frac{w}{s}\right)^2\cdot(t) + 0{,}0006\left(\frac{w}{s}\right)^2\cdot(t) + 195{,}039\left(\frac{w}{s}\right)^3 \\ & + 0{,}0000063(t)^3\end{aligned}$$

The efficiency of this approach has also been validated by evaluating the absolute relative deviation (ARD %) as a measure of predictability, adopting this formula:

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

Thanks to this equation, ARD values equal to 3,0133 % and 0,799% have been found for the two tested samples, indicating in both cases a result lower than 10 % and confirming the high efficiency of this method in determining the effects of parameters on CO_2 uptake of the analyzed material.

6.2 Effective Absorption

Having said that, in the previous chapter, the theoretical CO_2 absorption has to be compared with the result effectively obtained with the accelerated carbonation process, now it's important to cite and describe the possible test that can be used to find the real values of carbon dioxide uptake. The most used approaches in the literature studies are TGA (thermogravimetric analysis) and XRD (X-Ray diffraction).

6.2.1 Thermogravimetric analysis - TGA

The first one, TGA, represents a method useful to monitor the mass changes, in terms of percentage or absolute value, in the sample as a function of temperature and time. Hence it generates a graph representing the thermic decomposition.

An example of the TGA curve is described by (Dos Reis et al., 2020), where the results obtained with the non-carbonated material (recycled sand) have been compared with the ones determined with carbonated samples of the same type of sorbent, as shown in Figure 8. As previously anticipated, the following image indicates the differences existing between the virgin aggregates curve and the carbonated aggregates trends to evaluate the carbon oxide uptake. In particular, it's possible to see that all the second ones exhibit a higher mass reduction than the first one, an expected result because the carbonation products are degraded in this range of temperature. The decomposition of Calcium carbonates ($CaCO_3$) happens when the temperature values are between 550 °C and 1000 °C, the zone in which the differences between the weight losses are the highest. Thus, thanks to the TG mass changing, CO_2 absorption can be calculated by considering the mass before and after carbonation.

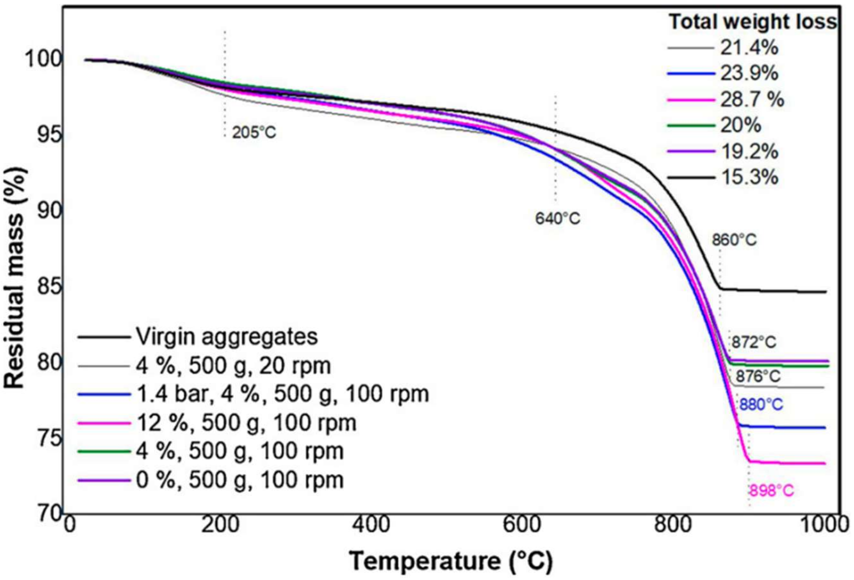


Figure 8: TGA curves of non-carbonated and carbonated materials (Dos Reis et al., 2020)

6.2.1 X-Ray diffraction – XRD

The XRD method is adopted to identify and characterize the mineralogical structure of the analyzed materials and, so, to quantify the presence of Calcium carbonate ($CaCO_3$) crystals. The procedure consists in irradiating a sample with X-Rays and measuring the intensity and the inclination of the rays diffracted by the material to keep information about its components. Like in the process described in the previous chapter, also in this case, the analysis has to be done both on virgin and carbonated sorbents to compare them and note their differences in terms of composition.

An example of a result obtained with this test is presented in Figure 9, which reveals the changes in the peak intensities registered at 25° and 35° (2theta).

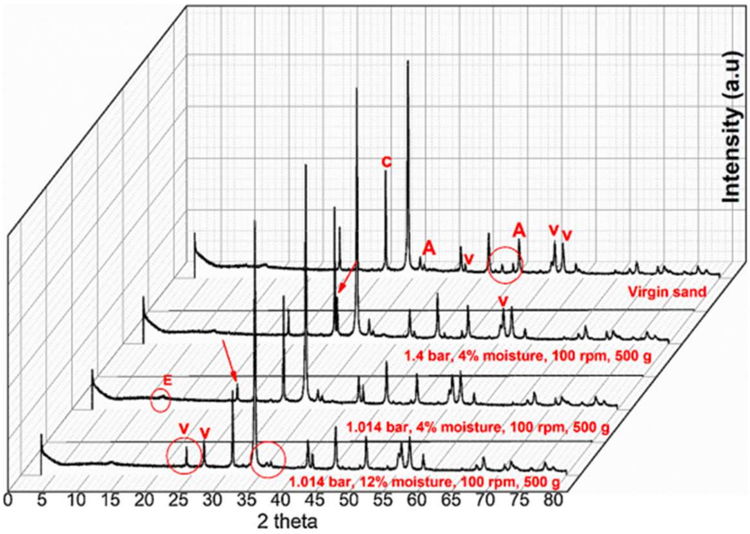


Figure 9: XRD results obtained by (Dos Reis et al., 2020)

PART II – SCIENTIFIC PAPER

1. ACCELERATED CARBONATION

1.1 Accelerated Carbonation inside the IPCC Neutrality Carbon Policy

Climate change is the great and main problem that characterizes and will characterize the 21st century. As can be seen in Figure 1, according to the latest report published by the " Intergovernmental Panel on Climate Change " - IPCC in March 2023, the average temperature of the Earth's surface in the decade 2011-2020 was 1.1°C higher than the value for the period between 1850 and 1900, and the main cause is due to anthropogenic activities and related emissions of greenhouse gases, especially carbon dioxide (CO_2) and methane (CH_4), into the atmosphere.

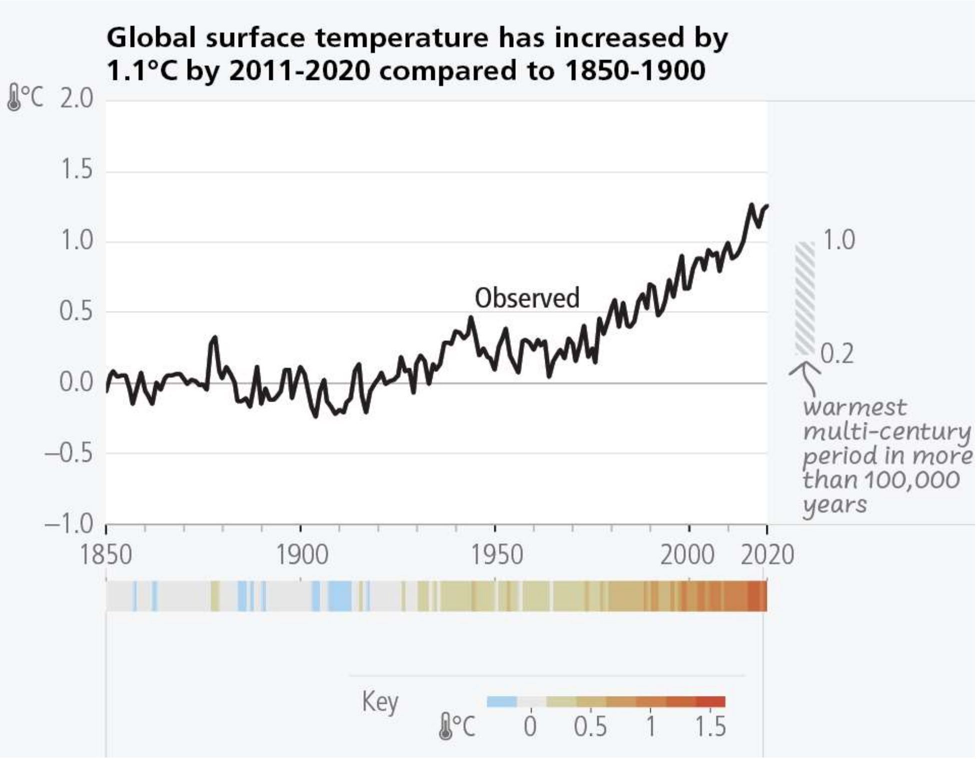


Figure 1: Trend of the Earth's average surface temperature over the period 1850 – 2020 2020 (IPCC – Climate Change 2022 – Mitigation of Climate Change).

It was also noted that the CO_2 concentration measured in the atmosphere in 2019 was 410 ppm, higher than any other value measured in the last two million years.

In fact, the period between 2010 and 2019 was characterized by the highest amount of greenhouse gases ever emitted into the atmosphere, although the percentage growth was 1.3%/year, which was less than that one measured in the previous decade, 2000 - 2009. As earlier mentioned, Climate Change is due to the emissions of CO_2 and other gases that have been produced, in ever-increasing quantities, by anthropogenic activities such as industrial processes, energy production processes based on fossil fuels usage, and transport for over a century. It's possible, therefore, to understand how human behavior has been fundamental to the warming of the atmosphere, land, and especially the warming of the oceans, which accounts for 91% of the temperature increase in the climate (IPCC, 2023).

The effects of climate change are important and numerous and, for this reason, must be carefully studied. In particular, as far as the sea is concerned, there has been a significant increase in the average sea level of 0.20 m between 1901 and 2018, with a growth rate that, from values of 1.3 mm/year and 1.9 mm/year corresponding to the periods 1901 - 1971 and 1971 - 2006 respectively, rose to 3.7 mm/year in the years between 2006 and 2018. In addition, a large ice retreat in the Arctic Sea was also observed in the period 2010 - 2019. As can be seen in Figure 2, however, numerous issues are most attributable to human action.

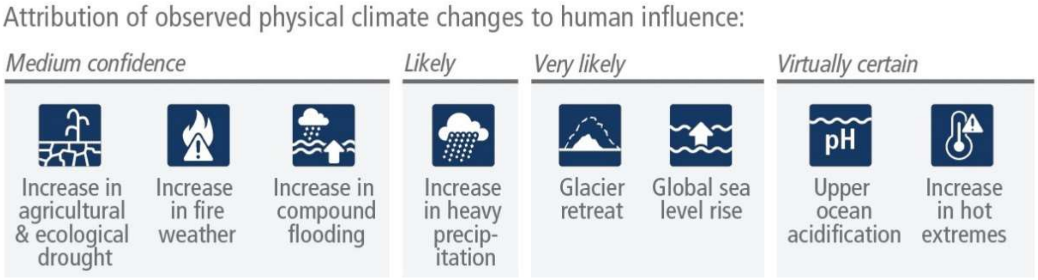


Figure 2: List of climate changes that are potentially or certainly caused by anthropogenic activities 2020 (IPCC – Climate Change 2022 – Mitigation of Climate Change).

This is important to understand how man-made CO_2 emissions must be rapidly reduced to limit the negative effects on the environment and people's daily lives as much as possible. For this reason, in 2015, the member states of the United Nations Framework Convention on Climate Change - UNFCCC entered into an international treaty called the Paris Agreement regarding the mitigation and adaptation actions necessary to tackle climate change and promote sustainable development.

Through this agreement, the goal was set to keep the increase in the Earth's average temperature well below 2°C compared to the pre-industrial period, continuing efforts not to exceed a 1.5°C increase and, thus, avoid serious and, perhaps, irreversible consequences. However, according to the latest IPCC report, CO_2 emissions must be reduced by more than 50 % to achieve this goal, while the amount of carbon dioxide emitted into the atmosphere must be reduced by about 30% to stay within 2°C. It is, therefore, essential to increase efforts and adopt better environmental policies to achieve carbon neutrality because, in this way, emissions will continue to rise, causing an increase of 2.2°C to 3.5°C by 2100. Carbon neutrality is the condition in which carbon dioxide emissions into the atmosphere and its removal or absorption occur in equal amounts, a condition that must be achieved by 2050 to meet the 1.5°C target.

Thus, it has been noted how important it is to invest in processes that can reduce the concentration of CO_2 in the atmosphere, either by reducing emissions or by absorbing certain quantities. In this context, processes for the capture and storage (CCS) or utilization (CCU) of carbon dioxide are crucial. However, they can be considered removal technologies (CDR) only if they are applied to biogenic CO_2, i.e. CO_2 released naturally by natural cycles or directly captured from the air. Thanks to these technologies, it is possible to store CO_2 in geological reservoirs or reusable by-products, thus stimulating the development of a circular economy related to carbon utilization.

As can be seen in Figure 3, different techniques can be used to capture and use or store carbon dioxide, each one based on a different emission source and methodology used to capture CO_2.

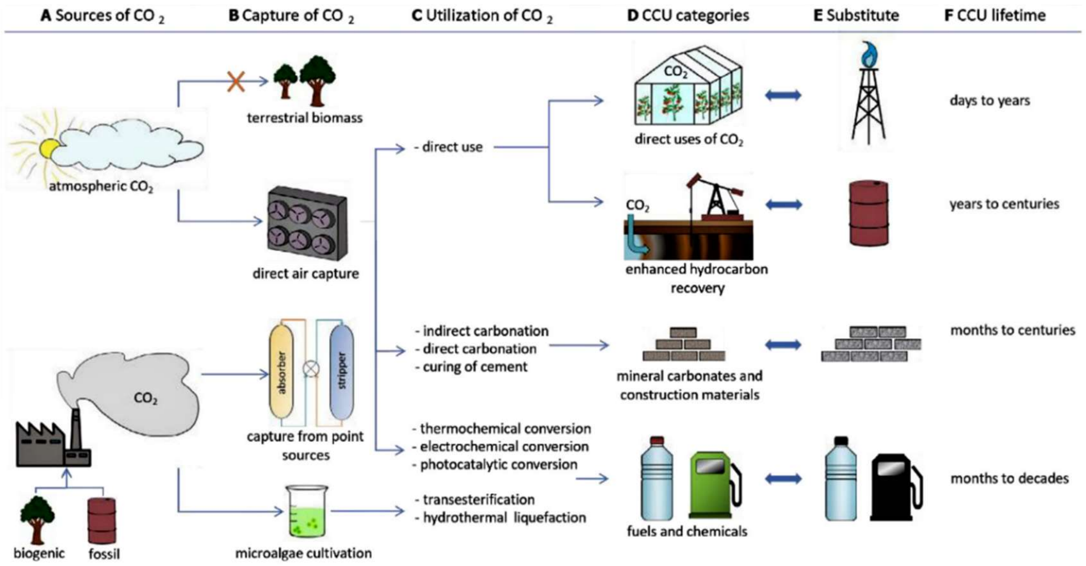


Figure 3: Description of possible carbon dioxide capture and utilization techniques (Guo et al. 2022).

Nevertheless, as mentioned earlier, storing and utilizing carbon dioxide is only considered a CDR when DACCS techniques are being considered, so when absorbing it from the air: a process, however, whose cost is very high and whose efficiency is still particularly low.

A more effective solution is to capture CO_2 from the chimneys of energy plants or industries where it is produced in concentrations of around 10%. As shown in Figure 4, several CCS techniques can be used, including the reaction under investigation in this study: accelerated carbonation.

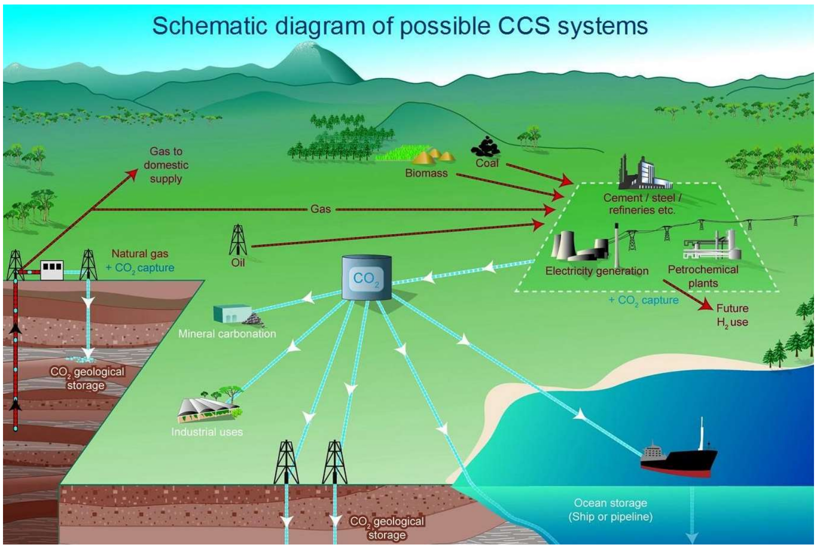


Figure 4: Outline of possible carbon dioxide capture and storage (CCS) techniques (IPCC - carbon dioxide Capture and Storage 2004).

Accelerated carbonation, whose detailed workings will be discussed in later chapters, is a chemical reaction whereby waste materials such as steel slag, fly ash, dust generated by incineration plants, or cement and clay waste are used to absorb CO_2. Carbon dioxide reacts with the metal oxides present in the material, producing carbonates and, thus, precipitating to a form that can remain stable for hundreds of years. Accelerated carbonation, therefore, is a process that, although still in the development phase, is very important in stimulating the industrial transition towards the goal of carbon neutrality, as it reduces emissions produced by anthropogenic processes that cannot be eliminated.

This process can be direct when the material is carbonated through a single step or indirect if metal oxides are extracted from the material before it is carbonated. In this paper, however, only direct carbonation was considered since the samples used have been directly exposed to carbon dioxide without intermediate steps.

1.2 The Potential of the Critical Review

As already mentioned in the previous chapter, the subject of this thesis is accelerated carbonation. First of all, a critical analysis was carried out to well understand the subject, the different techniques that can be adopted, and the amount of CO_2 that can be absorbed by different materials. This analysis has been performed starting from a previous study (Danelon, 2022), which was then revised and expanded.

The process was carried out based on the methodology described in 'The PRISMA 2020 statement: an updated guideline for reporting systematic reviews' (Page et al., 2021). In this way, among the articles found in the literature, it was possible to rigorously select only those that were truly inherent to the purpose of the thesis. The adopted procedure consists of four steps: identification, selection, eligibility, and, finally, inclusion. First, articles that could have useful information for the study were identified by searching the Scopus database for the following words: ("accelerated" AND "carbonation") AND ("aggregates" OR "fly ashes") AND ("CO_2"). In doing so, only articles containing both accelerated carbonation and CO_2 as keywords and one between aggregates and fly ashes have been searched. In the previous study, on which the revision and extension of the critical analysis are based, 194 articles were found in this way, 6 of which were removed before the next selection phase as they could not be downloaded, being duplicates or written in the Korean language.

In addition, 17 papers were found on other databases such as 'ScienceDirect' and 'ResearchGate'.

Subsequently, a further 146 articles with different scopes to the present analysis concerning the physical, chemical, and mechanical properties of materials were removed through the selection phase. In this way, 42 articles from Scopus and 17 from other sites were selected. Of these 42, due to the passage of eligibility, a further 20 were removed as not presenting a quantitative and precise analysis of the amount of CO_2 absorbed, while from the 17 ones, five were removed because they were already cited from other previously analyzed articles or they did not contain the formulas necessary to convert the carbonates produced into grams of carbon dioxide sequestered per kilogram of material used.

Thus, a total of 34 scientific articles were included and analyzed. However, since this study was concluded in March 2022, it was decided to first review the entire critical analysis and then extend it to the publication year 2023.

Therefore, by following the same procedure, through the various steps described above, it has been possible to add four other articles published between 2022 and 2023, which allowed us to reach a total number of 38 papers that can be used in order to have a thorough understanding of the different conditions used for carbonation and the different results found. Figure 5 shows the diagram of the Prism methodology on which this paper was based.

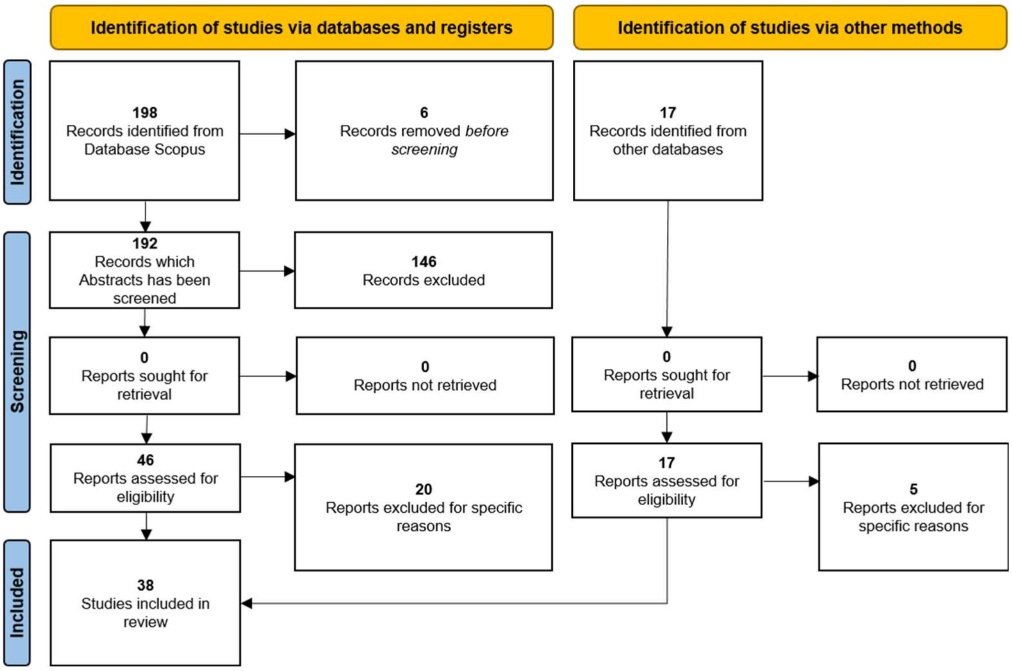


Figure 5: 2020 Prism Diagram used to determine scientific articles for use in critical analysis (Page et al. 2021).

Then, the papers included in the critical analysis have been analyzed using different parameters to compare the quantities of CO_2 sequestered under different operating conditions by each different material. The classes used to study these papers are: type of reactor used in the test, type of material analyzed, experimental design adopted, variables tested, operating conditions of the process, experimental scale, method used to calculate the carbon dioxide absorbed, and CO_2 sequestration results obtained.

Concerning the reactor used in the carbonation test, two types of it have been identified: the open reactor and the closed reactor. The first of these is characterized by the fact that the flow of gas is controlled by a flow meter which allows its transport inside the chamber where the reaction takes place to be regulated, while in the second, the exchange of gas between the cylinder from which it arrives and the combustion chamber is prevented.

In the latter case, before placing the samples of material to be tested inside the incubator, one proceeds by processing the chamber with a vacuum pump and injecting the gas into it, allowing the CO_2 parameters, such as pressure and concentration, to be varied only before closing the valves that allow the gas to pass through. However, both configurations are equipped with sensors to regulate the temperature and pressure inside the chamber where carbonation takes place.

The materials undergoing accelerated carbonation have been divided into ten categories.

The dominant class, for which the largest number of analyses was conducted, is recycled cement aggregates (RCA), which include compounds derived from construction cement waste, then crushed in recycling plants, or obtained from the demolition of concrete pipes or other such compounds. Ten studies were performed using recycled cement aggregates subjected to particle size reduction, of which two carbonation tests have been conducted by using the RCAs in monolithic form, using blocks formed from water, cement, and RCA used as aggregates. The second most widely used category concerns fly ashes: ashes coming from incineration plants, residues obtained from atmospheric pollution control, or ashes generated by energy production processes using coal and by other combustion processes.

Again, the cases in which this type of dust was used in granular form were differentiated from the processes that, instead, tested them in monolithic form. In the former case, nine studies were identified, while in the latter two. Concerning Bottom Ashes from municipal solid waste incinerators, six different studies were identified in which, in all cases, the material was analyzed in granular form.

This was followed by steel or iron slag, in granular form, from the electric arc furnace (EAF), blast furnace (BF), and basic oxygen furnace (BOF), of which six different analyses were studied.

Finally, five studies were examined concerning concrete in monolithic form, formed from the mixture of water, cement, and other aggregates, two concerning natural aggregates whose composition was not precisely defined, and only one study respectively for natural lime mortar, used for masonry in historic buildings, and for cement mortar, which can be used for new constructions.

For each study, the relevant experimental design, the methodology adopted for carbonation, the variables characterizing the study, and the value of the process parameters at which the carbonation reaction was conducted, have been described.

These parameters are temperature (°C), pressure (bar), humidity (%), and, in the case of CO_2, the flow rate of the incoming gas stream (L/min) and the concentration of the gas (%). Numerous values were evaluated for all these factors; in particular, temperature values in the range 20 °C - 200 °C, a pressure between 0.1 bar and 100 bar, and relative humidity between 50 % and 99 % have been analyzed, although most of the studies were conducted at ambient temperature and pressure ($T = 25 \pm 5$ °C, $P = 1$ bar) and humidity of 50 %.

A distinction was also made between the cases in which the process was developed in a dry route or using an aqueous solution. In the former case, the materials were dried before being placed in the incubator, while in the latter, they were tested by adding different amounts of distilled water to determine the change in absorbed CO_2 related to the various liquid-to-solid ratio (L/kg) values. In this case, 20 wet and 18 dry-route studies have been considered.

A further fundamental characteristic to subdivide the identified papers consists in the operating conditions, which, in this case, have been categorized into four subclasses: very mild, mild, intensive, and very intensive. The first category, very mild, has been identified in the presence of ambient temperature ($T < 35$ °C), pressure value of 1 bar, and carbon dioxide concentration between 0 % and 20 %; the second, mild, was assigned to the cases where there ambient temperature ($T < 35$ °C) and pressure of 1 bar was considered, but the CO_2 concentration ranged between 20 % and 100 %; The third, intensive, is for values of temperatures and pressures respectively equal to 35 °C and 1 bar, and CO_2 concentrations lower than 20 %; finally, the fourth group, very intensive, was assigned to analyses conducted at temperatures greater than 35 °C, pressures above 1 bar and CO_2 concentrations between 20 % and 100 %.

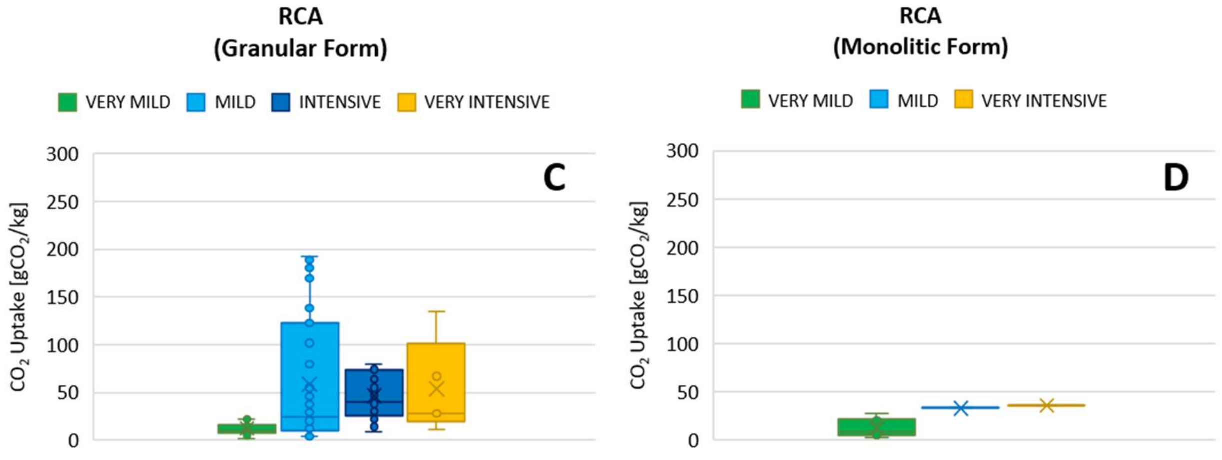
For each experiment, the scale, so its duration and, if present, the mass of the sample used, as well as the analyses carried out to characterize the material and to determine its physicochemical characteristics, have been indicated.

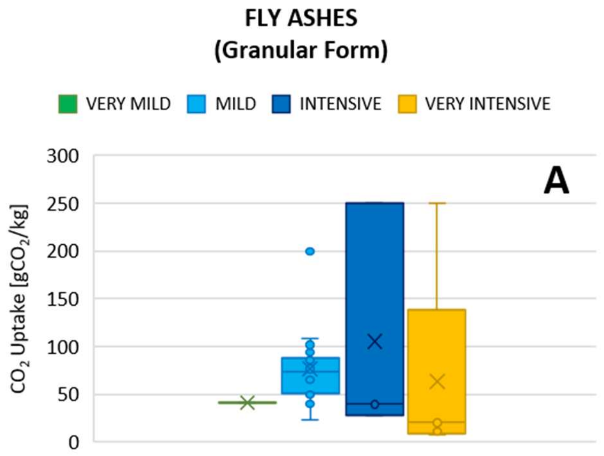
The different tests identified are: total carbon content (TC), water content, metal content, loss of ignition (LOI), X-ray diffraction (XRD), leaching test, thermogravimetry (TG), differential thermal analysis (DTA), acid/base neutralization capacity test (ANC/BNC), X-Ray fluorescence (XRF), scanning electric microscope (SEM) test, sieving, water absorption analysis, specific surface area study (SSA), pH measurement and electrical conductivity analysis. Finally, before reporting the results on the amounts of CO_2 sequestered ($g_{CO2}/kg_{sorbent}$) through each specific carbonation process, how these theoretical and actual results could be calculated has been described.

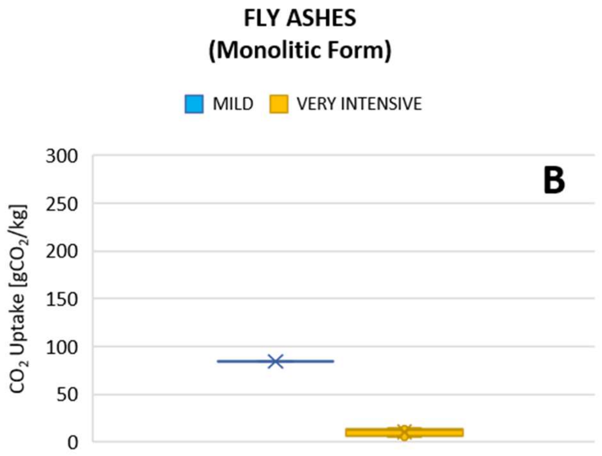
The methods used are: calcimetry analysis, thermogravimetric analysis (TG) together with differential thermal analysis (DTA) or differential thermogravimetric analysis (DTG), the method based on the mass curve or mass gain of the material, thermal decomposition, determination of inorganic carbon and use of the ideal gas law.

1.2.1 Results

Figure 6 shows the results for the amount of CO_2 absorbed by each type of material used concerning the operating condition (very mild, mild, intensive, and very intensive) adopted. However, it must be noted that in many analyses, not all variables considered in the critical analysis have been found in the papers. In particular, 20/38 articles did not report the mass (g) of the samples tested, 9/38 did not describe the pressure value (bar) used, 2/38 did not write the temperature (°C) considered, 2/38 did not describe the duration of the process, 20/38 did not state the relative humidity (%), 3/38 did not state the particle size of the material, 30/38 did not state the CO_2 flow value (L/min) and, finally, 6/38 did not define the CO_2 concentration. In the cases where pressure and temperature values were missing, environmental values of 1 bar and 25 °C have been respectively assumed.




FLY ASHES
(Granular Form)
VERY MILD
MILD
INTENSIVE
VERY INTENSIVE
A
CO$_2$ Uptake [gCO$_2$/kg]
300
250
200
150
100
50
0


FLY ASHES
(Monolitic Form)
MILD
VERY INTENSIVE
B
CO$_2$ Uptake [gCO$_2$/kg]
300
250
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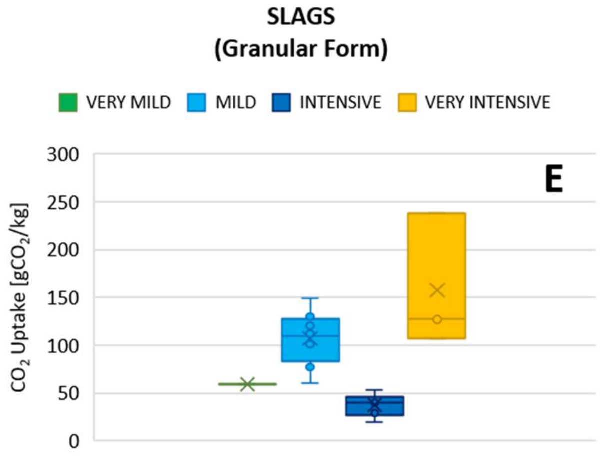
SLAGS
(Granular Form)
VERY MILD
MILD
INTENSIVE
VERY INTENSIVE
E
CO$_2$ Uptake [gCO$_2$/kg]
300
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200
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100
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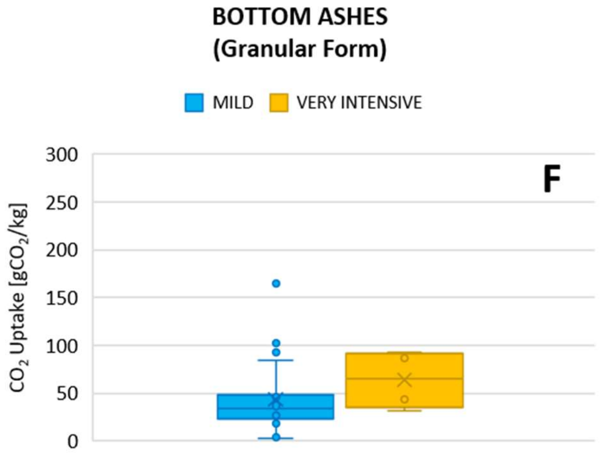
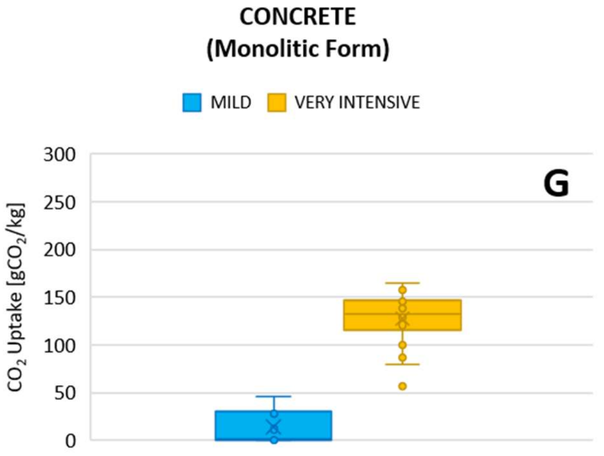

BOTTOM ASHES
(Granular Form)
MILD
VERY INTENSIVE
F
CO$_2$ Uptake [gCO$_2$/kg]
300
250
200
150
100
50
0


CONCRETE
(Monolitic Form)
MILD
VERY INTENSIVE
G
CO$_2$ Uptake [gCO$_2$/kg]
300
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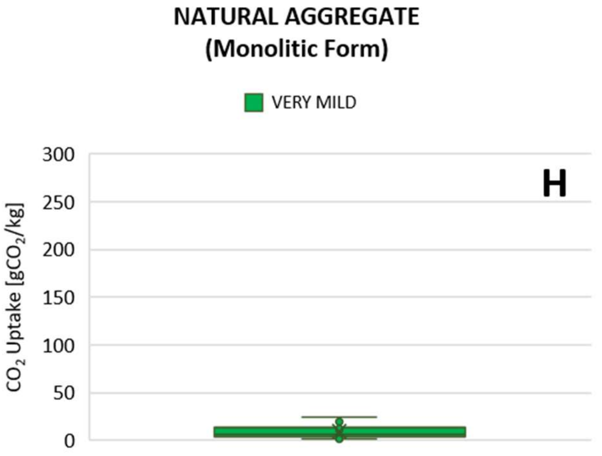
NATURAL AGGREGATE
(Monolitic Form)
VERY MILD
H
CO$_2$ Uptake [gCO$_2$/kg]
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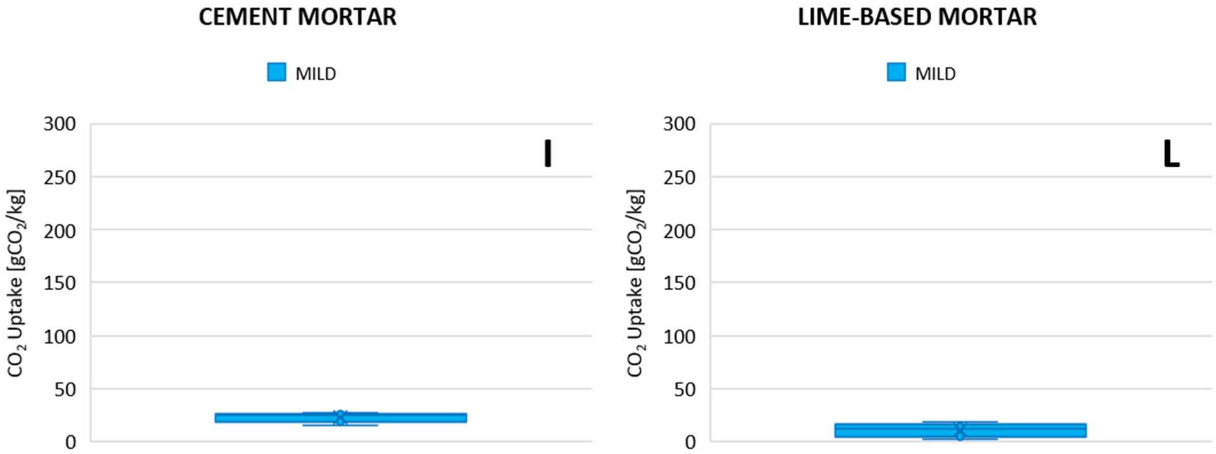


Figure 6: Results for CO_2 sequestered by (A) fly ashes in granular form, (B) fly ashes in monolithic form, (C) RCA in granular form, (D) RCA in monolithic form, (E) slag in granular form, (F) bottom ashes in granular form, (G) concrete in monolithic form, (H) natural aggregates in monolithic form, (I) cement mortar, and (L) lime mortar.

In particular, it can be observed that the maximum amount of CO_2 absorbed is 250 $g_{CO2}/kg_{dry\_sorbent}$, obtained using fly ashes in the granular form under operating conditions that fall into the intensive and very intensive class, although, in the latter case, the value is far removed from the others found results. With this type of material, without considering the very mild class where only one value can be relied on, the results found are good in all the categories: in the mild condition, 20 data have been evaluated in the range of 23.42 - 200 $g_{CO2}/kg_{dry\_sorbent}$, in the intensive only three values between 27.5 - 200 $g_{CO2}/kg_{dry\_sorbent}$ and in the very intensive five values of which four values between 7.66 - 27.05 $g_{CO2}/kg_{dry\_sorbent}$ and one, as mentioned above, of 200 $g_{CO2}/kg_{dry\_sorbent}$.

On the other hand, looking at the results obtained again with the fly ashes, but in monolithic form, it can be seen that in the mild condition, they are in line with what was observed before, while in the very intensive condition, they are much lower than before.

The material on which the most data are available is represented by the recycled cement aggregates (RCAs) in granular form, in which 13 results have been found in the very mild class with an absorbed CO_2 in the range of 2.08 - 21.8 $g_{CO2}/kg_{dry\_sorbent}$, 83 in the mild between 3.4 - 192.7 $g_{CO2}/kg_{dry\_sorbent}$, 38 in the intensive between 9 - 79 $g_{CO2}/kg_{dry\_sorbent}$ and, finally, 5 in the very intensive between 11 - 135 $g_{CO2}/kg_{dry\_sorbent}$. Again, although there is little data to confirm this, the results obtained using the same material in monolithic form appear similar in the very mild condition to those obtained with granular form, while in the mild and very intensive categories, they are less than the previous ones.

This may be because a finer grain size allows greater exposure of the oxides within the sample and also better CO_2 diffusion. In the other materials (slag, bottom ashes, and concrete), it can be seen that the maximum amounts of CO_2 sequestered were obtained using very intensive conditions. Finally, for natural aggregates, cement mortar, and lime mortar, the results are very low, although it must be considered that very little data was found to refer to.

In conclusion, it can be stated that according to this critical analysis, the material that absorbs the most carbon dioxide is fly ashes. However, it should be noted that, in general, all materials are characterized by great variability in the results, given by the different conditions to which they were subjected during carbonation; among these, there is a great dependence on the duration of the process in that the longer the test lasts, the better the final results as the greater the degree of carbonation.

1.3 Method Principles

Carbonation is a natural process that occurs when carbon dioxide, present in the atmosphere, reacts with the metal oxides of alkaline metals that can be found in nature, causing the formation of silicates and, thus, the consumption of CO_2. Nevertheless, in nature, this reaction takes several years to take place as the concentration of CO_2 in the atmosphere is 0.04%, and the available alkaline materials are not so accessible that carbon dioxide can easily diffuse into them. For this reason, numerous studies and experimental analyses are being conducted in an attempt to shorten the duration of this reaction by focusing on so-called accelerated carbonation, which, as mentioned earlier, is the subject of this paper.

This process can be conducted in situ or ex-situ; in the former case, the activity is developed like geological storage, while in the latter, various processes are involved that require an additional amount of energy that can hardly be offset by the energy released by carbonation. The latter methodology is carried out by extracting the materials, transporting them to the designated location and preparing them, altering their grain size, and, if necessary, increasing their reactivity.

Therefore, the use, recycling, and possible loss of additives and catalyzing agents must also be considered, as well as the final disposal of the carbonates and by-products generated. The reaction that develops when CO_2 reacts with metal oxides is:

$$MO + CO_2 \rightarrow MCO_3 + heat$$

M represents the bivalent metal considered (Calcium, Magnesium, Potassium, or Sodium), while MO is the metal oxide used as a reactant, and MCO_3 represents the carbonates produced. The heat generated by the reaction, on the other hand, depends on the metal used and the material containing the oxides; in the case of Calcium oxide (CaO), it consists of 46% of the heat released by the reaction forming carbon dioxide (393.8 kJ/mol_{CO2}) (IPCC Special Report on Carbon dioxide Capture and Storage - 2004).

Since carbonates formed from the oxides of Sodium and Potassium are very soluble in water, most studies on carbonation are based on Magnesium and Calcium ones. The latter, however, are very reactive but do not occur in large quantities in nature. For this reason, it is preferred to use industrial wastes instead of siliceous rocks as sources of metal oxides because, unlike the rocks that are present on Earth in larger quantities but are difficult to access, the formers are certainly found in smaller quantities but represent an easily accessible, immediately available and, above all, very reactive source of oxides.

The materials most frequently used as a source of metal oxides are alkaline industrial wastes such as steel slag, fly ash, dust from incineration plants, or waste materials from the construction sector such as cement and clay waste.

These materials have a multitude of calcium oxides; in particular, there is an amount of CaO, expressed as a percentage by weight of the material in question, of 65 % in the poor from coal-fired power plants, 20 %, and 35 %, respectively in dust and fly ash from municipal solid waste incinerators, and 65 % in steel slag and cement waste (IPCC Special Report on Carbon dioxide Capture and Storage - 2004).

Thus, accelerated carbonation, in addition to being a useful technique to decrease CO_2 emissions into the atmosphere to achieve the goal of carbon neutrality, also allows for the development of the circular economy concept as it uses alkaline waste as a source of metal oxides. Moreover, this process reduces the release of heavy metals from the waste used, improving its environmental quality, and enhances the mechanical properties of cement aggregates, making them potentially reusable in the construction sector (Ndiaye et al., 2023) (Xuan et al., 2018). Given that Calcium oxides are the most prevalent species among others in alkaline waste, and knowing that Calcium carbonates are one of the most studied species in forced carbonation, the carbonation reaction can be rewritten using Calcium (Ca) as a bivalent metal:

$$CaO + CO_2 \rightarrow CaCO_3 + heat$$

The enthalpy of the reaction (ΔH) is - 181.15 kJ/mol$_{CO2}$, a negative value indicating the presence of an exothermic process, a reaction that produces heat in the form of energy that is transferred from the system to the environment.

However, since the aim of the research conducted on this reaction is to reduce its duration, the parameters that most influence the process have been identified to determine the values that allow maximum efficiency in CO_2 absorption. The most important variables are: liquid-solid ratio (L/S), test duration, temperature, pressure, CO_2 concentration, material particle size, and reactor speed.

The water content present in the material is an essential condition for carbonation; an optimal liquid-solid ratio improves the hydration and the mixing in the suspended phase of the sample, thus increasing the final adsorption efficiency. However, the diffusion of CO_2 in the sample can be decreased in case of insufficient or excessive L/S. In the former case, the amount of water is too little, while in the latter, the overabundance of liquid saturates the pores of the material, and both situations make the process inefficient (Kombathula, 2020) (Ndiaye et al., 2023). For recycled concrete aggregates, several analyses reported that increasing the water content from 0 % to 4 % by mass of the material used also the amount of CO_2 absorbed can be increased, while above 4 %, the efficiency is reduced, thus establishing an optimum value of 4 % (Dos Reis et al., 2020). These results were also confirmed by other studies that reported an optimal water content for recycled concrete aggregates of 4-5% (Ndiaye et al., 2023). For ash, on the other hand, an optimal liquid-solid ratio of 0.2-0.3 g/kg was measured (Schnabel et al., 2021).

Moreover, carbonation is very fast at the beginning of the process and then stabilizes as the reaction time increases until its completion, so when maximum CO_2 absorption within the material is reached (Chang et al., 2015).

Regarding the temperature, it's known that, as mentioned earlier, carbonation is an exothermic reaction, so, according to Le Chatelier's principle, by increasing the temperature, the reaction equilibrium is shifted to the right, and product formation is inhibited. Temperature also affects the solubility, the transport speed of the reactant, and the water vapor condition. A high temperature decreases the solubility of CO_2 and calcium oxide in the solution and, therefore, delays the reaction, but, on the other hand, allows for a rapid movement of the substances involved, accelerating the diffusion of carbon dioxide in the material, and increases the amount of water vapor in the pores of the sample, improving the movement of CO_2 within them. Studies that have been carried out to investigate how the rate of carbonation varies as a function of temperature have shown that optimal conditions generally occur when it has values in the range of 0 °C – 100 °C for different cementitious materials (Xu et al., 2022). This result is confirmed by other analyses, which, studying recycled cement aggregates, determined an increase in reaction kinetics from a temperature of 40 °C to 60 °C, showing that the reduction in CO_2 solubility appears to be a secondary phenomenon to the increase in its diffusion, although an upper limit to the temperature increase needs to be determined (Ndiaye et al., 2023) (Kombathula, 2020).

Another important parameter to consider is pressure, whose increase causes a growth in the reaction speed. Increasing the pressure generates two contrasting phenomena: the solubility of CO_2 in water is improved, making the process more efficient, but the diffusion coefficient of carbon dioxide in the material decreases, hindering carbonation (Ndiaye et al., 2023).

However, the first principle described is the dominant phenomenon, so it is possible to say that high pressure increases the rate of carbonation and thus increases the amount of CO_2 absorbed (Dos Reis et al., 2020).

The kinetics of the reaction can also be improved by influencing the concentration of CO_2 used in the process. Thus, faster carbonation can occur if a higher concentration is used, although numerous studies have confirmed the existence of a limit beyond which the process slows down. Initially, a high concentration of CO_2 accelerates the movement of carbon dioxide itself into the pores of the material, allowing it to reach even the deepest points in the sample, but values above 50 % in cementitious materials cause the pores to become saturated, making it more difficult for carbon dioxide to penetrate (Xu et al., 2022).

However, other studies have shown that even with a CO_2 concentration of 10 % in the flow, the process can be made faster (Baciocchi et al., 2009). Furthermore, it has been observed that an increase in CO_2 concentration to values above 15 % generally does not cause a significant improvement in the result (Dos Reis et al., 2020).

In addition to these factors, it is also necessary to assess the properties of the material being used and, in particular, its grain size. Samples composed of finer grains allow for a high dissolution of the minerals and a high exposure to CO_2, which, in turn, can diffuse efficiently on the surface of the material (Kombathula, 2020) (Dos Reis et al., 2020).

Finally, it was observed that the absorption of CO_2 in the material used also depends on the speed of rotation to which the latter was subjected during carbonation. Continuous stirring of the material improves the transfer of gas into it.

Confirming this, the amount of carbon dioxide sequestered with rotation speeds of 20 rpm and 100 rpm was found to be 2.4 and 4.6 times greater, respectively, than the results obtained with a static process (Dos Reis et al., 2020).

2. OBJECTIVES

This paper aims to analyze how the accelerated carbonation reaction can act on different materials and, thus, determine how much CO_2 can be absorbed into them. Each material was tested with different liquid-to-solid ratios, focusing in particular on the value L/S = 0.3 L/kg, as previous studies (Danelon, 2022) have shown that it can guarantee better results to gain a thorough understanding of the process.

All tests were carried out under the same operating conditions, using the same incubator for 4 hours for all samples. Secondly, the study focused on metal release to understand how this phenomenon varies when subjecting the material to carbonation to understand whether or not it renders the final material inert. These results were then also compared with data from the literature to have feedback on the analysis performed.

3. MATERIALS AND METHODS

3.1 Materials Description

The alkaline wastes used to perform accelerated carbonation are of four types: a powder derived from Portland cement, foundry sand, clayey silts, and, finally, fly ashes coming from an incinerator.

3.2 Experimental Scheme

The experimental design used as a reference for the analysis of the four previously mentioned materials can be seen in Figure 7.

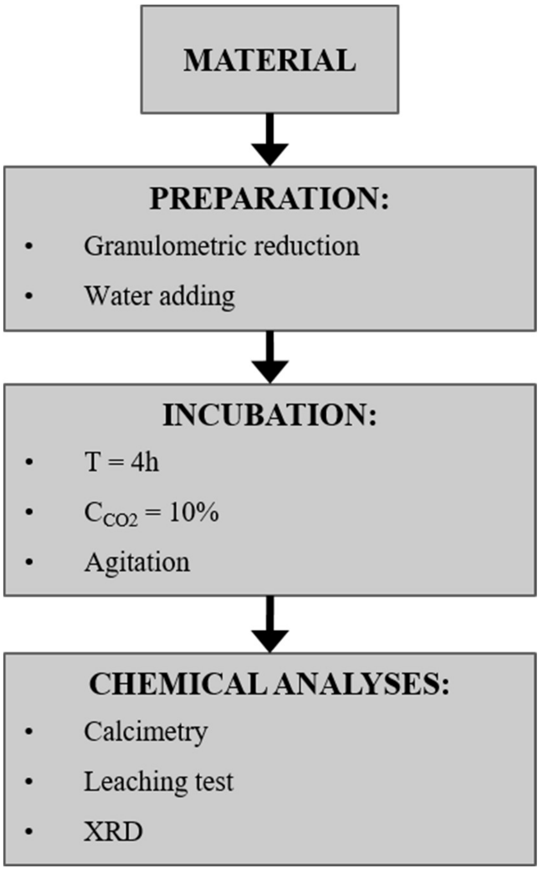


Figure 7: Experimental design used as a reference for the four materials analyzed in the thesis.

For the evaluation of the forced carbonation process, the materials were prepared in terms of grain size and Liquid-to-Solid (L/S) ratio before being placed in the incubator and exposed at a CO_2 concentration set at 10% v/v for a period equal to four hours. Furthermore, the samples showing the best performance in terms of L/S ratio in preliminary tests have been shaken manually during incubation to also simulate the contribution of possible material handling. Before and after forced carbonation, the materials have been characterized through calcimetry tests and chemical analysis.

3.3 Materials Preparation

Before performing the accelerated carbonation test, the Portland material and foundry sand were prepared by grain size reduction to 4 mm using a 'Retsch SM 2000' mill, the setup of which is shown in Figure 8. The clayey silts and fly ashes, on the other hand, as they are characterized by high moisture content, as can be seen in Table 3, could not be subjected to the action of the mill and, therefore, did not undergo particle size reduction.

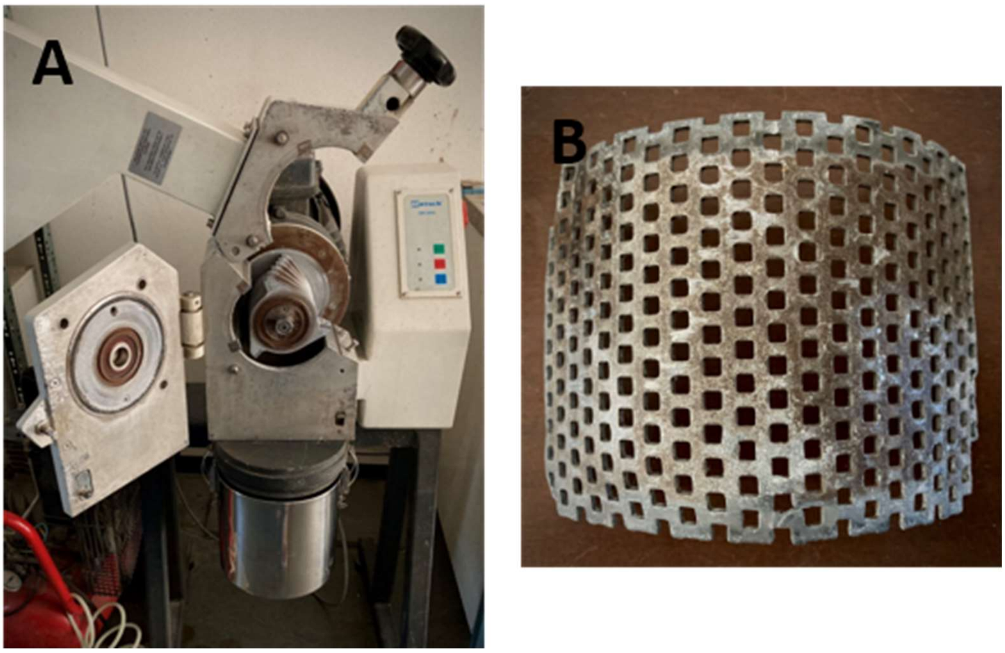


Figure 8: Mill setup used for particle size reduction. (A) Mill setup, (B) 4mm reduction mesh.

Different Liquid-to-Solid ratios (0, 0.2, 0.3) were applied to determine the influence that the degree of moisture can have on the final results of the Portland test. The amount of distilled water added to the sample (L/S = 0.0) has been measured by multiplying the weight of the dry sample (200 g) by the Liquid-to-Solid ratio to be achieved, resulting in values of 40 mL and 60 mL relative to L/S ratios of 0.2 and 0.3, respectively.

For the foundry sand, on the other hand, in addition to the initial material (L/S = 0.0), only the optimum Liquid-to-Solid ratio of 0.3 was tested, even if this latter has been analyzed with and without stirring in the incubator.

Finally, clayey silts, already having an initial moisture content of 0.23 and thus an L/S of 0.3, always considering a dry sample mass of 200 g, have been analyzed as row materials and by adding distilled water until a liquid-to-solid ratio of 0.6 was reached with and without agitation to study the variation in the final results, while for the fly ashes, characterized by an initial humidity of 0.28 and a ratio L/S = 0.4, only the initial condition has been tested.

All tests were carried out in triplicate. The following tables show the total solids and moisture values measured on both pre-and post-carbonation materials.

Table 1: Total Solids and Moisture of Tested Portland Material Samples.

PORTLAND						
ID	PORT-L/S=0.0		PORT-L/S=0.2		PORT-L/S=0.3	
CONDITION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION
HUMIDITY	0,074	0,061	0,216	0,160	0,273	0,205
TOTAL SOLIDS	0,926	0,939	0,784	0,840	0,727	0,795

Table 2: Total Solids and Moisture of Tested Foundry Sand Samples.

SAND						
ID	SAND-L/S=0.0		SAND-L/S=0.3		SAND-L/S=0.3 - MIX	
CONDITION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION
HUMIDITY	0,048	0,044	0,260	0,208	0,260	0,202
TOTAL SOLIDS	0,952	0,956	0,740	0,792	0,740	0,798

Table 3: Total Solids and Moisture of Clayey Silts Samples Tested.

CLAY						
ID	CLAY-L/S=0.3		CLAY-L/S=0.6		CLAY-L/S=0.6 - MIX	
CONDITION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION	BEFORE INCUBATION	AFTER INCUBATION
HUMIDITY	0,228	0,183	0,375	0,335	0,342	0,205
TOTAL SOLIDS	0,772	0,817	0,625	0,665	0,658	0,795

Table 4: Total Solids and Moisture of fly ashes Samples Tested.

FLY ASHES		
ID	BFLY-L/S=0.4	
CONDITION	BEFORE INCUBATION	AFTER INCUBATION
HUMIDITY	0,280	0,203
TOTAL SOLIDS	0,720	0,797

Regarding the L/S ratios, the measured humidity value has been rounded to the tenth.

3.4 Carbonation Test

For the carbonation tests, samples of described materials, each one with a dry weight of 200 g, have been prepared and subsequently placed in aluminum trays. The various types of Portland material samples tested in this way are shown in Figure 9, the foundry sand samples analyzed are presented in Figure 10, and, finally, the clayey silt samples can be seen in Figure 11.

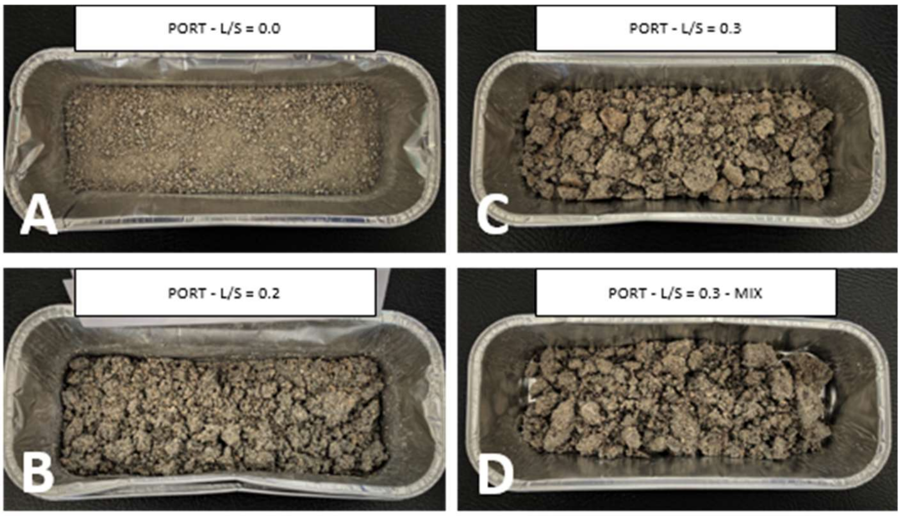


Figure 9: Several analyzed samples of Portland material. (A) L/S = 0.0, (B) L/S = 0.2, (C) L/S = 0.3, (D) L/S = 0.3 mixed every hour in the incubator.

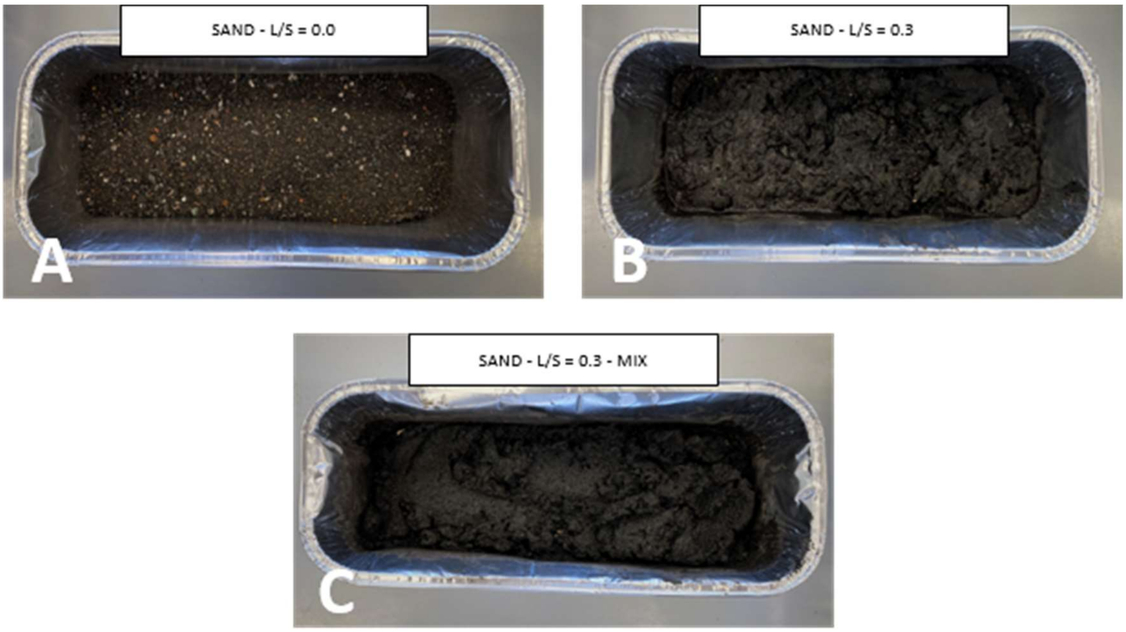


Figure 10: Several analyzed samples of foundry sand. (A) L/S = 0.0, (B) L/S = 0.3, (C) L/S = 0.3 mixed every hour in the incubator.

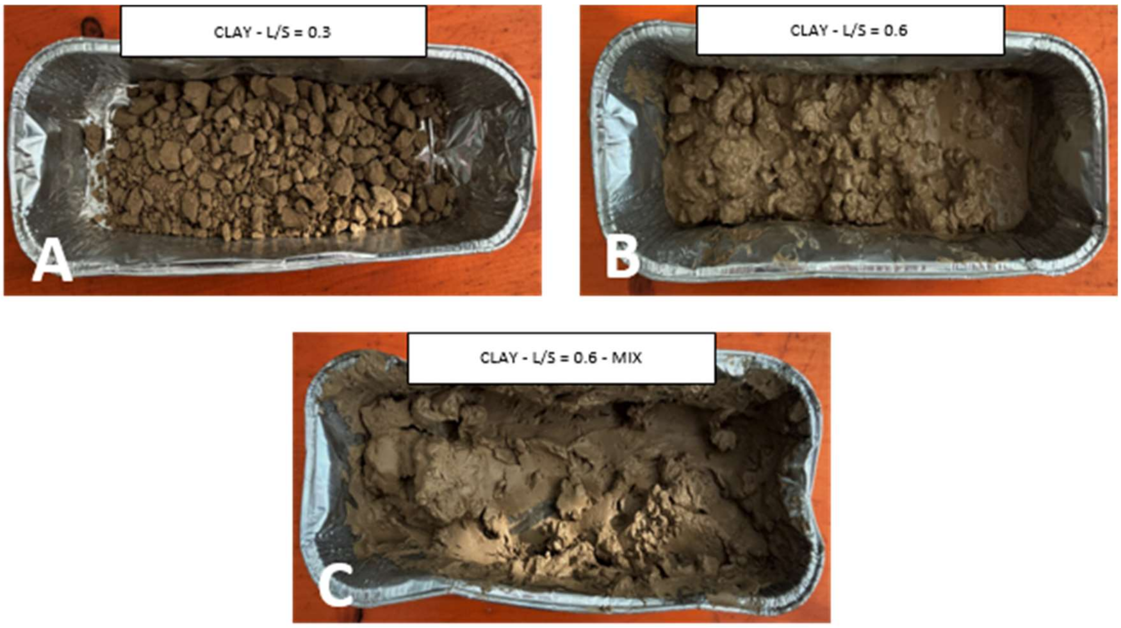


Figure 11: Different analyzed samples of clayey silts. (A) L/S = 0.3, (B) L/S = 0.6, (C) L/S = 0.6 mixed every hour in the incubator.

3.4.1 Incubation

To carry out the accelerated carbonation, the trays containing the material have been placed inside a static chamber: a N-BIOTEK NB-203 CO_2 incubator at a constant temperature of 35 °C and a CO_2 concentration set at 10% v/v to simulate the concentration of carbon dioxide that can be found in chimney smokes.

By using the static reactor, it was possible to keep these parameters constant within the chamber for the duration of the process, which in this case was chosen to be 4 hours.

The choice of incubation time is a compromise between the need to guarantee operating times compatible with industrial practice and the achievement of a sufficient level of saturation of the maximum carbonation capacity as a function of time. In fact, previous experience has shown that considering three different incubation times (2, 4, and 8 hours), most of the CO_2 uptake (62.86%) occurs in the first two hours, while the remaining 37.14% occurs in the next 6 hours (Danelon, 2022). It can, therefore, be stated that the rate of carbonation is greatest in the first few hours and then decreases in the following hours until a stable CO_2 uptake value is reached. These results were also confirmed by several authors (Yuan et al., 2022) (Schnabel et al., 2021) (Ulkwattage et al., 2013) who performed forced carbonation on materials such as recycled concrete aggregates, metal slag, and incineration ashes.

In Figure 12 below, the incubator setup can be seen in detail. It consists of a CO_2 cylinder connected to the chamber via a small tube in which a valve is positioned to select the gas concentration inside the reactor. At the front, there is a small monitor that allows the values of parameters useful for the process to be set, while inside the incubator is the temperature probe and the compartments where the samples to be tested can be placed.

In the samples with the best results, the material was also tested by simulating agitation during incubation by stirring it every hour inside the incubator in a way to stimulate the CO_2 diffusion inside it.

Figure 12: CO_2 incubator setup used. (A) closed incubator with a dashboard for choosing temperature and CO_2 concentration, (B) a CO_2 cylinder connected to the incubator, and (C) an open incubator with shelves on which to place samples.

3.5 Calcimetry Analyses

The success of the process was then analyzed by comparing the results obtained from the calcimetry tests conducted on the initial material and the data obtained from the calcimetry tests conducted on the materials subject to carbonation; in particular, three calcimetry tests were conducted for each sample.

Analyses were conducted using the 'Dietrich-Fruhling' calcimeter shown in Figure 13.

With this instrument, it is possible to determine the amount of carbon dioxide absorbed by the material by referring to the following chemical reaction:

$$CaCO_{3(s)} + 2HCl_{(l)} \rightarrow CaCl_{2(l)} + H_2O_{(l)} + CO_{2(g)}$$

As can be seen, hydrochloric acid (HCl) reacts with the calcium carbonates ($CaCO_3$) within the material, producing carbon dioxide in gaseous form. Thus, by calculating the amount of CO_2 produced in the material pre- and post-carbonation, it is possible to determine the total uptake of carbon dioxide in the sample.

Figure 13: Calcimeter used for the analysis of pre-and post-carbonation materials.

As can be seen, the calcimeter consists of a metal stand on which a container of demineralized water is placed, a graduated bottle in which the material and hydrochloric acid (HCl) are placed, a graduated cylinder that can hold a maximum of 100 mL of liquid and a final venting vessel.

The procedure involves taking a small amount of material and placing it inside the graduated bottle in which a small test tube containing HCl acid (35% v/v) is also placed.

Before the analysis, the graduated cylinder is brought to a level of 100 mL, and only after closing the hermetic stopper of the bottle the container is shaken to make the acid react with the material, producing CO_2: the production of the gas causes the decrease of the water level, which is discharged into an appropriate container. Thus, after reaching a stable condition, it is possible to read the volume of CO_2 produced, which is useful for calculating the number of carbonates present in the material later on. The volume of CO_2 produced with the post-carbonation material must be greater than the amount obtained with the samples tested before carbonation to confirm that, with the use of the incubator, the amount of calcium carbonate increased, and CO_2 has been absorbed by the tested compound.

In this study, the first stage of calcimetry has been used to decide on the amount of material and the volume of HCl to be used to not underestimate the presence of carbonates.

Initially, for the Portland material, 5 g was tested by reacting it with 5 mL of HCl, but a fast consumption of 100 mL of water was observed with an underestimation of the amount of $CaCO_3$ that led to a subsequent decrease in the material tested, finally choosing an amount of approximately 0.5 g. In addition, different sizes of graduated bottles were tested, following the principle that smaller bottles correspond to more uniform bottoms and ensure a better distribution of the acid, which in larger containers tended to accumulate at the sides of the bottom, causing less contact with the material. It was also observed that, at the end of the process in the CO_2 incubator, the Portland material showed a great change in color between the top and bottom, indicating successful carbonation only in the most exposed part of the compound. This is the reason why three further samples of Portland material were made with a liquid-to-solid ratio of 0.3, with the compound being mixed every hour during the incubation time.

For foundry sands and clayey silts, on the other hand, 5 g of material and 5 ml of HCl were used for the former, and 0.5 g of material and 5 ml of HCl for the latter, respectively.

3.6 Chemical Characterization Analyses

Chemical analyses have been performed before and after the completion of the carbonation tests to gain a more detailed and thorough understanding of the process.

3.6.1 XRD analysis

The materials have been analyzed using XRD (X-Ray Diffraction) technology to know the detailed composition of the samples and observe possible differences between the composition of the material pre- and post-accelerated carbonation. The procedure consists of irradiating a sample with X-rays and measuring the intensity and inclination of the rays refracted by the material, which is necessary information to understand the quantity of the components present in it. In this study, the XRD analysis was carried out, for the Portland material, on the uncarbonated sample (L/S = 0.0) and the sample with L/S ratio = 0.3 agitated during incubation.

3.6.2 Leaching tests

Leaching tests were also carried out to assess the possible leaching of compounds in the pre-and post-carbonation materials. Analyses were conducted according to the UNI EN 12457-2:2004 standard containing information about the procedures to be followed in carrying out the test. A necessary condition for the final results to be true is that the material tested must be dry. The standard indicates the use of a ratio of 10 L of leach, in this case, distilled water, to 1 kg of dry material.

Therefore, the moisture content of the material has been taken into account in each test to maintain the ratio specified by the standard.

For all the materials tested, leaching tests have been conducted on the initial material, which did not undergo carbonation, and on the material which, after carbonation, presented the greatest quantities of CO_2 absorbed: the material with an L/S equal to 0.3 mixed in the case of portland and foundry sand, and the material as is, which already had an L/S = 0.3, in the case of clayey silts. Again, all tests were carried out in triplicate.

Then, the materials were placed inside containers with a capacity of 1 L each, in quantities equal to 100 g of dry compound each. In particular, for the Portland carbonate material, only one bottle has been filled with a smaller quantity equal to 50 g. In doing so, 1 L of distilled water was used for the 100 g quantities, while in the case of the 50 g sample, 500 mL of solution was poured in. As can be seen in Figure 14, the finished containers were placed in a 'Rotax' rotary mixer to ensure continuous mixing for 24 hours. At the end of this process, the supernatant obtained was passed through a 45 µm mesh filter using a vacuum pump.

Finally, the filtered solution was chemically analyzed to determine the substances present in it.

Subsequently, the procedure was repeated for foundry sand. In this case, the material did not settle after the 24-hour mixing time, and the vacuum pump could not work efficiently; thus, the liquid obtained was subjected to centrifugation to separate the coarse fraction from the supernatant and increase the efficiency of the subsequent filtration. The filtered liquid was then obtained from each sample using the equipment described above in the same manner.

The methodology was then repeated for the clayey silts, for which, however, the use of centrifugation before filtering was not necessary.

Figure 14: Rotax 6.8 rotary mixer used for the leaching tests.

A very important parameter, whose variation must be assessed employing leaching tests, is the pH. Indeed, it is the chemical parameter that most influences and controls the leaching of substances present in a compound in more or less significant quantities. Small changes in this variable can cause significant increases or decrease in the concentrations of a substance in the leachate. In particular, pH can change when, as in this case, alkaline materials are exposed to CO_2 and are, therefore, subjected to carbonation. This chemical reaction, through the absorption of CO_2, transforms oxides into carbonates, causing a decrease in pH and thus generating a change in the solubility and release of certain substances present in the initial compound (Saveyn et al., 2014).

4. RESULTS

4.1 Calcimetry Tests

Calcimetry analyses were used to determine the amount of carbon dioxide sequestered by each material due to the carbonation reaction. Firstly, starting from the volume of CO_2 absorbed by the sample, measured through the calcimeter described above, it was possible to determine the relative quantity in moles thanks to the ideal gas law:

$$pV = nRT$$

Where p is the pressure (1 atm), T is the temperature (298.15 K), R is the gas constant (0.082057338 L · atm/K · mol), V is the measured volume (L), and n is the number of moles (mol).

Then, knowing that the molecular weight of carbon dioxide is 44 g/mol, the mass of CO_2 was calculated by multiplying this value by the amount of carbon dioxide in moles. Next, the value for the mass of CO_2 relative to the dry mass of the material used in the calcimeter was found by multiplying the total mass (g) by the percentage of total solids in it. This calculation was carried out for each test before measuring the average of this data for each sample tested, the relative standard deviation, to finally, obtain the amount of CO_2 absorbed, removing from this data the carbon dioxide measured through the calcimeter tests on the non-carbonate material as it was initially present in the material itself. The results thus obtained can be seen in Figure 15, which shows the amount of carbon dioxide sequestered by each material as a function of the different L/S ratios used. Thus, it can be seen that in materials with a Liquid-Solid ratio of 0.0 L/kg, CO_2 uptake was 0, confirming what was announced in Chapter 1.3, namely that accelerated carbonation requires a certain amount of water to be efficient, especially when, as in this case, working at room temperature. In fact, in all four materials, the greatest results were shown in cases where water was present in the sample and, in particular, when an L/S ratio of 0.3 L/kg was used for Portland residues, foundry sand, and clayey silts while an L/S = 0.4 L/kg for the fly ashes. In cases where the samples were mixed every hour in the incubator, a slight increase in performance was shown, indicating that according to what has been observed in articles found in the literature, mixing the material improves gas transfer within the material itself and, therefore, increases the amount of CO_2 that can be absorbed. The maximum value in terms of carbon dioxide sequestered was 15 g_{CO2}/kg_{DW} obtained in the clayey silts, while a value of 11 g_{CO2}/kg_{DW} and 8 g_{CO2}/kg_{DW} was achieved in the Portland material and foundry sand, respectively.

In the clayey silts, moreover, since the Liquid-Solid ratio in the initial material was already 0.3 L/kg, two further conditions were tested, both with L/S = 0.6 L/kg, with the latter also subject to mixing. In both cases, the carbon dioxide absorbed was 0 g_{CO2}/kg_{DW}, confirming the fact that in the presence of too much water, the pores of the material are saturated, hindering the diffusion of CO_2 within them and, therefore, making the process inefficient.

Finally, concerning the accelerated carbonation of fly ashes, a much higher average carbon dioxide uptake was observed than in previous cases, amounting to 85 g_{CO2}/kg_{DW}.

It can be stated that the amount of CO_2 absorbed in the Portland material is comparable to the values found in the literature with the carbonation of recycled cement aggregates (RCA) in granular form, tested under 'very mild' conditions (T < 35 °C, P = 1 bar and C_{CO2} = 0 - 20 %). The data obtained with foundry sands and incineration slag are lower than the values found through the critical analysis concerning the slags and fly ashes uptakes, while the measurement of carbon dioxide sequestered through clayey silts, analyzed without particle size reduction, is in line with the quantities determined in the literature using natural aggregates even if they were prepared in a monolithic form and the studies are only a few.

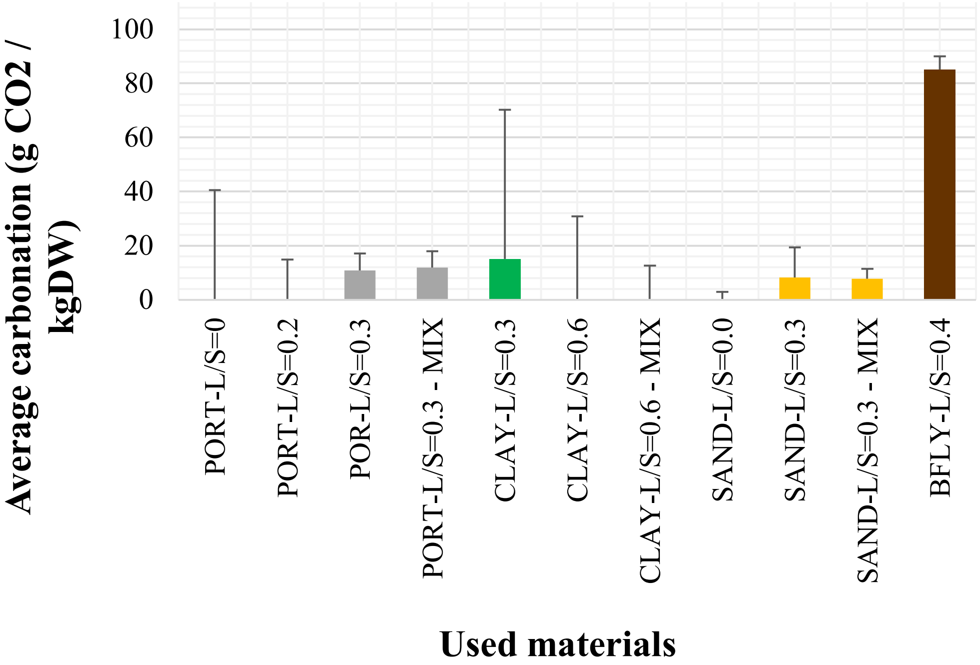


Figure 15: Experimental results obtained for the various materials concerning the different L/S ratios analyzed.

4.2 Leaching tests

As explained in Section 3.6.2, all the analyzed materials were subjected to leaching tests to determine any changes caused by carbonation in the mobility of metals within the material. The results of these analyses can be seen below; Table 5 presents the data for the Portland material, as it initially was pre-carbonation and with L/S = 0.3 post-carbonation mixed hourly, Table 6 shows the results for the foundry sands, as it initially was with L/S = 0.3 post-carbonation mixed hourly, while Table 7 shows the analyses for the clayey silts, as it initially was pre-carbonation and with L/S = 0.3 post-carbonation.

The results highlighted in red are those showing concentrations of the metals greater than the limits dictated by Legislative Decree No 22 of 5 February 1997 (version coordinated with the Ministerial Decree of 5 April 2006) regarding the identification of non-hazardous waste subject to simplified recovery procedures. Concerning the Portland material, a significant reduction in the values of Barium and Chromium was seen comparing the data obtained before and after the carbonation reaction, respectively, from 1.4 - 1.6 mg/L to 35 - 42 μg/L for the former and from 38 - 68 μg/L to values of 24 and less than 10 for the latter, while the concentration of Nickel slightly increased.

The high values of Barium and Chromium in the non-carbonated Portland material are also confirmed by some test reports on the same type of material. For foundry sands and clay silts, on the other hand, no values above the limits were recorded in both cases, before and after incubation. In the foundry sands, only a small reduction in Copper (Cu) was observed, from 14 - 23 μg/L to values below 10 μg/L, and Molybdenum (Mo), whose concentrations fell from 30 - 41 μg/L to 22 - 25 μg/L, while Barium (Ba) slightly increased from initial values of 14 - 19 μg/L to final values of 18 - 29μg/L. Finally, in the clayey silts, the concentrations of all metals were less than 10 μg/L except for Vanadium (V), of which a small decrease was seen from values of 13 - 15 μg/L to values of 11 μg/L.

About the Portland material, a reduction in pH between the initial material, having a pH = 13, and the carbonate material, characterized by a pH = 12, was also highlighted. Concerning this parameter, the results obtained were compared with data found in the literature, referring to the document called “Study on methodological aspects regarding limit values for pollutants in aggregates in the context of the possible development of end-of-waste criteria under the EU Waste Framework Directive” (Saveyn et al., 2014). In addition, five test reports on the same type of material, waste from Portland cement, were considered, which, in line with what was found in the end-of-waste tests, showed Barium and Chromium values above the legal limit.

Thus, as reported in Figure 16 and Figure 17, the variations in the release of both of the previously mentioned elements as a function of the pH were then analyzed.

In this way, examining the information contained in the document concerning 'Recycled Concrete' (RecCon), an increase in the average release of both Barium and Chromium was observed from 5 mg/kg to 10 mg/kg for the former and from 0.07 mg/kg to 0.3 mg/kg for the latter with the change in pH from 13 to 12. This change, however, is in contrast to the findings of the cutoff tests, whereby a reduction in both Barium and Chromium was observed in all three samples tested.

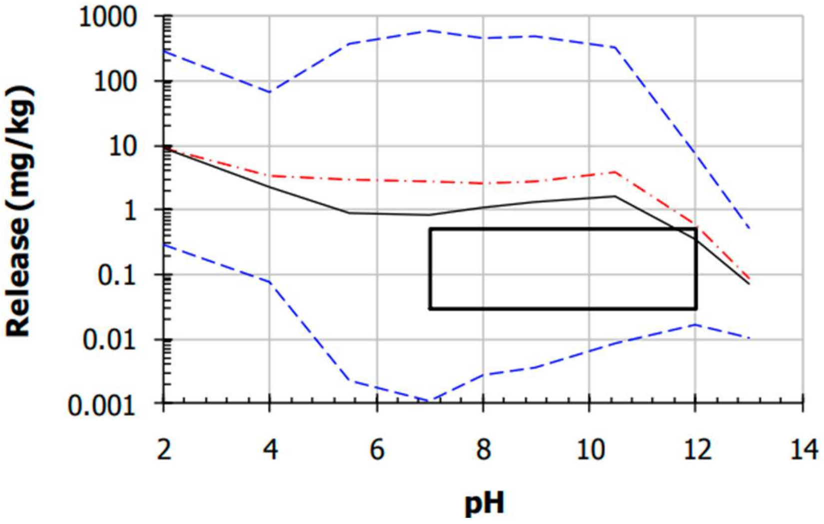


Figure 16: pH-dependent release of Cr (Saveyn et al. 2014).

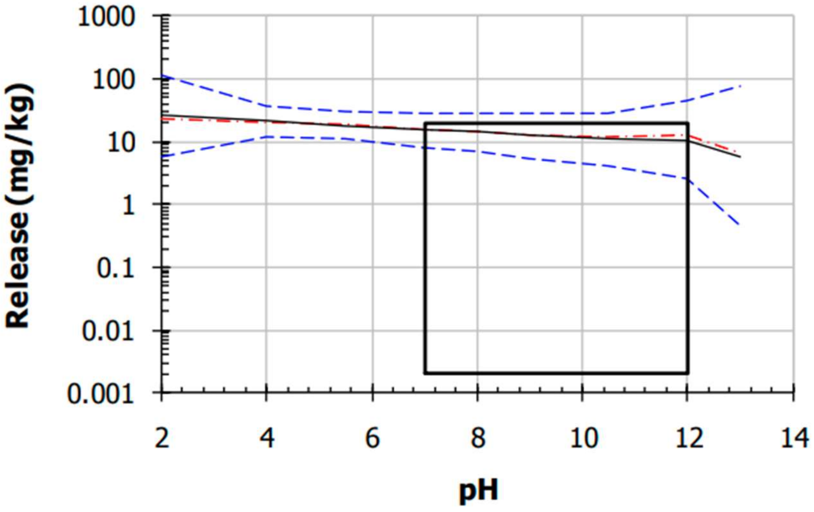


Figure 17: pH-dependent release of Ba (Saveyn et al. 2014).

Table 5: Results of the yield test performed on Portland material.

CONDITION	ID	Ba (µgBa/l)		Be (µgBe/l)		Cd (µgCd/l)		Co (µgCo/l)		Cr (µgCr/l)		Cu (µgCu/l)		Mo (µgMo/l)		Ni (µgNi/l)		Pb (µgPb/l)		V (µgV/l)		Zn (µgZn/l)	
		AV.	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**
PRE-CARBONATION	PORT - L/S = 0.0	1517	121	< 10	0	< 10	0	< 10	0	56	16	42	28	< 10	0	17	12	< 10	0	< 10	0	< 10	0
POST-CARBONATION	PORT - L/S = 0.3 - MIX	39	4	< 10	0	< 10	0	< 10	0	15	9	48	23	13	1	38	14	< 10	0	< 10	0	< 10	0

Table 6: Results of the release test performed on foundry sands.

CONDITION	ID	Ba (µgBa/l)		Be (µgBe/l)		Cd (µgCd/l)		Co (µgCo/l)		Cr (µgCr/l)		Cu (µgCu/l)		Mo (µgMo/l)		Ni (µgNi/l)		Pb (µgPb/l)		V (µgV/l)		Zn (µgZn/l)	
		AV.	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**
PRE-CARBONATION	PORT - L/S = 0.0	17	2	< 10	0	< 10	0	< 10	0	< 10	0	16	7	35	6	< 10	0	< 10	0	< 10	0	11	1
POST-CARBONATION	PORT - L/S = 0.3 - MIX	22	6	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	24	1	< 10	0	< 10	0	< 10	0	17	10

Table 7: Results of the release test performed on clayey silts.

CONDITION	ID	Ba (µgBa/l)		Be (µgBe/l)		Cd (µgCd/l)		Co (µgCo/l)		Cr (µgCr/l)		Cu (µgCu/l)		Mo (µgMo/l)		Ni (µgNi/l)		Pb (µgPb/l)		V (µgV/l)		Zn (µgZn/l)	
		AV.	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**	**AV.**	**S.D.**
PRE-CARBONATION	PORT - L/S = 0.0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	14	1	< 10	0
POST-CARBONATION	PORT - L/S = 0.3 - MIX	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	< 10	0	11	0	11	1

4.3 XRD Analysis

As previously said, the XRD tests have been performed only on Portland residues and, in particular, on the non-carbonated sample with an L/S = 0.0 and on the carbonated one characterized by an L/S = 0.3 mixed during the incubation time.

Thus, it has been possible to determine the mineralogical composition of the interested material to observe its variation due to the occurring of accelerated carbonation. In particular, Figure 18 presents the structure image of the two samples studied with a scale of 100 µm and with different colors representing different minerals (Al, Ca, Si, and Mg).

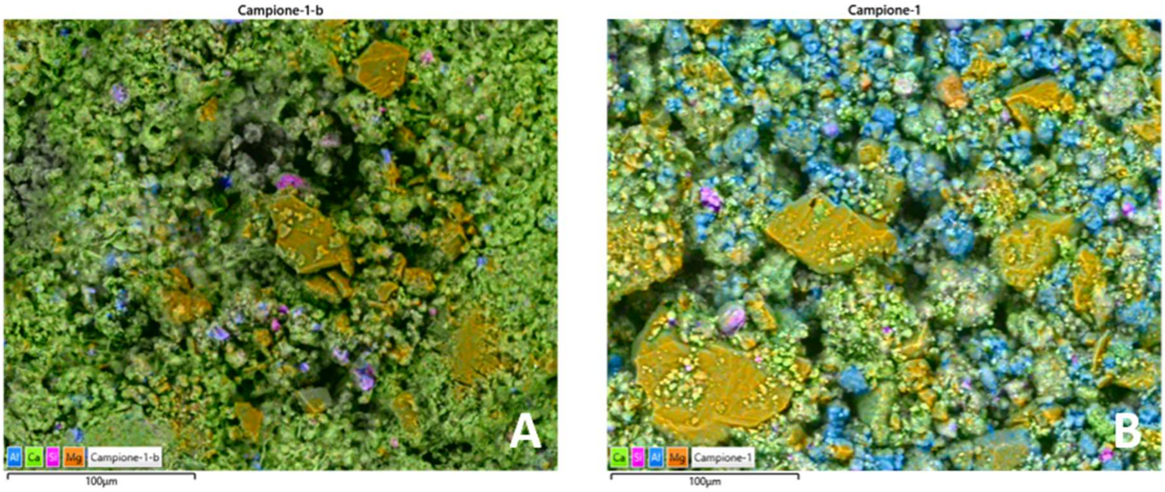
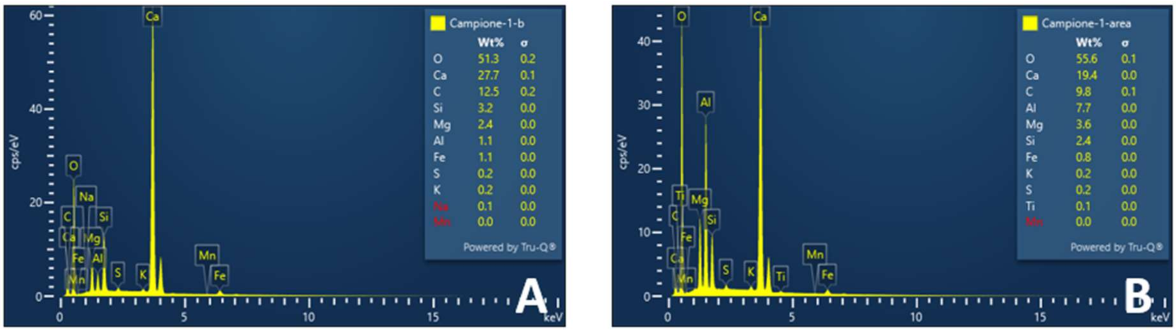


Figure 58: Mineralogical structure of the Portland residues: (A) non-carbonated sample with L/S = 0.0, (B) carbonated sample with L/S = 0.3 mixed during the incubation.

Furthermore, the X-Ray Diffraction method permits to obtain a graph called diffractogram that shows the presence of each mineral and oxide both in terms of percentage. Concerning the mineral composition, the comparison of the two analyzed samples is reported in Figure 19, whereas regarding the oxide composition, it’s possible to see Figure 20.



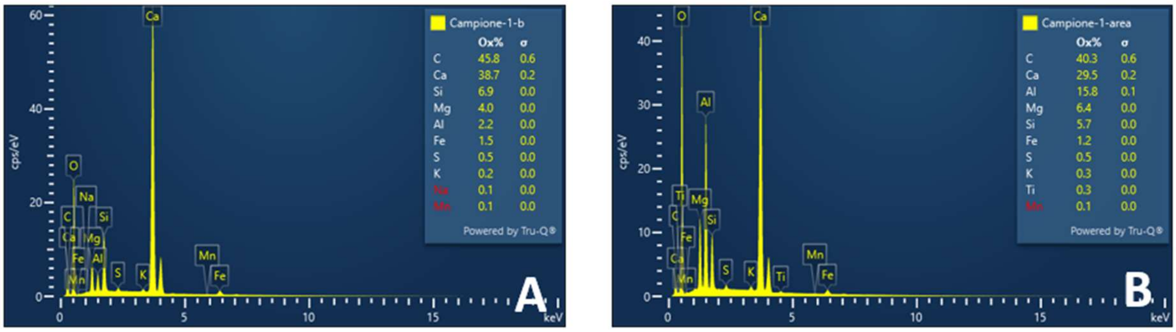
Campione-1-b

	Wt%	σ
O	51.3	0.2
Ca	27.7	0.1
C	12.5	0.2
Si	3.2	0.0
Mg	2.4	0.0
Al	1.1	0.0
Fe	1.1	0.0
S	0.2	0.0
K	0.2	0.0
Na	0.1	0.0
Mn	0.0	0.0

Campione-1-area

	Wt%	σ
O	55.6	0.1
Ca	19.4	0.0
C	9.8	0.1
Al	7.7	0.0
Mg	3.6	0.0
Si	2.4	0.0
Fe	0.8	0.0
K	0.2	0.0
S	0.2	0.0
Ti	0.1	0.0
Mn	0.0	0.0

Figure 19: Mineral composition (%): (A) non-carbonated sample with L/S = 0.0, (B) carbonated sample with L/S = 0.3 mixed during the incubation.



Campione-1-b

	Ox%	σ
C	45.8	0.6
Ca	38.7	0.2
Si	6.9	0.0
Mg	4.0	0.0
Al	2.2	0.0
Fe	1.5	0.0
S	0.5	0.0
K	0.2	0.0
Na	0.1	0.0
Mn	0.1	0.0

Campione-1-area

	Ox%	σ
C	40.3	0.6
Ca	29.5	0.2
Al	15.8	0.1
Mg	6.4	0.0
Si	5.7	0.0
Fe	1.2	0.0
S	0.5	0.0
K	0.3	0.0
Ti	0.3	0.0
Mn	0.1	0.0

Figure 20: Oxides composition: (A) non-carbonated sample with L/S = 0.0, (B) carbonated sample with L/S = 0.3 mixed during the incubation.

Finally, the graphs showing the minerals present in the samples with their names are shown in Figure 21.

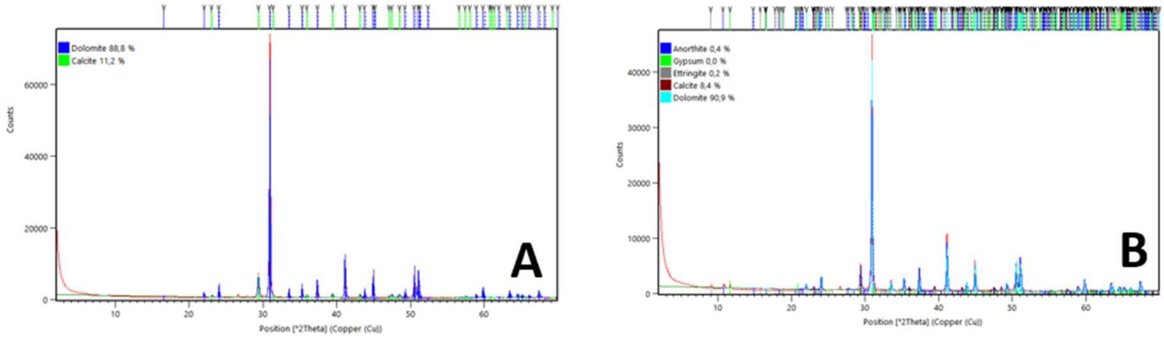


Figure 21: Minerals present in the samples: (A) non-carbonated sample with L/S = 0.0, (B) carbonated sample with L/S = 0.3 mixed during the incubation.

It's possible to see that the mineral composition of non-carbonated and carbonated materials remained almost the same. In fact, the dominant presence of Dolomite ($C_2Ca_1Mg_1O_6$) with 88,8 %, and Calcite ($C_1Ca_1O_3$) with 11,2 presented in the non-carbonated samples have also been found in the carbonated residues even if they are also characterized by Anorthite ($Al_2Ca_1O_8Si_2$) and Ettringite ($H_{64}Al_2Ca_6O_{50}S_3$) but in a minimal quantity (0,4 % and 0,2 %).

So, it's possible to conclude that with a CO_2 concentration only equal to 10 %, the accelerated carbonation is not so efficient for the Portland residues, and their composition does not change too much. This is confirmed also by the carbon uptake results analyzed in the critical review and obtained in Mild conditions, while with Mild, Intensive, and Very Intensive operative conditions an higher quantity of carbon dioxide could be absorbed by granular RCAs.

4.3.1 Theoretical CO_2 uptake

Thanks to the oxide composition of the non-carbonated Portland sample characterized by a Liquid-to-Solid ratio of 0.0 L/kg, it has been possible to measure the amount of CO_2 that, in theory, can be absorbed by the material by using the Steinour equation:

$$CO_2(\%, max) = 0{,}785 \cdot (CaO - 0{,}7SO_3) + 1{,}091MgO + 1{,}420Na_2O + 0{,}935K_2O$$

Thus, it has been determined a potential carbon dioxide uptake equal to 35 % that corresponds to 350 $g_{CO2}/kg_{sorbent}$, a value too much higher than the effective one determined with the experimental analysis.

5. CONCLUSIONS

In conclusion, thanks to this study, it was possible to confirm some of the information about forced carbonation reported in the literature.

Firstly, it was seen that the presence of water in the samples is a fundamental condition for the efficient absorption of CO_2 into the tested material. However, if present in excessive amounts, as reported in clayey silts with L/S = 0.6, it hinders the diffusion of carbon dioxide, making the process ineffective.

In all materials analyzed, the maximum amount of CO_2 adsorbed was obtained using a liquid-solid ratio of 0.3 L/kg with results ranging from 8 g_{CO2}/kg_{DW} to 15 g_{CO2}/kg_{DW}. In addition, by mixing the samples every hour during the incubation period, slightly better results were obtained, thus verifying the hypothesis that this activity improves the diffusion of CO_2 within the pores of the tested material by sequestering more of it.

The only exception was the incineration slag, which already had an L/S ratio of 0.4 L/kg and did not undergo a particle size reduction to 4 mm and absorbed 85 g_{CO2}/kg_{DW} of CO_2, a much better result than the previous ones, which may be due to the greater number of oxides available within the slag compared to the other materials.

Concerning the leaching of metals, on the other hand, a strong reduction in the concentrations of Barium and Chromium was observed, from 1.4 - 1.6 mg/L to 35 - 42 μg/L for the former and from 38 - 68 μg/L to values of 24 and less than 10 for the latter, respectively, in the Portland carbonate material compared to the non-carbonate material, which had values above the legal limits. This result, although in contrast to the data reported in the literature (Saveyn et al. 2014), allows us to state that forced carbonation, besides being an important process for reducing carbon dioxide in the atmosphere, can also be considered an inerting reaction of the tested materials that improve their environmental quality. For foundry sands and clayey silts, on the other hand, no substantial changes were found as the metal concentrations were already low in the initial non-carbonated materials.

PART III – ANNEX

In this third part, all the graphs, tables, and data used for the thesis are reported. So, in the following annexes, it’s possible to see all the information used in this thesis, starting from the critical review and finishing with the CO_2 absorption results obtained by the accelerated carbonation for all the tested materials.

ANNEX I: CRITICAL REVIEW – INCLUDED PAPER

This annex is related to the 38 papers included using the Prisma method. All of them have been analyzed in order to be compared to each other, so here, in this section, it’s possible to see the main features of each scientific article found in the literature.

REFERENCE	OBJECTIVE OF THE PAPER	TYPOLOGY OF REACTOR	SORBENT TYPOLOGY	EXPERIMENT DESIGN	VARIABLES	OPERATIVE CONDITIONS	SCALE OF EXPERIMENT	CHARACTERIZATION OF THE SORBENT	METHODS TO ASSES CARBONATION UPTAKE	RESULTS [gCO_2/kg_{dw}]
G. Cappai, S. Cara, A. Muntoni, M. Piredda. Application of accelerated carbonation on MSW combustion APC residues for metal immobilization and CO2 sequestration. 2011 doi: 10.1016/j.jhazmat.2011.04.013	Application of an accelerated carbonation treatment to APC residues, in order to assess the potential of the process in terms of sequestration of the CO2 emitted from WtE point sources as well as the influence on the environmental behaviour of the residue under concern.	Open reactor with magnetic stirrer.	APC ash samples coming from WtE plant (Boiler ashes) and pm collected by the bag filters. (Fly ashes)	The applied accelerated carbonation treatment consisted of a single step aqueous-route process. Slurry route.	Ambient conditions (T=20 ◦C and atmospheric pressure). L/S = 2.5	Mild	Time: about 3hr needed to reach Ph = 8.22. Mass of sorbent: no present	Preliminary charachterization: Metal and anioin content, total carbon content (TC), loss on ignition (LOI). Mineralogy: XRD, Thermogravimetric (TG), differential thermoanalyses (DTA)	The CO2 sequestration potential of APC ash was evaluated by both calcimetry and TG analyses.	200 g/kg
Dongxing Xuan, Chi Sun Poon. Sequestration of carbon dioxide by RCAs and enhancement of properties of RAC by accelerated carbonation. 2018. doi:10.1016/B978-0-08-102480-5.00016-6	Adopting the accelerated carbonation technique to improve the quality of RCAs (Recycled Concrete Aggregates)	Batch reactor.	RCAs (Recycled Concrete Aggregates)	Pressurised carbonation ŸŸ	(100% CO2) -> first Pressurization at P=0.5bar, then P=0.1-5bar), RH=50 ± 5%, T=25 ± 3°C.	Very Intensive	Time: 24hr Mass of sorbent: no present	The composition is given: Size of aggregates (mm): 10-20, 5-10, 2.36-5, <2.36. Apparent density (kg/m^3): 2605, 2624, 2610, 2572. Water absorption (%): 7.2, 6.1, 15.0, 16.4. Cement content (%): 11.2, 16.8, 22.5, 24.	The CO2 sequestration potential of APC ash was evaluated by TG analyses.	(P=5 bar) up to 28,2 gCO2/Kg dry sorbent. Data are deduced from plots.
		ŸOpen reactor.		Flow-through carbonation	Ambient T and P, mixture of pure CO2 and air, flowrate controlled from 1.0 to 10 L/min), RH=50 ± 5%, T=25 ± 3°C.	Mild				Different values according to the carbonation time 6,5 (xx) ; 12,6 (xx) ; 24,0;19,5; 19,5 gCO2/Kg dry sorbent
Sushanth Kombathula. Sequestration of carbon dioxide in steel slag. 2020.	Developing an industrial process to sequester CO2 in metallurgical slag. Study the effects of parameters(T, t, shape) on the carbonation speed and efficency	Open reactor (tubular furnace)	EAF (Electric Arc Furnace) slugs with high concentration of CaO.	Observe how yeld of carbonation changes with time.	Time = 1, 2, 4, 6hr and T = 200°C, Flow rate CO2 = 3L/min, grain size < 0.25mm.	Intensive	Time: at least up to 6hr. Mass of sorbent: not present.	The EAF slug composition is given (CaO = 38%, FeO = 26%, SiO_2 = 13.3%, MgO = 13.2%, Al_2O_3 = 8.27%)	Not present.	0 - 40 - 44 - 53 gCO_2/Kg --> 1 - 2 - 4 - 6 hr

REFERENCE	OBJECTIVE OF THE PAPER	TYPOLOGY OF REACTOR	SORBENT TYPOLOGY	EXPERIMENT DESIGN	VARIABLES	OPERATIVE CONDITIONS	SCALE OF EXPERIMENT	CHARACTERIZATION OF THE SORBENT	METHODS TO ASSES CARBONATION UPTAKE	RESULTS [gCO_2/kg_{dw}]
	and propose a process to sequester CO2 by using steel slags			Study how T affects the yeld of carbonation.	Time = 2hr, T1 = 200°C, T2 = 360°C, Flow rate CO2 = 3L/min, grain size < 0.25mm.					40 - 20 gCO2/Kg --> 200 - 360 °C
				Investigate how shape of slag affects the yeld of carbonation.	Powder and pellet (pressed powder), Time = 2hr, T = 200°C, Flow rate CO2 = 3L/min, grain size < 0.25mm.					40 - 0 gCO2/Kg --> powder - pelle
R. Baciocchi, G. Costa, A. Polettini, R. Pomi, V. Prigiobbe. Comparison of different reaction routes for carbonation of APC residues. 2009.	Identify the most promising conditions for a full scale application.	Open reactor	APC residues of a MSW incineration plant (Fly Ashes)	Dry route -> gas-solid reaction between CO_2 and APC residues.	T = 300-500°C, P = 1bar, CO_2-argon mix (10-50% vol CO_2).	Intensive	Time: 10min to 48hr Mass of sorbent: 44mg of APC residues	Water content, Loss of Ignition (LoI), element and majoe anion content, carbonate content, mineralogy, acid/base neutralization capacity (ANC/BNC), leaching behaviour.	A Thermogravimetric system (TGA, Netzsch STA 409 CD) coupled with Gas Chromatography (GC, 2-channel Varian Micro GC, equipped with a Molsieve-5A and a Poraplot-U columns) for measurement of the evolving CO2.	max 250gCO2/kg
		Slurry reactor		Liquid route	T = 30-50°C, L/S = 0.02, 0.1, 0.2, 0.4, 0.6 L/kg, CO_2 = 100%, P = 1, 3, 5, 7, 10bar, Humidity = 75%.	Very Intensive	Time: 10min to 48hr Mass of sorbent: 1g of dry ash		By measuring the weight gain upon carbonation and the increase in the carbonate content of the samples, measured by calcimetry analysis, accounting for the	

REFERENCE	OBJECTIVE OF THE PAPER	TYPOLOGY OF REACTOR	SORBENT TYPOLOGY	EXPERIMENT DESIGN	VARIABLES	OPERATIVE CONDITIONS	SCALE OF EXPERIMENT	CHARACTERIZATION OF THE SORBENT	METHODS TO ASSES CARBONATION UPTAKE	RESULTS [gCO_2/kg_{dw}]
									weight increase upon carbonation.	
Hilal El-Hassan, Yixin Shao. Carbon Storage through Concrete Block Carbonation Curing. 2014.	Develop a carbonation curing process that can be used to replace steam curing for CMU production.	Batch reactor.	concrete slab samples.	Concrete slab samples after initial curing were placed in a sealed chamber. The chamber was placed on a digital balance to obtain the mass curve of concrete during carbonation.	T = 25°C, RH = 50%, P = 0.7-1bar, CO_2 = 99.5%.	Mild	□Time: 2-4hr and 96h. □Mass of sorbent: 677g	The mixture composition is given (Cement = 88g, Water = 35g, Expanded Slag = 554g).	Three methods used to estimate carbon uptake: □Mass gain. □Mass curve. □Thermal decomposition analysis.	0.3-45.8gCO2/kg
Xiaoliang Fang, Dongxing Xuan, Chi Sun Poon. Empirical modellinf og CO_2 uptake by recycled concrete aggregates under accelerated carbonation conditions. 2017. doi: 10.1617/s11527-017-1066-y	Develop an empirical model to estimate the CO2 uptake of RCAs with different material characteristics in relation to various accelerated carbonation conditions.	Batch reactor.	RCA (Recycled Concrete Aggregates).	Pressurixed carbonation -> carbonation chamber was first vacuumed and then pure CO_2 was injected into the chamber.	100% CO_2, P = 0.5-0.1bar, T = 25°C. RH = 50 ± 5%.	Mild Very Intensive	□Time: 1, 3, 5, 7, 24hr were tested. □Mass of sorbent: not present.	The characteristics of the two types of RCAs (size (mm), Specific density (kg/m^3), Water absorption (%), Cement content (%)) are given.	Thermogravimetric methods.	□Mild (P = 0.1bar) 7; 12.8; 24; 19; 19 gCO2/kg; □Intensive (P = 5bar): 28 gCO2/kg
				Flow-through carbonation -> flow was a mixture of pure CO2 and air.	Flowrate = 1.0-10L/min, ambient T and P. RH = 50 ± 5%.					Mild: □flow through test: 5,8 to 12,4 gCO2/kg
Yxin Shao, Vahid Rostami, Zhen He, Andrew J. Boyd. Accelerated carbonation of Portland Limestone Cement. 2014. doi: 10.1061/(asce)mt.1943-5533.0000773	Study the accelerated carbonation of PLC to examine the possibility of replacing OPC (Ordinary Portland Cement) by PLC in carbonation	Batch reactor.	PLC (Portland Limestone Cement)	The initially air cured samples were placed in the chamber and carbon dioxide was injected.	CO_2 = 99.5%, P = 0.15MPa, T = 25°C, RH = 60%.	Very Intensive	□Time: 2hr. □Mass of sorbent: not present.	The chemical composition of PLC cement is known (Ca0 = 59.8%, SiO_2 = 20.6..).	□Mass gain method that compares the sample mass before and after carbonation. The difference represented the	121, 129, 128gCO2/kg

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	curing of precast concrete products.								carbon dioxide uptake. Infrared based carbon analyzer (CW800, Eltra).	
Sereng M., Djerbi A., Omikrine Metalssi O., Dangla P. and Torrenti J.. Accelerated carbonation of recycled concrete aggregates. 2021.	Obtain the maximum of CO_2 uptake on RCA with a simultaneous improvement of their properties, through the parametric study of carbonation parameters.	Slurry reactor	RCA	RCA carbonation test is carried out by setting a water content on dry RCA at the initial state through the immersion of RCA during 24 hours and after by a controlled drying in order to obtain the desired water content. In a second part, samples were kept in a desiccator as a curing chamber. Finally, at the end of accelerated carbonation test, RCA are dried at 80°C (until constant mass) to eliminate free water in RCA.	Grain size = 1-4mm, 10-20mm, CO_2 = 15% and 100%, Atmospheric Pressure.	Very Mild (CO_2 = 15%) and Mild (CO_2 = 100%).	Time: 24hr. Mass of sorbent: not present.	The Portlandite contents are determined by thermogravimetric analysis (NETZCH STA 449 F5 Jupiter). WAC (in percentage) is determined by difference between mass of RCA at SSD (Saturated Surface Dry) state (mSSD) and mass of dry RCA (mdried).	To determine CO2 uptake (in g/kg), a mass monitoring is achieved to study the weight gain after accelerated carbonation test. It is determined by the ratio between the mass of dry RCA before carbonation $m_{ini\ dried}$, and the mass of dry RCA after carbonation $m_{final\ dried}$.	Average values of 8.6, 17.3, 49.9, 10.4, 21.8, 8.5, 21.8, 6.5, 10.4, 14.9, 20.4 gCO_2/kg. (see figures of chapter 3)
Kaliyavaradhan S.K., Ling T.C., Mo K. H.. CO_2 sequestration of fresh concrete slurry waste: Optimization of CO2 uptake and feasible use as a potential cement binder. 2020. doi: 10.1016/j.jcou.2020.101330	The main objective of this work is to maximize CO2 uptake of the CSW waste powder by optimizing the carbonation process.	Open, Slurry reactor	Fresh CSW (Concrete Slurry Waste)	Some process conditions (T, RH and CO_2) were kept constant, and only the L/S ratio and reaction time were varied.	T = 20°C, RH = 65%, CO_2 = 20%, L/S = 0-0.7, grain size < 300µm.	Mild	Time: 1-168hr. Mass of sorbent: 10g.	The oxide composition was examined by X-ray fluorescence spectroscopy (XRF). The pH was measured using pH meter. The true density of CSW was measured by AccuPyc II 1340 series, micromeritics instrument. The particle size distribution by Mastersizer 2000,	The weight of CO_2 was quantified by TGA using a Rigaku TG-DTA 8121H. About 20mg ground sample was heated from room T to 1000°C at heating rate of 10°C/min in a nitrogen atmosphere with a flowrate of 30mL/min.	The maximum CO_2 uptake of CSW at optimazed conditions was 204.35g/kg.

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								Malven panalytical. □The mineralogical composition was analyzed by XRD. □The specific surface area, pore size and pore volume were analysed using VET and BJH method. □The leaching of eavy metals was determined using TCLP.		
Furcas C., Balletto G., Naitza S., Mazzella A.. Evaluation of CO_2 Uptake under Mild Accelerated Carbonation Conditions in Cement-Based and Lime-Based Mortars. 2014. doi: 10.4028/www.scientific.net/AMR.980.57	Explore the CO2 sequestration capacity of a cement-based (CM) and a lime-based mortar (LM) over the first 28 days of curing.	Batch reactor	Cement-based Mortar (CM)	Four samples were cast in four small cylindrical plastic molds and immediately sealed with para-film paper to prevent early carbonation. The experimental apparatus was kept under vacuum (in order to exclude the uptake of atmospheric gases), and the Sample Cell was charged allowing the reaction between CO_2 and the sample.	T = 25°C, P = 1bar, CO_2 = 99%.	Mild	□Time: 1, 7, 28days. □Mass of sorbent: 45g.	□The primary characteristics of CM and LM are given. □The mineral composition of the mortars was evaluated by powder XRD analysis.	X-ray diffraction (XRD) and calcimetry analysis (Dietrich-Frehling). CO2 content was measuerd also recording the differnce of pressure in the chamber by Redlich-Kwong (RKS) equation of state.	RKS -> 19.1, 25.5, 26.4 for 1, 7, 28d. Calcimetry -> 15.6, 24.0, 27.4 fro 1, 7, 28d.
			Lime-based Mortar (LM)							RKS -> 5.0, 11.0, 16.1, fro 1, 7, 28d. Calcimetry -> 2.3, 13.7, 18.1 for 1, 7, 28d.
Yi Z., Wang T., Guo R.. Sustainable building material from CO_2 mineralization slag: Aggregate for concretes and effect of CO_2 curing. 2020.	Provide an economical way of making full use of the CO_2 mineralization slag and CO_2 gas. Discusses the effects of CMS aggregate on CO2 sequestration capacity and mechanical property during CO2 curing.	Batch reactor	OPC (Ordinary Portland Cement)	Before CO2 curing, a vacuum is created in the chambers for 3 min through suction, and then the inlet valve is opened to supply the surge chamber with CO2 gas to a certain pressure. During the curing process, the pneumatic valve is used to regulate CO2 pressure in the curing chamber. Subsequently, when the curing process is	T = 40 ± 2°C, RH >= 90%, CO_2 = 99.9%, P = 0.5, 1.0, 1.5, 2.0, 2.5MPa. During the CO_2 curing process, P and T are recorded every 5s.	Intensive	□Time: up to 2hr. □Mass of sorbent: not present.	The chemical composition of CMS was detemined by XRF.	The calculation of the CO2 uptake is based on the actual gas equation.	It depends on the P and L/S. 57-160gCO2/kg

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				completed the exhaust valve is opened to balance the internal and external pressures on the curing chamber.						
Yuan Q., Yang G., Zhang Y., Wang T., Wang J., Romero C. E.. Supercritical CO$_2$ coupled with mechanical force to enhance carbonation of fly ash and heavy metal solidification. 2020.	Analyze the fly ashes absorption capacity.	Batch reactor	Coal-fired plant fly ashes	The carbonation experiments of fly ash were carried out on a supercritical CO$_2$ platform. Dried samples were put in a Teflon container of an autoclave, and a certain amount of deionized water was added according to the liquid-to solid ratio and then the air tightness was checked.	Slurry route. L/S = 1, 5, 10, 20, 30, 50, 100, 200, 30mL/g, T = 25, 40, 60, 80°C, P = 1, 8MPa.	Intensive	□Time: up to 24hr □Mass of sorbent: 10g	The composition of the raw ashes is given. Mainly composed of SiO$_2$, CaO, Al$_2$O$_3$, Fe$_2$O$_3$)	The carbonation efficiency of the fly ash was tested by a thermogravimetric analyzer (SDT, model: Q60).	□P = 1MPa -> 27.5gCO$_2$/kg$_{fly\_ashes}$ □P = 8MPa -> 39.4gCO$_2$/kg$_{fly\_ashes}$
Suescum-Morales D., Fernandez-Rodriguez J. M., Jimenez J. R.. Use of carbonated water to improve the mechanical properties and reduce the carbon footprint of cement-based materials with recycled aggregates. 2022	The evolution of the carbon sequestration capacity of porous CBMs made with NA or RMA at early curing ages using normal or carbonated water as the mixing and/or curing solvent were investigated.	Batch reactor	□RMA (Recycled Masonry Aggregate) from a recycling plant in Cordoba. □Siliceous NA (Natural Aggregate). □Portland Cement.	Air Curing -> Normal climatic chamber was used.	CO2 = 0.04%, RH = 65 ± 10%, T = 21 ± 2°C.	Mild	□Time: 1, 3, 7d. □Mass of sorbent: 2574g (NA), 3507g (RMA).	□Chemical composition: Wavelength dispersive X-ray fluorescence spectrometry (ZSX PRI MUS IV, Rigaku, 4 kW power). □Morphology and composition of the scalactites: , scanning electron microscopy (SEM) and mapping were performed with a JEOL JSM 7800 F. □The acidities of water curing were measured with an MM40 digital pH meter (CRISON Instruments) at the ages of 1, 3, 7d.	Thermogravimetric analysis (TGA) and Differential Thermal Analysis (DTA).	Values between 1,16 to 16,5gCO2/kg
		Slurry reactor		Water curing -> small individual containers (V = 3L).	ŸŸT = 21 ± 2°C.					

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Rostami V., Shao Y., Boyd A. J.. Durability of concrete pipes subjected to combined steam and carbonation curing. 2011. doi: 10.1016/j.conbuildmat.2011.03.025	Improve durability performance of concrete pipes and explore the possibility of using concrete pipe to sequester carbon dioxide.	Batch reactor	Concrete pipes.	In the carbonation setup the CO2 of 99.5% purity, simulating the recovered CO2 from industrial point sources, was injected into a sealed chamber and the interior pressure was regulated at a constant of 1.5 bar so that the consumed CO2 could be replenished. Using a digital balance, the mass gain due to CO2 uptake was recorded throughout the carbonation process.	CO2 = 99.5%, P = 1.5bar, T = 24°C, RH = 30%.	Very Intensive	Time: 2hr. Mass of sorbent: not present.	The mixtures were proportioned using ordinary Portland cement (CSA Type GU) at 286 kg/m3 , coarse aggregates 730 kg/m3 , fine aggregates 1050 kg/m3 and water 100 kg/m3 .	Mass curve and Mass gain methods.	87, 89, 79 g/kg
Zhang D., Shao Y.. Early age carbonation curing for precast reinforced concretes. 2016. doi: 10.1016/j.conbuildmat.2016.03.048	Develop an early carbonation curing process for wet-mix precast concrete with high slump.	Batch reactor, Slurry reactor.	OPC (Ordinary Portland Cement)	The conditioned concrete was carbonated in a pressure chamber with a CO2 gas of 99.8% purity and at varied duration and pressure.	L/S = 0.4, 0.3. CO2 = 99.8%, RH = 65 ± 5%, T = 25 ± 3°C, P = 1-5bar. PH was recorded in order to monitor carbonation activity.	Intensive	Time: 12hr. Mass of sorbent: not present.	The mixture proportions of concrete are given.	Mass curve and Mass gain methods.	Mass gain method: 101.3, 158.1 gCCO2/kgdrycementcontent Mass curve method: 113.3, 166.9 gCO2/kgdrycementcontent From 1 to 5bars: 99.5 to 146.0 gCO2/kg
Dos Reis G. S., Cazacliu B., Artoni R., Torrenti J., Hoffmann C. S., Lima E. C..Coupling of attrition and accelerated carbonation for CO2 sequestration in recycled concrete aggregates. 2021. doi: 10.1016/j.clet.2021.100106.	Investigate such a process coupling attrition and carbonation for CO2 sequestration and to improve coarse RCA.	Batch reactor with rotating drum	Coarse RCA (Recycled Concrete Aggregate)	Dry concrete aggregates were simply immersed in water for a given time to prepare samples with different initial moisture values. The system consisted of a CO2 cylinder, a vacuum pump, a controlled injector of CO2 and horizontal drum	P = 0.75bar, T = 22°C, CO2 = 100%	Mild	Time: 1 to 7d. Mass of sorbent: 500g.	Particle size distribution: sieving, while a VDG40 video grader determined the evolution of the particle shape according to the standard French XP P 18-566-2002. Chemical composition: EDX analysis was obtained through a scanning electron microscope (SEM) equipped with	Analyze the CO2 capture performance through the difference (increase) in the dried mass before (mini dried) and after (mfinal dried) the tests.	Values between 7.5 and 79.5gCO_2/kg

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				vessel totally air-proof.				energy-dispersive X-ray spectroscopy (TESCAN 3, Sweden). Physical properties: the concrete aggregates' density and water absorption were determined according to NF EN 1097–6.		
Schnabe K., Bruck F., Pohl S.. Technically exploitable mineral carbonation potential of four alkaline waste materials and effects on contaminant mobility. 2021. doi: 10.1002/ghg.2063	Examine the CO2 sequestration potential of four dry-discharged waste materials using theoretical and experimental approaches.	Batch reactor	Biomass Bottom Ash (BBA) Biomass Fly Ash (BFA) Fly ashes from refuse-derived fuel incineration (RFA) Stainless Steel Slag (SSS)	Tests were conducted in a fixed bed setup. The reactor was a gas-tight glass vessel equipped with a sample rack. The reactor was instrumented with a piezoresistive pressure sensor (OxiTop, WTW, Weilheim, Germany) with a tolerance of ± 1 hPa to record the pressure drop induced by CO2 uptake and with wireless temperature/humidity sensors.	CO2 = 100%, L/S = 0.1, 0.2, 0.3, 0.4, 0.5, Atmospheric pressure, grain size < 3mm.	MIld	Time: 168hr. Mass of sorbent: 110 ± 10g.	Moisture content determined gravimetrically. ANC determined by titrating aqueous suspensions using a Mettler-Toledo T70 titrator equipped with a DGi111-SC pH-electrode. Elemental composition determined by XRF using a SpectroScout ES with a Motex Magnum TSR-B5008-RH2 X-ray source. Mineral phases identified using XRD. Solid phase constituents quantified with TGA.	The CO2 sequestration potential of the materials was alternatively calculated using the ANC (Acid Neutralization Capacity) and StE (Steinour equation).	BBA -> 92,84gCO_2/kg BFA -> 43,56 gCO_2/kg RFA -> 76,56 gCO_2/kg SSS -> 111.76 gCO_2/kg
Suescum-Morales D., Kalinowska-Wichrowska K., Fernandez J.M., Jimenez J. R.. Accelerated carbonation of fresh cement-based products containing recycled masonry aggregates for CO_2 sequestration. 2021. doi: 10.1016/j.jcou.2021.101461	Investigate how accelerated carbonation effects on the mechanical properties of a porous cement-based material made replacing natural sand with different percentages by volume	Batch reactor	RMA (Recycled Masonry Aggregates) Natural Aggregates (NA)	The samples were cured under two hardening environments and tested at three different ages.	Climatic Chamber: CO2 = 0.04%, RH = 65 ± 10%, T = 21°C ± 2°C, atmospheric pressure.	Very Mild	Time: 1, 3, 7days. Mass of sorbent not present.	RMA composition (red ceramic, mortar…) determined according to UNE-EN 933-11:2009. Particle size distribution obtained according g to UNE-EN 933-1:2012 before	Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) were performed for the raw materials and hardened mortar samples at 1, 3,	Values between 1.573 - 26.35 gCO_2/kg sorbent. (See Table n.7)

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	(0, 50, and 100 %) of the fine fraction of RMA.				Accelerated carbonation chamber: CO2 = 5%, RH = 65 ± 10%, T = 21°C ± 2°C, atmospheric pressure.			discarding the particles greater than 4 mm. □ Sand equivalent, dry bulk density, and water absorption were measured according to the following European standards: UNE-EN 933- 8:2012+A1 , UNE-EN 1097-6:2013 and UNE-EN 1097-6:2013. □Chemical composition studied with XRF. □Mineral composition determined with XRD.	and 7 days of curing. The amount of CO$_2$ sequestered by the aggregates was determined by the difference between the quantity of carbonate content after and before CO$_2$ exposure.	
Dos Reis G. S., Cazacliu B. G., Artoni R., Torrenti J.. Effect of the accelerated carbonation treatment on the recycled sand physicochemical characteristics through the rolling carbonation process. 2020. doi: 10.1016/j.jcou.2020.101181	Proposes an efficient method for accelerating the carbonation process, called “rolling carbonation” for improving the physicochemical properties of the recycled sand made from the inert part of CDW. Provides a new alternative to solve some pro blems that are associated with reusing the inert part of CDW as recycled sands.	Batch reactor	Recycled sand	The scheme of the experimental setup used in this work consists of a CO$_2$ loading system composed of a vacuum pump (for the first loading), a CO$_2$ cylinder and a mass flow controller for dosing the gas into the drum (before rotation); There is also a drum, lying on free rollers, which is directly set in motion by the motor.	T = 22°C, P$_1$ = 1.014, P$_2$ = 1.4bar, V$_{rotation}$ = 20, 60, 100rpm, CO$_2$ $_{Partial\ Pressure}$ varied from 0.58bar to 1.2bar.	Mild	□Time: 7h □Mass of sorbent: 500, 1500g	□Morphology of the recycled sands observed with a scanning electron microscope (SEM) (TESCAN 3, Sweden). □Functional groups of the recycled sands were determined using a Fourier Transform Infrared Spectroscopy (FTIR) with the ATR (Attenuated Total Reflectance) accessory, Bruker Spectrometer, alpha model. □Thermogravimetric (TGA) and Differential thermogravimetric (DTG) analyses were made by using a TA Instruments model SDT Q600	The mass increase by carbonation, i.e. the difference between the dry mass of the aggregates before (mO) and after (mF) carbonation, was measured to evaluate the carbonation level after the accelerated test by introducing a correction coefficient α considering the stoichiometry.	Values between 4.2 - 47.4 gCO$_2$/kg sorbent. (See Table n.2)
Berber H., Tamm K., Leinus M., Kuusik R., Tonsuaadu K., Paaver P., Uibu M.. Accelerated carbonation technology granulation of industrial waste: Effects of mixture composition on product properties. 2019. doi: 10.1177/0734242X19886646	Aims to simplify the ACT granulation process by carrying out premixing, hydration, accelerated carbonation, and combined carbonation–	Open, Slurry reactor	Different mixtures of: MSWI FA, MSWI APC residues, OSA samples, CKD,	Different mixtures were tested by ACT granulation in order to optimise the granule composition by varying the share of MSWI ash from 100% to 1%.	Room temperature, CO$_2$ = 20%. Flow rate = 100L/h First step: L/S = 0.2 - 0.3w/w.	Mild	□Time = 30min + 20 min □Mass of sorbent = 200g	□The Brunauer, Emmett, and Teller (BET)- specific surface area (SSA) was determined using a Kelvin 1042 sorptometer. □The particle size	The carbon dioxide binding degree (BDCO$_2$), which is considered to be one of the main indicators of carbonation efficiency, was	Values from 23 to 108gCO$_2$/kg$_{waste.}$ (See Table n. 3)

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	granulation of wastes in the same apparatus. The main focus of the current study is to incorporate hazardous MSWI ash into mixtures of OSA, CKD, and quarry fines in order to produce granulated products that could be reused or declassified.		Quarry fines, and OPC.	Premixing, hydration, and carbonation (Step 1), followed by combined carbonation-granulation (Step 2), were all carried out in an Eirich GmbH and Co EL1-type intensive mixer/granulator by operating under different regimes.	Second step: L/S = 0.2 - 0.5w/w.			distribution (PSD) was determined using a Horiba LA-950 laser-scattering PSD analyser. The chemical composition was determined by using both X-ray fluorescence (XRF) spectroscopy (Rigaku Primus II) and quantitative X-ray diffraction (XRD)(Bruker D8 Advanced) methods.	calculated by ta ratio between $CO_{2granules}$ and $CO_{2\ max}$, where CO2granules is the analytically determined CO2 content of the granules and CO_{2max} is the maximum possible CO_2 content of the granulated mixture of ash, OPC, and quarry fines in percentage.	
Wang D., Zhu J., He F.. Quantification and micro-mechanisms of CO2 sequestration in magnesia lime-fly ash/slag solidified soils. 2019. doi: 10.1016/j.ijggc.2019.102827.	This study attempts to propose two approaches to evaluate the CO2 uptake amount of reactive magnesia-lime-fly ash/slag solidified soils, rather than soil improvement proved by previous findings.	Batch reactor	Fly Ashes	The carbonation device for soil specimens consists of three parts, i.e. confining pressure loading system, CO2 circulation system and carbonation test platform.	T = 20°C ± 2°C, RH > 95%, CO_2 pressure = 150kPa, CO_2 = 100%, confining pressure = 300kPa.	Intensive	Time = 0.5, 1, 3, 6, 12, 24h Mass of sorbent = not present.	Physical parameters are determined based on the Chinese standard. Chemical properties are given.	CO2 uptake efficiency defined by direct weight gain and by indirect TGA/DSC analysis.	t = 6h -> values from 4.87 to 13.42gCO_2/kg. t=24h -> values from 5.72 to 14.66gCO_2/kg.
Kim J. H., Kwon W. T.. Semi-Dry Carbonation Process Using Fly Ash from Solid Refused Fuel Power Plant. 2019. doi: 10.3390/su11030908	This study investigated the utilization of fly ash as a solid sorbent material for CO2 capture via semi-dry carbonation reaction illustrating the possibility of replacing the wet process with the semi-dry process.	Open reactor	Fly Ashes	CO_2 capture was conducted in a round-bottom flask with a single neck. The mixture of fly ash and water was delivered into the flas. The rotation speed was fixed to shake the mixture and the CO_2 stream was flow into the rector using the regulator and the flow meter to control the CO_2 concentration.	RH = 25, 50, 75, 100%, $V_{rotation}$ = 40rpm, CO_2 = 99.99%, Flow_rate = 10L/min, T = 25°C.	Mild	Time: 60min Mass of sorbent: 200g	The morphologies and structures were analyzed by SEM (JSM-6700F). The crystal structures were investigated by XRD (X' pert3 powder). The particle size was measured using a lased difraction particle size analyzer (PSA) (Mastersizer S Ver. 2.15). The elemental compositions were investigated using XRF (Primus2).	Stechimetric equation	101.7 gCO_2/kg

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Pei S., Pan S., Gao X., Fang Y., Chiang P.. Efficacy of carbonated petroleum coke fly ash as supplementary cementitious materials in cement mortars. 2018. doi 10.1016/j.jclepro.2018.01.055	Investigate the changes in physico-chemical properties of clinker before and after accelerated carbonation, examine the feasibility of utilizing treated PCFA as SCMs with respect to composition, workability and durability, determine the improvement of blended cement substituted with carbonated PCFA by mathematic models and estimate the environmental and economic benefits of waste-to-resource supply chain in the case of PCFA.	Slurry, RPB reactor	Fly Ashes (PCFA)	Accelerated carbonation of PCFA was performed in a petrochemical plant whose exhausted stream provided CO2 for carbonation reaction. The major component of the carbonation equipment is a rotating packed bed (RPB) reactor, which can generate a high centrifugal force via the high-speed rotation, thereby resulting in an enhanced mass transfer rate between liquid and gas phases.	Ambient pressure and temperature, L/S = 10m^3/kg, flowrate = 0.2m$_3$/min, carbonation completed as soon as pH dropped below 6.3.	Mild	☐Time: not present ☐Mass of sorbent: not present	The compositions of PCFA were examined by X-Ray Fluorescence (XRF) and chemical titration analysis.	The amount of CO2 fixation on PCFA (noted as ThCO2) can be calculated by the Steinour equation	84.5 gCO$_2$/kg
Ulkwattage N.L., Ranjith P.G., Li X.. Steel-making slag for mineral sequestration of carbon dioxide by accelerated carbonation. 2016. doi: 10.1016/j.measurement.2016.10.057	Investigate the effect of three process parameters (pressure, temperature and water-to-solid ratio) on the rate and efficacy of the mineral sequestration of steel-making slag at laboratory-scale.	Slurry, batch reactor.	Steel slags	Carbonation tests were carried out inside a laboratory-scale reactor connected to a CO2 gas source through a plumbing system. The test set-up consists of (1) a CO2 supply, (2) a reactor cell, (3) a heating unit, (4) a stirring unit and (5) a monitoring and recording unit.	L/S = 0.25, 0.5, 1, 2, 3, CO_2 pressure = 1, 2, 3, 4, 5, 6MPa, T = 20, 50, 80°C, $V_{rotation}$ = 60rpm.	Intensive	☐Time: 48h ☐Mass of sorbent: 100g	Mineral composition analyzed with XRF method	In order to quantity the CO2 sequestration, CO2 pressure reduction during the tests was used in the ideal gas law equation.	29.47gCO$_2$/kg

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Lombardi L., Carnevale E.A., Pecorini I.. Experimental evaluation of two different types of reactors for CO_2 removal from gaseous stream by bottom ash accelerated carbonation. 2016. doi: 10.1016/j.wasman.2016.09.038	Investigate the possibility of improving the way the contact between bottom ash and landfill gas takes place.	Fixed bed reactor	Bottom Ashes	Stainless steel cylindrical tank. The reactor can be opened from the top, by means of a semispherical cap,for loading and unloading the BA. The gas flows into the reactor from the bottom to the top (upflow configuration).The ashes are placed on one/three drilled plates sustained by dedicated supports and covered by a geotextile fabric which retains the small particles.	CO_2 = 100%, Room Temperature, Atmospheric Pressure, Gas_flowrate = 400Nml/min, Particle size = 5, 10mm, number of layers = 1, 3.	Mild	Time: 8 - 14h Mass of sorbent: 6kg	The moisture content was measured after drying for 24h at 105°C. For pH measurement, the BA was dried, reduced in size, sieved at 5mm and mixed in the ratio of 10g of BA to 100mL of distilled waster (UNI EN 12457-2). The pH was measured by means of a pH-meter.	The amount of removed CO2 was estimated by means of a mass balance, i.e. from the difference between the inlet and the outlet volume of gas, at a given time of the experiment. The calcium car bonate content of BA samples collected before and the end of each test was also measured by calcimetry analysis using a Dietrich Früling calcimeter.	Single layer and 10mm: 24 - 48 gCO_2/kg. Single layer and 5mm: 34 - 46g/kg. 3 layers and 10mm: 27 - 47g/kg. 3 layers and 5mm: 34 - 48g/kg
		Rotating drum reactor		Provides continuous mixing of the solids. A large circular opening 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.	CO_2 = 100%, Room Temperature, Atmospheric Pressure, Filling Ratio (FR) = 10, 20, 30%, Rotating Speed (RS) = 2.5, 5rpm.		Time: 8 - 14h Mass of sorbent: 3, 6, 9kg			RS = 2.5rpm and FR = 10%: 33 - 37g/kg. RS = 2.5rpm and FR = 20%: 25 - 31g/kg. RS = 2.5rpm and FR = 30%: 19 - 23g/kg. RS = 5rpm and FR = 20%: 25 - 31g/kg
Xuan D., Zhan B., Poon C. S.. Development of a new generation of eco-friendly concrete blocks by accelerated mineral carbonation. 2016. doi: 10.1016/j.jclepro.2016.06.062	Accelerated mineral carbonation as an alternative method to produce a new generation of eco-blocks in Hong Kong is the subject.	Batch reactor	Concrete blocks made by RCA + PC + water	In this study, an airtight steel-cylindrical chamber with a volume of approximately 100 L, controlled under a maximum gas pressure, was used. Before CO2 gas was	CO_2 > 99.5%, P = 0.1bar and 5bar, T = 25°C, RH = 50% for 6h	Mild. Intensive.	Time: 1, 2, 3, 6, 18, 24h Mass of sorbent: not present.	The properties of all aggregates were determined by BS EN 1097-6.	Thermal analysis by using a furnace device was performed to measure the amount of CO2 sequestration by the samples.	0.1bar -> 165gCO_2/kg_{cement} 5bar -> 180gCO_2/kg_{cement}

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				injected, the chamber had been vacuumed to 0.6 Bar by a vacuum pump.						
Xuan D., Zhan B., Poon C. S., Zheng W.. Carbon dioxide sequestration of concrete slurry waste and its valorisation in construction products. 2016. doi: 10.1016/j.conbuildmat.2016.03.109.	Investigate the CO2 sequestration potential of concrete slurry waste (CSW) and its valorisation with fine recycled concrete aggregates (FRCAs) for the production of sustainable construction products by direct gas-solid mineral carbonation.	Batch reactor	Concrete Slurry Wastes (CSW) FineRCA (FRCA)	The CSWs samples, FRCAs and the prepared mixture specimens were carbonated in an airtight stainless steel vessel with a volume of about 33L. Before the CO2 gas was injected in the vessel, it was vacuumed. A commercially sourced CO2 gas was then injected in the vessel. The pressure in the chamber was controlled by a gas regulator.	$CO_2 > 99.5\%$, P = 0.1bar, T = 25°C, RH = 50% for 6h	Mild	□Time: 1, 3, 24, 48, 96, 144h. □Mass of sorbent: not present.	□The elemental compositions of CSWs, FRCAs and cement were examined by using X-ray fluorescence spectroscopy (Element analyser, JEOL JSX-3201Z). □The surface morphologies of the samples before and after min eral carbonation were observed under a scanning electron micro scopy with energy dispersive X-ray spectroscopy (SEM-EDX, JEOL Model JSM-6490). □X-ray diffraction analysis (XRD, Rigaku Smartlab) was conducted.	A thermogravimetric analyser (TGA/DSC, Netzch STA 449C, Jupiter) was employed to determine the overall carbonate content in the samples	□110gCO_2/kg_{dryCSW}. □52$gCO_2/kg_{mixture}$. □20gCO_2/kg_{FRCA}.
Chang E-E., Pan S-Y., Yang L., Chen Y-H., Kim H., Chiang P-C.. Accelerated carbonation using municipal solid waste incinerator bottom ash and cold-rolling wastewater: Performance evaluation and reaction kinetics. 2015. doi: 10.1016/j.wasman.2015.05.001	Evaluate the performance of aqueous carbonation using MSWI-BA coupled with cold-rolling wastewater (CRW) via a slurry reactor for CO2 fixation.	Slurry reactor	MSWI-BA (Municipal Solid Waste Incinerator Bottom Ash) with CRW (Cold-rolling wastewater).	The ground MSWI-BA was sieved into three categories and dried to eliminate moisture. The MSWI-BA was stored in small air tight containers and then placed in a larger capped container. In addition, high-pressure CO2 gaswas	$CO_2 = 99\%$, L/S = 5, 10, 20, 40mL/g. CO_2 Pressure = 1atm, Flow rates = 0.2, 0.5, 1.0, 1.5 L/min, Room Temperature.	Mild	□Time: 120min. □Mass of sorbent: not present.	□The chemical properties were measured by ASTM method C114 using X-ray fluorescence (XRF, PW2430, Philips, Netherland). □The densities and BET surface areas were analyzed by a pycnometer (Accupyc 1340, Micromeritics Instrument Corporation, USA) and	The carbonation conversion of the MSWI-BA before and after carbonation reaction were examined by a thermogravimetric and differential scanning calorimetry (TG–DSC, STA6000, PerkinElmer, USA).	max value of 102g/kg.

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				supplied. CRW produced from a steel manufacturer was used as the liquid agent during the carbonation reaction in a slurry reactor.				a low-temperature N2-adsorption BET apparatus (ASAP2010, Micromeritics Instrument Corporation, USA), respectively. □The particle size distribution (PSD) of the MSWI-BA was obtained by laser diffraction (Beckman Coulter LS 230), which was adapted from the ISO 13320:2009 method. □The mineralogical characteristics of MSWI-BA before and after the carbonation were determined by XRD.		
Ulkwattage N.L., Ranjith P.G., Yellishetty M., Bui H.H., Xu T.. A laboratory-scale study of the aqueous mineral carbonation of coal fly ash for CO2 sequestration. 2014. doi: 10.1016/j.jclepro.2014.03.005	Investigate the effect of two reaction parameters (fly ashewater content and temperature) on the mineral CO2 sequestration of three types of Australian brown coal fly ashes.	Continuosly stirred autoclave reactor	Coal fly ashes	The bulk material was sieved with a 1.18 mm sieve to obtain uniform-sized particles for testing. The samples were then oven dried at 105 °C to a constant weight, to remove all the moisture in the samples. Carbonation tests were carried out inside continuously stirred autoclave reactors connected to a plumbing system. The system included a reactor cell, gas inlet unit, gas outlet unit, heating unit, stirring unit and a data acquisition unit.	L/S = 0.1, 0.5, 1, at CO_2 Pressure = 3MPa and T = 40°C. v_{mixing} = 60rpm T = 20, 40, 60, 80°C at L/S = 0.3 and CO2 Pressure = 3MPa. v_{mixing} = 60rpm.	Intensive	□Time: 10h. □Mass of sorbent: 300g	Morphological analysis was done using a scanning electron microscope Nova Nano 450 which is equipped with a field emission gun. The mineralogy of the fly ashes was determined by X-ray diffraction spectroscopy using a PANalytical Empyrean diffractometer coupled with Cu-Ka radiation at 45 kV and 40 mA. ŸElemental analysis was conducted using the X-ray fluorescence method. ŸThe Brunauer, Emmett and Teller (BET) single point method with N2 was used to measure the surface area of the particles. ŸThe absolute and bulk	The carbonation Pressure drops were measured at each time interval. The pressure reduction data were converted to the mole number of CO_2 sequestered in each case, by applying ideal gas law.	10.71, 20.13, 27.05 gCO_2/kg.

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								densities were determined using a Micromeritics Accupyc 1330 helium pycnometer and Micromeritics Autopore 9520 mercury porosimeter, respectively.		
Morone M., Costa G., Polettini A., Pomi R., Baciocchi R.. Valorization of steel slag by a combined carbonation and granulation treatment. 2013. doi: 10.1016/j.mineng.2013.08.009	Evaluate the applicability of combined granulation and carbonation as a valorization technique for BOF slag and to compare the effects of this process with those exerted by granulation under atmospheric air, so to analyze the influence of carbonation on particle aggregation and on the properties of the products	(Laboratory scale granulator) Slurry reactor	BOF (Basic Oxygen Furnace) Steel Slags.	Granulation–carbonation experiments were performed using a 180 W laboratory scale granulator with a diameter of 0.3 m and a height of 0.23 m, equipped with a blade. Air-dried slag and deionized water were premixed in a plastic bag, at the liquid to solid ratio that resulted to be optimal for the granulation of this material at the tested conditions. In order to obtain a homogeneous initial particle size distribution for all the experiments, the mixture was then pushed through a 2 mm sieve.	L/S = 0.12, CO_2 = 100%, P = 1bar, T = 23 ± 2°C, v_{mixing} = 24rpm.	Mild	Time: 20, 60, 90, 120min. Mass of sorbent: approximately 500g.	The main chemical constituents of the BOF slag are determined by alkaline fusion with $Li_2B_4O_7$ at 1050°C and ICP-OES analysis. The particle density was assessed applying the UNI EN 1097-6 procedure. The mineralogical composition was determined by powder XRD analysis with Cu Ka radiation using a Philips Expert Pro diffractometer. The carbonate content was evaluated by Inorganic Carbon analysis using a Shimadzu TOC VCPH analyser equipped with a SSM-5000A solid sampler.	The extent of carbonation of the slag, resulting from the reaction of the alkaline phases with either atmospheric CO2 (granulation tests) or pure CO2 (granulation–carbonation tests), was assessed by measuring the Inorganic Carbon (IC) content of the material before and after each treatment.	values beetwen 120 and 149 gCO_2/kg
Jiang J., Tian S., Zhang C.. Influence of SO2 in incineration flue gas on the sequestration of CO2 by municipal solid waste incinerator fly ash. 2013. doi: 10.1016/S1001-0742(12)60142-9	The influence of CO_2 content and presence of SO_2 on the sequestration of CO_2 by municipal solid waste incinerator (MSWI) fly ash was studied by investigating the carbonation reaction of MSWI fly ash with	Open reactor	MSWI (Municipal Solid Waste Incinerator) Fly Ashes.	The fly ash was skived into smaller particles which could pass through a 200-mesh screen. By applying a continuous reaction laboratory simulated system, the simulated reaction gas flowed continuously from	CO2 = 12% and 100%, instantaneous flow = 0.01L/min, ambient temperature, L/S = 0.2	Mild	Time: up to 100min. Mass of sorbent: 100g and 150g.	Microstructure analysis of the BET specific surface area, T-plot micropore volume, and BJH adsorption pore volume (pore radius, 1.7–300 nm) of the fly ash were analyzed using	Thermo-gravimetric analysis (TGA) and differential scanning calorimetry (DSC) were used to investigate CO_2 uptake into and release from fly ash.	100% CO_2 -> 87gCO_2/kg 12% CO_2 -> 41gCO_2/kg

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	different combinations of simulated flue gas.			the bottom of the reactor to react with the fly ash sample.				ASAP2010 (Micromeritics Co., USA). The elemental compositions of the raw fly ash and the carbonated fly ash after CO2 capture were analyzed by using XRF (XRF-1800 analyzer, Shimadzu, Japan).		
Ulkwattage N.L., Ranjith P.G., Wang S.H.. Investigation of the potential of coal combustion fly ash for mineral sequestration of CO2 by accelerated carbonation. 2013. doi: 10.1016/j.energy.2012.12.048	Evaluate the potential for CO_2 sequestration of coal fly ash and to study the effect of CO_2 pressure and solid-to-liquid ratio on the rate and the efficiency of CO_2 sequestration.	Slurry reactor	Coal Fly Ashes	The ash samples initially contained large clods of ash and needed to be broken into individual particles for testing. The aqueous mineral carbonation reaction was carried out in side a pressure chamber connected to a CO_2 gas inlet line which provides it with the gas from a cylinder which is pressurized by the pump to the required pressure.	T = 40°C, L/S = 0.0 to 0.7, CO_2 Pressure = 2MPa to 6MPa.	Intensive	Time: 10h. Mass of sorbent: 300g.	The chemical composition was analyzed by X-ray spectroscopy analysis.	Since the volume of water and all the other operational parameters are the same, the pressure drop due to carbonation (P carbonation) can be found from the difference between P_{total} and P_{water}. The sequestered quantity of CO_2 by mineralization can be quantified using the ideal gas law considering that CO_2 is an ideal gas.	7.66gCO_2/kg
Nam S.-Y., Seo J., Thriveni T., Ahn J.-W.. Accelerated carbonation of municipal solid waste incineration bottom ash for CO2 sequestration. 2012. doi: 10.1080/12269328.2012.732319	Determine the degree of influence of several main factors, such as the optimum temperature, solid-to-water ratio and CO2 concentration, on the process of accelerated carbonation of the bottom ash.	Slurry reactor, batch reactor.	MSWI-BA (Municipal Solid Waste Incinerator Bottom Ash)	The ash was dried and the ferric metal was removed through a magnetic separation process. The bottom ash was sieved by mechanical shaking using stainless steel mesh screens with openings that meet the DIN4188 standards. Then the carbonation occurred in a double-walled sealed cylinder-shped reactor.	Humidity = 75%, CO_2 =10 - 30%, L/S = 0.1 - 0.3, T = 20, 40, 60°C.	Very Mild/Mild	Time: up to 25min. Mass of sorbent: 100g	Chemical composition is given.	The amounts of CO_2 sequestrated were calculated theoretically, based on chemical composition of the bottom ash, and the experimental amounts of CO_2 sequestrated were determined by direct CO_2 reaction.	3, 4, 5, 18, 50, 84, 165 gCO_2/kg

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Chang E.-E., Pan S.-Y., Chen Y.-H., Chu H.-W., Wang C.-F., Chiang P.-C.. CO_2 sequestration by carbonation of steelmaking slags in an autoclave reactor. 2011. doi: 10.1016/j.jhazmat.2011.08.006	Investigate the carbonation of several steelmaking slags, including ultra-fine (UF) slag, fly-ash (FA) slag, and blended hydraulic slag cement (BHC), in an autoclave reactor.	Autoclave reactor.	Steel slags (UF, FA and BHC)	The aqueous carbonation of UF slag, FA slag, and BHC were conducted in an autoclave reactor that contained distilled water at a designated temperature.	T = 40 - 160°C, L/S = 10, CO2 Pressure = 700psi (5MPa) - 1300psi (9MPa), Grain size < 44µm.	Intensive	□Time: 5min to 12h. □Mass of sorbent: not present	□The chemical compositions were characterized by inductively coupled plasma atomic emission spectroscopy (ICP-AES). □The contents of each metal in the extracted solution were measured by the ICP-AES method.	The conversion of the carbonation products was determined quantitatively by thermogravimetric analysis (TGA) (TGA-51,Shimadzu) and qualitatively by X-ray diffraction (XRD) (X' Pert Pro, PANalytical) and scanning-electron microscopy (SEM) (JSM-6500F, JEOL).	UF slag -> 127gCO_2/kg FA slag -> 107gCO_2/kg BHC slag -> 283gCO_2/kg
Dundar B., Tugluca M. S., Ilcan H., Sahin O.. The effects of various operational- and materials-oriented parameters on the carbonation performance of low-quality recycled concrete aggregate. 2023. doi: 10.1016/j.jobe.2023.106138	Examination of the parameters affecting the carbonation performance of low-quality recycled concrete aggregates (RCA) obtained from end-of-life buildings and having low mechanical and chemical properties.	Batch reactor.	RCAs (Recycled Concrete Aggregates)	A lab-scale carbonation reactor was used to provide an accelerated carbonation process for low-quality RCAs. The reactor was designed to provide a reliable, controlled environment by arranging the relative humidity, pressure, temperature, and CO_2 concentration for the carbonation reaction. . In order to keep the CO2 contact at a high level and to provide homogeneous carbonation, a mixer providing rotational mixing was designed for the carbonation reactor.	Case 1 - Humidity variation: RH = 50, 70, 90%. T = 90°C, P = 1bar, Grain Size = 2,00 - 4,75mm, CO_2 = 15%, $v_{Rotation}$ = 1rpm.	Intensive	□Time: 48h. □Mass of sorbent: not present	□A scanning electron microscopy (SEM) was used to examine morphological changes because of the carbonation process. □The crystalline structures of carbonated and uncarbonated RCAs were assessed by X-ray diffraction (XRD) analysis. □The compressive strength tests were performed on the 7-day and 28-day-old 50 mm-cubic specimens to evaluate the differences in the mechanical performance of mortar mixtures containing carbonated and uncarbonated RCAs.	Thermogravimetry (TG/DTG) analysis was performed to assess the chemical composition of carbonated and uncarbonated RCAs by focusing on the consumption of calcium hydroxide (CH) and the formation of different forms of calcium carbonate (CC).To calculate the CO2 uptake rate of RCAs after the accelerated carbonation process, the differences between CC amount of carbonated and uncarbonated RCAs were considered.	RH = 50% -> 37,2gCO_2/kg RH = 70% -> 74gCO_2/kg. RH = 90% -> 63,6gCO_2/kg.
					Case 2 - Temperature: T = 50, 90°C. RH = 70%, P = 1bar, Grain Size = 2,00 - 4,75mm, CO_2 = 15%, $v_{Rotation}$ = 1rpm.	Intensive	□Time: 2, 4, 6, 12, 24, 48, 72, 96, 120h. □Mass of sorbent: not present			T = 50°C: 2h -> 15gCO2/kg. 4h -> 22gCO2/kg 6h -> 25gCO2/kg. 12h -> 35gCO2/kg. 24h -> 50gCO2/kg. 48h -> 74gCO2/kg. 72h -> 76gCO2/kg. 96h -> 77gCO2/kg. 120h -> 78gCO2/kg. T = 90°C: 2h -> 22gCO2/kg. 4h -> 30gCO2/kg 6h -> 35gCO2/kg. 12h -> 42gCO2/kg. 24h -> 55gCO2/kg. 48h -> 75gCO2/kg.

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										72h -> 77gCO2/kg. 96h -> 78gCO2/kg. 120h -> 79gCO2/kg.
					Case 3 - Pressure: P = 1, 2, 3bar. RH = 70%, T = 90°C, Grain Size = 2,00 - 4,75mm, CO_2 = 15%, $v_{rotation}$ = 1rpm.	Intensive	Time: 2, 4, 6, 48h. Mass of sorbent: not present			Duration = 2h: P = 1bar -> 23gCO2/kg. P = 2bar -> 19gCO2/kg. P = 3bar -> 9gCO2/kg. Duration = 4h: P = 1bar -> 32gCO2/kg. P = 2bar -> 25gCO2/kg. P = 3bar -> 14gCO2/kg. Duration = 6h: P = 1bar -> 34gCO2/kg. P = 2bar -> 28gCO2/kg. P = 3bar -> 18gCO2/kg. Duration =48h: P = 1bar -> 74gCO2/kg. P = 2bar -> 66gCO2/kg. P = 3bar -> 64gCO2/kg.
					Case 4 - Particle size: Grain Size =2,00-4,75, 4,75-9,50, 9,50-14,00mm. RH = 70%, T = 90°C, P = 1bar, CO_2 = 15%, $v_{rotation}$ = 1rpm.	Intensive	Time: 48h. Mass of sorbent: not present			Grain Size = 2,00-4,75mm -> 74gCO_2/kg. Grain Size = 4,75-9,50mm -> 38gCO_2/kg. Grain Size = 9,50-14,00 -> 31gCO_2/kg.

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Ndiaye S., Condoret J.-S., Bourgeois F., Cassayre L., Camy S. High-pressure carbonation of mortar as a model for recycled concrete aggregates. 2023. doi: 10.1016/j.supflu.2023.105932	Study the role of CO2 pressure, material water content and temperature on the kinetics of the carbonation reaction of mortar powders and cylindrical mortar samples, in order to evidence the potential of RCA to attempt maximizing CO2 storage and improving reaction kinetics.	Autoclave reactor.	RCAs (Recycled Concrete Aggregates)	Runs implementing high pressure CO2 were carried out using a SEPAREX SFE200 set-up (Separex Company, Nancy, France) with an autoclave. The CO_2 was fed into the autoclave using a volumetric metallic membrane pump to reach the desired pressure, which usually took a few minutes. The pressure was kept constant using a CO_2 syringe pump (260D syringe pump, Teledyne ISCO, Nebraska, USA) allowing injection of CO2 to maintain a constant pressure during the experiment, as CO_2 was consumed by the reaction.	CO_2 = 100%, T = 40 - 80°C, P = 20 - 100bar, Grain Size (powder) < 0,198mm.	Very Intensive	Time: 15min - 48h. Mass of sorbent: powder -> 30g. cylinder -> 220g.	Dried solids (reactants and products) were characterized by X-ray diffraction (XRD) using a Bruker D8 Advance X-ray Diffractometer. Scanning electron microscopy (SEM) investigations were performed on the solid samples to investigate morphology, size and texture of the reaction products obtained during various experiments. The particle size distribution was measured by laser diffraction without dispersant using a Malvern MS3000. The specific surface areas and the porosity of the powders were determined by the BET and BJH nitrogen adsorption method. X-Ray Fluorescence spectroscopy (XRF) was performed on ground mortar powder to determine its elemental composition.	Thermogravimetric analysis and total C analysis were used to assess the extent of carbonation of the carbonated mortar. TGA was mainly used to detect the presence of carbonates, while total carbon analysis was used to quantify their amount.	11gCO2/kg, 135gCO2/kg, 67gCO2/kg
Soares E. G., Castro-Gomes J.. The role of biomass bottom ash in Carbonated Reactive Magnesia Cement (CRMC) for CO2 mineralisation. 2022. doi: 10.1016/j.jclepro.2022.135092	Evaluates the incorporation of biomass bottom ashes on Carbonated Reactive Magnesia Cement-based (CRMC) mortars.	Batch reactor.	Biomass Bottom Ashes (BS)	The 20 fresh specimens were placed together into a pressurised carbonation chamber of about 75 litters, where a partial vacuum pressure of 0.2 bar was applied, and, soon after it, the accelerated carbonation curing	Grain Size = 0.125 - 4mm, < 0,125mm, < 10mm, P = 0,7bar, CO2 > 99%, T = 50 ± 2°C, RH > 99%.	Very Intensive	Time: 24h. Mass of sorbent: not present.	The oxide compositions were estimated through Energy-dispersive X-ray spectroscopy (SEM-EDX) using a Hitachi S-3400N apparatus. The particle size distributions of BSb, BSm, and YS were obtained through the sieving method.	The theoretical CO2 adsorption (TCO2) was estimated through Huntzinger's formula. Thermogravimetry and Derivative thermogravimetry (TG-DTG) analysis were carried out on a	Grain Size < 10mm -> 86,9gCO2/kg. Grain Size < 0,125mm -> 32,1gCO2/kg. Grain Size < 10mm -> 93,3gCO2/kg. Grain Size = 0,125 - 4mm -> 43,10gCO2/kg.

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				period of 24 h was applied.				X-ray diffraction (XRD) analyses were performed in a D-Max III/C (Rigaku) apparatus to qualitatively analyse the crystalline phases. A scanning electron microscope (SEM) model S-2700 (Hitachi) was used to obtain the microstructure images.	SDT Q-50 (TA Instrument).	
Wang A., Ren P., Zeng Q., Ling T.-C.. Performance investigation and optimization of the granulation-CO_2 concentration for the production of high-strength BOFS aggregates. 2022. doi: 10.1016/j.jcou.2022.102160	Optimize the CO_2 concentration during granulation of BOFS and investigate the impact on the subsequent post air or CO_2 curing of the produced aggregates.	Batch, Slurry reactor.	Basic Oxygen Furnace Slags (BOFS)	For CC, the fresh BOFS aggregates were placed in a chamber at temperature of 20 ◦C, relative humidity (RH) of 65% and 20% CO_2 concentration for 4 days because CO_2 uptake would reach a plateau at 4 days according to previous studies.	T = 20°C, RH = 65%, CO2 = 0, 20, 40, 60, 80, 100%, L/S = 0,17, 0,18, 0,21, 0,25, 0,27, Grain Size < 4mm.	Mild	Time: 96h. Mass of sorbent: not present.	The macroscopic pore structure of aggregate was characterized by Xray computed tomography (XCT) with a XTH255/320 LC XCT equipment (Nikon, Japan). An HVS-1000Z microhardness tester with an applied load of 0.01 kg and dwell time of 10 s was used to determine the microhardness values of the observed aggregate.	CO_2 mass and CO_2 uptake of aggregates were determined using thermogravimetric analysis (Rigaku 8121 H).	CO2 = 0%, L/S = 0,17 -> 59,20gCO2/kg. CO2 = 20%, L/S = 0,17 -> 100,90gCO2/kg. CO2 = 40%, L/S = 0,18 -> 130gCO2/kg. CO2 = 60%, L/S = 0,21 -> 108,30gCO2/kg. CO2 = 80%, L/S = 0,25 -> 77,30gCO2/kg. CO2 = 100%, L/S = 0,27-> 60,4 gCO2/kg.

ANNEX II: CRITICAL REVIEW – EXPERIMENTAL DATA

In this annex, all the variables' data, used in each study, are shown. These values are necessary to compare the analysis with each other and to know the reason for the different CO_2 absorption results. Thus, it's possible to know the values obtained from different methods of carbonation and finally compare them to those that come from this study.

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
FLY ASHES			x			1	20	3		200,00							2.5
RCA			x			0.1	25 ± 3	1	50 ± 5	6,50			5 to 10		> 99.5	x	
RCA			x			0.1	25 ± 3	3	50 ± 5	12,50			5 to 10		> 99.5	x	
RCA			x			0.1	25 ± 3	5	50 ± 5	24,00			5 to 10		> 99.5	x	
RCA			x			0.1	25 ± 3	7	50 ± 5	19,50			5 to 10		> 99.5	x	
RCA			x			0.1	25 ± 3	24	50 ± 5	19,50			5 to 10		> 99.5	x	
RCA			x			1	25 ± 3	24	50 ± 5	55,00			1.5		> 99.5		
RCA			x			1	25 ± 3	24	50 ± 5	32,00			3		> 99.5		
RCA			x			1	25 ± 3	24	50 ± 5	25,00			7.5		> 99.5		
RCA			x			1	25 ± 3	24	50 ± 5	27,00			15		> 99.5		

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA					x	5	25 ± 3	24	50 ± 5	28,20			5 to 10		> 99.5		
SLAGS				x		1	200	1		0,00			< 0.25	3			
SLAGS				x		1	200	2		40,00			< 0.25	3			
SLAGS				x		1	200	4		44,00			< 0.25	3			
SLAGS				x		1	200	6		53,00			< 0.25	3			
SLAGS				x		1	200	2		40,00			< 0.25	3			
SLAGS				x		1	360	2		20,00			< 0.25	3			
FLY ASHES	0.044			x		1	300 - 500	0.17 - 48	75			250			10 to 50	x	
FLY ASHES	1				x	> 1	30 - 50	0.17 - 48	75			250			100		0.02 to 0.6
CONCRETE MONOLITE	47.25		x			0.7 - 1	25		50	11,60			/		99.5	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
CONCRETE MONOLITE	48.26		x			0.7 - 1	25		50	0,60			/		99.5	x	
CONCRETE MONOLITE	28.84		x			0.7 - 1	25		50	0,30			/		99.5	x	
CONCRETE MONOLITE	76.45		x			0.7 - 1	25		50	28,30			/		99.5	x	
CONCRETE MONOLITE	35.58		x			0.7 - 1	25		50	0,80			/		99.5	x	
CONCRETE MONOLITE	45.91		x			0.7 - 1	25		50	0,40			/		99.5	x	
CONCRETE MONOLITE	44.33		x			0.7 - 1	25		50	29,80			/		99.5	x	
CONCRETE MONOLITE	38.76		x			0.7 - 1	25		50	0,80			/		99.5	x	
CONCRETE MONOLITE	34.92		x			0.7 - 1	25		50	0,60			/		99.5	x	
CONCRETE MONOLITE	47.26		x			0.7 - 1	25		50	30,50			/		99.5	x	
CONCRETE MONOLITE	55.4		x			0.7 - 1	25		50	0,90			/		99.5	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
CONCRETE MONOLITE	47.28		x			0.7 - 1	25		50	0,60			/		99.5	x	
CONCRETE MONOLITE	65.15		x			0.7 - 1	25		50	31,50			/		99.5	x	
CONCRETE MONOLITE	71.14		x			0.7 - 1	25		50	32,00			/		99.5	x	
CONCRETE MONOLITE	50.69		x			0.7 - 1	25		50	28,20			/		99.5	x	
CONCRETE MONOLITE	40.57		x			0.7 - 1	25		50	45,90			/		99.5	x	
CONCRETE MONOLITE	43.61		x			0.7 - 1	25		50	0,90			/		99.5	x	
RCA			x			0.1	25	1	50 ± 5	7,00			5 to 10		100	x	
RCA			x			0.1	25	3	50 ± 5	12,80			5 to 10		100	x	
RCA			x			0.1	25	5	50 ± 5	24,00			5 to 10		100	x	
RCA			x			0.1	25	7	50 ± 5	19,00			5 to 10		100	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA			x			0.1	25	24	50 ± 5	19,00			5 to 10		100	x	
RCA					x	5	25	24	50 ± 5	28,00			5 to 10		100	x	
RCA			x			1	25	24	50 ± 5	55,00			< 2.36	1	100	x	
RCA			x			1	25	24	50 ± 5	30,00			2.36 to 5	1	100	x	
RCA			x			1	25	24	50 ± 5	26,00			5 to 10	1	100	x	
RCA			x			1	25	24	50 ± 5	27,00			10 to 20	1	100	x	
RCA			x			1	25	3	50 ± 5	9,50			5 to 10	1	10	x	
RCA			x			1	25	5	50 ± 5	9,90			5 to 10	1	10	x	
RCA			x			1	25	7	50 ± 5	9,70			5 to 10	1	10	x	
RCA			x			1	25	24	50 ± 5	10,20			5 to 10	1	10	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA			x			1	25	3	50 ± 5	5,80			5 to 10	1	100	x	
RCA			x			1	25	5	50 ± 5	12,20			5 to 10	1	100	x	
RCA			x			1	25	7	50 ± 5	12,40			5 to 10	1	100	x	
RCA			x			1	25	24	50 ± 5	12,20			5 to 10	1	100	x	
RCA			x			1	25	3	50 ± 5	4,00			5 to 10	1	100	x	
RCA			x			1	25	5	50 ± 5	4,20			5 to 10	1	100	x	
RCA			x			1	25	7	50 ± 5	3,50			5 to 10	1	100	x	
RCA			x			1	25	24	50 ± 5	3,40			5 to 10	1	100	x	
RCA			x			1	25	3	50 ± 5	4,90			5 to 10	5	100	x	
RCA			x			1	25	5	50 ± 5	5,40			5 to 10	5	100	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA			x			1	25	7	50 ± 5	6,60			5 to 10	5	100	x	
RCA			x			1	25	24	50 ± 5	6,50			5 to 10	5	100	x	
RCA			x			1	25	3	50 ± 5	8,50			5 to 10	10	100	x	
RCA			x			1	25	5	50 ± 5	6,00			5 to 10	10	100	x	
RCA			x			1	25	7	50 ± 5	6,50			5 to 10	10	100	x	
RCA			x			1	25	24	50 ± 5	6,40			5 to 10	10	100	x	
CONCRETE MONOLITE					x	1.5	25	2	60	121,00			/		99.5	x	
CONCRETE MONOLITE					x	1.5	25	2	60	129,00			/		99.5	x	
CONCRETE MONOLITE					x	1.5	25	2	60	128,00			/		99.5	x	
RCA			x			1	25	24		8,60			1 to 4		100		0.072

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA			x			1	25	24		17,30			1 to 4		100		0.052
RCA			x			1	25	24		49,90			1 to 4		100		0.052
RCA		x				1	25	24		10,40			1 to 4		15		0.035
RCA		x				1	25	24		21,80			1 to 4		15		0.04
RCA		x				1	25	24		21,80			1 to 4		15		0.04
RCA		x				1	25	24		8,50			12 to 20		15		0.028
RCA		x				1	25	24		10,40			1 to 4		15		0.035
RCA		x				1	25	24		6.5			12 to 20		15		0.022
RCA		x				1	25	24		14,90			10 to 20		15		0.038
RCA			x			1	25	24		20,40			10 to 20		100		0.038

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO$_2$ UPTAKE [g/kg]			GRANULOMETRY [mm]	CO$_2$ PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	10		x			1	20	120	65	138,50			< 30		20		0,7
RCA	10		x			1	20	72	65	169,50			< 30		20		0,1
RCA	10		x			1	20	168	65	192,70			< 30		20		0,4
RCA	10		x			1	20	120	65	189,70			< 30		20		0,4
RCA	10		x			1	20	24	65	186,20			< 30		20		0,25
RCA	10		x			1	20	72	65	138,50			< 30		20		0,7
RCA	10		x			1	20	72	65	186,60			< 30		20		0,4
RCA	10		x			1	20	168	65	174,70			< 30		20		0,1
RCA	10		x			1	20	1	65	126,90			< 30		20		0,1
RCA	10		x			1	20	120	65	188,60			< 30		20		0,25

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	10		x			1	20	1	65	126,90			< 30		20		0,1
RCA	10		x			1	20	120	65	171,30			< 30		20		0,1
RCA	10		x			1	20	72	65	186,60			< 30		20		0,4
RCA	10		x			1	20	168	65	192,70			< 30		20		0,4
RCA	10		x			1	20	168	65	180,50			< 30		20		0,7
RCA	10		x			1	20	1	65	101,60			< 30		20		0,7
RCA	10		x			1	20	24	65	122,90			< 30		20		0,55
RCA	10		x			1	20	1	65	101,60			< 30		20		0,7
RCA	10		x			1	20	1	65	126,50			< 30		20		0,4
RCA	10		x			1	20	168	65	180,50			< 30		20		0,7

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	10		x			1	20	168	65	174,70			< 30		20		0,1
RCA	10		x			1	20	72	65	143,00			< 30		20		0,55
RCA	10		x			1	20	72	65	186,60			< 30		20		0,4
CEMENT MORTAR	45		x			1	25	1		19,10					99	x	
CEMENT MORTAR	45		x			1	25	7		25,50					99	x	
CEMENT MORTAR	45		x			1	25	28		26,40					99	x	
CEMENT MORTAR	45		x			1	25	1		15,60					99	x	
CEMENT MORTAR	45		x			1	25	7		24,00					99	x	
CEMENT MORTAR	45		x			1	25	28		27,40					99	x	
LIME-BASED MORTAR	45		x			1	25	1		5,00					99	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
LIME-BASED MORTAR	45		x			1	25	7		11,00					99	x	
LIME-BASED MORTAR	45		x			1	25	28		16,10					99	x	
LIME-BASED MORTAR	45		x			1	25	1		2,30					99	x	
LIME-BASED MORTAR	45		x			1	25	7		13,70					99	x	
LIME-BASED MORTAR	45		x			1	25	28		18,10					99	x	
CONCRETE MONOLITE					x	1.5	40 ± 2	2	>= 90	160,00			/		99.9		0.2
CONCRETE MONOLITE					x	1.5	40 ± 2	2	>= 90	158,00			/		99.9		0.25
CONCRETE MONOLITE					x	1.5	40 ± 2	2	>= 90	148,00			/		99.9		0.3
CONCRETE MONOLITE					x	1.5	40 ± 2	2	>= 90	135,00			/		99.9		0.35
CONCRETE MONOLITE					x	1.5	40 ± 2	2	>= 90	130,00			/		99.9		0.4

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
CONCRETE MONOLITE					x	5	40 ± 2	2	>= 90	57,00			/		99.9		0.25
CONCRETE MONOLITE					x	10	40 ± 2	2	>= 90	138,00			/		99.9		0.25
CONCRETE MONOLITE					x	15	40 ± 2	2	>= 90	165,00			/		99.9		0.25
CONCRETE MONOLITE					x	20	40 ± 2	2	>= 90	160,00			/		99.9		0.25
CONCRETE MONOLITE					x	25	40 ± 2	2	>= 90	142,00			/		99.9		0.25
FLY ASHES	10			x		10	25	5		27,50			0,0000152				
FLY ASHES	10			x		80	80	5		39,40			0,0000152				30
NATURAL AGGREGATE	2574	x				1	21 ± 2	24	65 ± 10	1,51			<0,5		0.4	x	
NATURAL AGGREGATE	2574	x				1	21 ± 2	72	65 ± 10	5,17			<0,5		0.4	x	
NATURAL AGGREGATE	2574	x				1	21 ± 2	168	65 ± 10	5,89			<0,5		0.4	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
NATURAL AGGREGATE	2574	x				1	21 ± 2	24	65 ± 10	11,83			<0,5		0.4	x	
NATURAL AGGREGATE	2574	x				1	21 ± 2	72	65 ± 10	13,76			<0,5		0.4	x	
NATURAL AGGREGATE	2574	x				1	21 ± 2	168	65 ± 10	13,82			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	24	65 ± 10	2,08			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	72	65 ± 10	5,45			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	168	65 ± 10	8,32			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	24	65 ± 10	14,01			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	72	65 ± 10	15,85			<0,5		0.4	x	
RCA	3507	x				1	21 ± 2	168	65 ± 10	16,55			<0,5		0.4	x	
CONCRETE MONOLITE					x	1.5	24	2	30	87,00			/		99.5	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
CONCRETE MONOLITE					x	1.5	24	2	30	89,00			/		99.5	x	
CONCRETE MONOLITE					x	1.5	24	2	30	79,00			/		99.5	x	
CONCRETE MONOLITE					x	5	25 ± 3	12	65 ± 5		99,5	146	/		99.8		0.3 - 0.4
CONCRETE MONOLITE			x			1	25 ± 3	12	65 ± 5	100,00			/		99.8		0.3 - 0.4
CONCRETE MONOLITE					x	2	25 ± 3	12	65 ± 5	126,00			/		99.8		0.3 - 0.4
CONCRETE MONOLITE					x	3	25 ± 3	12	65 ± 5	128,00			/		99.8		0.3 - 0.4
CONCRETE MONOLITE					x	4	25 ± 3	12	65 ± 5	136,00			/		99.8		0.3 - 0.4
CONCRETE MONOLITE					x	5	25 ± 3	12	65 ± 5	145,00			/		99.8		0.3 - 0.4
RCA	500		x			0.75	22	96		7,50			10 to 20		100	x	
RCA	500		x			0.75	22	72		15,90			10 to 20		100	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	500		x			0.75	22	72		28,90			10 to 20		100	x	
RCA	500		x			0.75	22	72		37,90			10 to 20		100	x	
RCA	500		x			0.75	22	72		39,20			10 to 20		100	x	
RCA	500		x			0.75	22	96		45,80			10 to 20		100	x	
RCA	500		x			0.75	22	168		79,50			10 to 20		100	x	
RCA	500		x			0.75	22	72		45,50			8 to 10		100	x	
NATURAL AGGREGATE		x				1	21 ± 2	24	65 ± 5	1,57			< 2		0,04	x	
NATURAL AGGREGATE		x				1	21 ± 2	72	65 ± 5	5,21			< 2		0,04	x	
NATURAL AGGREGATE		x				1	21 ± 2	168	65 ± 5	5,87			< 2		0,04	x	
NATURAL AGGREGATE		x				1	21 ± 2	24	65 ± 5	3,30			< 2		5	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
NATURAL AGGREGATE		x				1	21 ± 2	72	65 ± 5	19,93			< 2		5	x	
NATURAL AGGREGATE		x				1	21 ± 2	168	65 ± 5	23,96			< 2		5	x	
RCA MONOLITE		x				1	21 ± 2	24	65 ± 5	2,14			< 2		0,04	x	
RCA MONOLITE		x				1	21 ± 2	72	65 ± 5	5,50			< 2		0,04	x	
RCA MONOLITE		x				1	21 ± 2	168	65 ± 5	8,35			< 2		0,04	x	
RCA MONOLITE		x				1	21 ± 2	24	65 ± 5	8,62			< 2		5	x	
RCA MONOLITE		x				1	21 ± 2	72	65 ± 5	20,59			< 2		5	x	
RCA MONOLITE		x				1	21 ± 2	168	65 ± 5	27,10			< 2		5	x	
RCA	500		x			1	22	7		17,20			< 6			x	
RCA	500		x			1	22	7		30,40			< 6			x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	500			x		1.4	22	7		47,40			< 6			x	
RCA	1000			x		1.4	22	7		41,20			< 6			x	
RCA	500		x			1	22	7		30,80			< 6			x	
RCA	500		x			1	22	7		12,00			< 6			x	
RCA	1500		x			1	22	7		15,30			< 6			x	
RCA	500		x			1	22	7		4,20			< 6			x	
RCA	500		x			1	22	7		9,20			< 6			x	
RCA	500		x			1	22	7		15,50			< 6			x	
FLY ASHES	200		x			1	25	0.5		45,62			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		39,71			4 to 10	1,667	20		0.2 to 0.5

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
FLY ASHES	200		x			1	25	0.5		108,06			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		54,45			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		23,42			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		85,52			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		88,31			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		67,67			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		71,67			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		80,96			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		93,82			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		65,71			4 to 10	1,667	20		0.2 to 0.5

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
FLY ASHES	200		x			1	25	0.5		64,72			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		80,28			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES	200		x			1	25	0.5		49,08			4 to 10	1,667	20		0.2 to 0.5
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	4,87			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	10,22			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	13,05			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	6,96			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	11,66			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	6	> 95	13,42			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	5,72			/		100	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	12,41			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	13,33			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	5,99			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	13,79			/		100	x	
FLY ASHES MONOLITE					x	1.5	20 ± 2	24	> 95	14,66			/		100	x	
FLY ASHES	200		x			1	25	1	> 25	101,70			0,002 to 0,130	10	99.9	x	0.002 to 0.130
FLY ASHES MONOLITE			x			1	25			84,50			/	200			10
SLAGS	100			x		> 1	40	48		29,47			< 0.1				3
BOTTOM ASHES			x			1	25	8 to 14			24	48	5 to 10	0.4	100	x	
BOTTOM ASHES			x			1	25	8 to 14			34	46	5 to 10	0.4	100	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
BOTTOM ASHES			x			1	25	8 to 14			27	47	5 to 10	0.4	100	x	
BOTTOM ASHES			x			1	25	8 to 14			34	48	5 to 10	0.4	100	x	
BOTTOM ASHES			x			1	25	8 to 14			33	37	5 to 10	0.4	100	x	
BOTTOM ASHES			x			1	25	8 to 14			25	31	5 to 10	0.4	100	x	
BOTTOM ASHES			x			1	25	8 to 14			19	23	5 to 10	0.4	100	x	
CONCRETE			x			0.1	25	144	50	110,00			< 0.15		> 99.5	x	
RCA			x			0.1	25	144	50	52,00			< 5		> 99.5	x	
RCA			x			0.1	25	144	50	20,00			< 5		> 99.5	x	
BOTTOM ASHES			x			1	25	2		102,00			0.125 to 0.35	0.5	99		10
FLY ASHES	300				x	30	60	10		10,71			0.32		100		0.3

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
FLY ASHES	300				x	30	60	10		20,13			0.116		100		0.3
FLY ASHES	300				x	30	60	10		27,05			0.054		100		0.3
SLAGS	500		x			1	23 ± 2	1.5			120	149	< 10		100		0.12
FLY ASHES	100		x			1	25	1.2		87,00				0.01	100		0.2
FLY ASHES	150	x				1	25	1.2		41,00				0.01	12		0.2
FLY ASHES	300				x	30	40	10		7,66			0.9		100		0.0 to 0.7
BOTTOM ASHES	100		x			1	20	0.416667	75	3,00			> 4.75		20		0.2
BOTTOM ASHES	100		x			1	20	0.416667	75	4,00			2.36 to 4.75		20		0.2
BOTTOM ASHES	100		x			1	20	0.416667	75	5,00			1.18 to 2.36		20		0.2
BOTTOM ASHES	100		x			1	20	0.416667	75	18,00			0.6 to 1.18		20		0.2

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
BOTTOM ASHES	100		x			1	20	0.416667	75	50,00			0.3 to 0.6		20		0.2
BOTTOM ASHES	100		x			1	20	0.416667	75	84,00			0.15 to 0.3		20		0.2
BOTTOM ASHES	100		x			1	20	0.416667	75	165,00			< 0.15		20		0.2
SLAGS					x	5 to 9	160	12		127,00			< 0.044		100		10
SLAGS					x	5 to 9	160	12		107,00			< 0.044		100		10
SLAGS					x	5 to 9	160	12		238,00			< 0.044		100		10
FLY ASHES	110 ± 10		x			1	25	168		43,56			< 3		100		0,3
FLY ASHES	110 ± 10		x			1	25	168		76,56			< 3		100		0,2
BOTTOM ASHES	110 ± 10		x			1	25	168		92,84			< 3		100		0,3
SLAGS	110 ± 10		x			1	25	168		111,76			< 3		100		0,2

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA MONOLITE			x			0.1	25	24	50	33,00			/		> 99.5	x	
RCA MONOLITE					x	5	25	24	50	36,00			/		> 99.5	x	
RCA				x		1	90	48	50	37,20			2,00 - 4,75		15	x	
RCA				x		1	90	48	70	74,00			2,00 - 4,75		15	x	
RCA				x		1	90	48	90	63,60			2,00 - 4,75		15	x	
RCA				x		1	50	2	70	15,00			2,00 - 4,75		15	x	
RCA				x		1	50	4	70	22,00			2,00 - 4,75		15	x	
RCA				x		1	50	6	70	25,00			2,00 - 4,75		15	x	
RCA				x		1	50	12	70	35,00			2,00 - 4,75		15	x	
RCA				x		1	50	24	70	50,00			2,00 - 4,75		15	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA				x		1	50	48	70	74,00			2,00 - 4,75		15	x	
RCA				x		1	50	72	70	76,00			2,00 - 4,75		15	x	
RCA				x		1	50	96	70	77,00			2,00 - 4,75		15	x	
RCA				x		1	50	120	70	78,00			2,00 - 4,75		15	x	
RCA				x		1	90	2	70	22,00			2,00 - 4,75		15	x	
RCA				x		1	90	4	70	30,00			2,00 - 4,75		15	x	
RCA				x		1	90	6	70	35,00			2,00 - 4,75		15	x	
RCA				x		1	90	12	70	42,00			2,00 - 4,75		15	x	
RCA				x		1	90	24	70	55,00			2,00 - 4,75		15	x	
RCA				x		1	90	48	70	75,00			2,00 - 4,75		15	x	

MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA				x		1	90	72	70	77,00			2,00 - 4,75		15	x	
RCA				x		1	90	96	70	78,00			2,00 - 4,75		15	x	
RCA				x		1	90	120	70	79,00			2,00 - 4,75		15	x	
RCA				x		1	90	2	70	23,00			2,00 - 4,75		15	x	
RCA				x		1	90	4	70	32,00			2,00 - 4,75		15	x	
RCA				x		1	90	6	70	34,00			2,00 - 4,75		15	x	
RCA				x		1	90	48	70	74,00			2,00 - 4,75		15	x	
RCA				x		2	90	2	70	19,00			2,00 - 4,75		15	x	
RCA				x		2	90	4	70	25,00			2,00 - 4,75		15	x	
RCA				x		2	90	6	70	28,00			2,00 - 4,75		15	x	

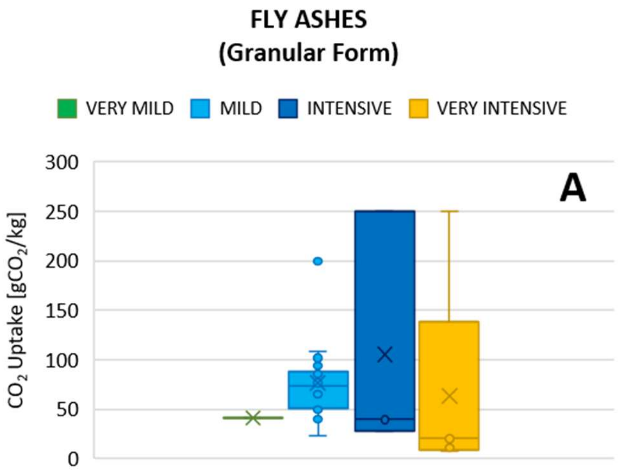
MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA				x		2	90	48	70	66,00			2,00 - 4,75		15	x	
RCA				x		3	90	2	70	9,00			2,00 - 4,75		15	x	
RCA				x		3	90	4	70	14,00			2,00 - 4,75		15	x	
RCA				x		3	90	6	70	18,00			2,00 - 4,75		15	x	
RCA				x		3	90	48	70	64,00			2,00 - 4,75		15	x	
RCA				x		1	90	48	70	74,00			2,00 - 4,75		15	x	
RCA				x		1	90	48	70	38,00			4,75 - 9,50		15	x	
RCA				x		1	90	48	70	31,00			9,50 - 14,00		15	x	
RCA	30				x	100	80	1		11,00			< 0,198		100	x	
RCA	30				x	100	80	1		135,00			< 0,198		100	x	

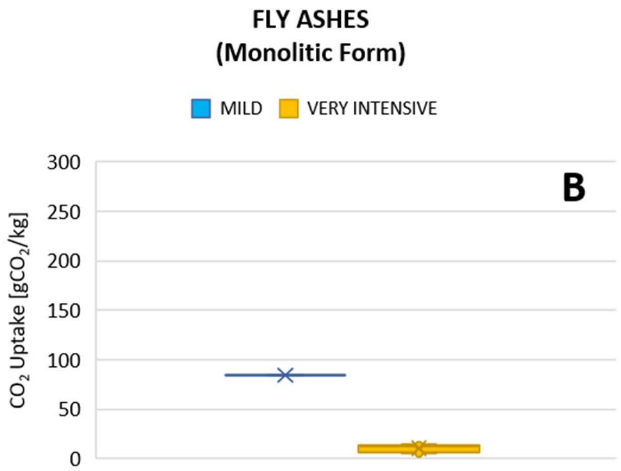
MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
RCA	30				x	100	80	1		67,00			< 0,198		100	x	
BOTTOM ASHES					x	0,7	50 ± 2	24	> 99	86,90			< 10		> 99	x	
BOTTOM ASHES					x	0,7	50 ± 2	24	> 99	32,10			< 0,125		> 99	x	
BOTTOM ASHES					x	0,7	50 ± 2	24	> 99	93,30			< 10		> 99	x	
BOTTOM ASHES					x	0,7	50 ± 2	24	> 99	43,10			0,125 - 4		> 99	x	
SLAGS		x				1	20	96	65	59,20			< 4		0		0,17
SLAGS			x			1	20	96	65	100,90			< 4		20		0,17
SLAGS			x			1	20	96	65	130,00			< 4		40		0,18
SLAGS			x			1	20	96	65	108,30			< 4		60		0,21
SLAGS			x			1	20	96	65	77,30			< 4		80		0,25

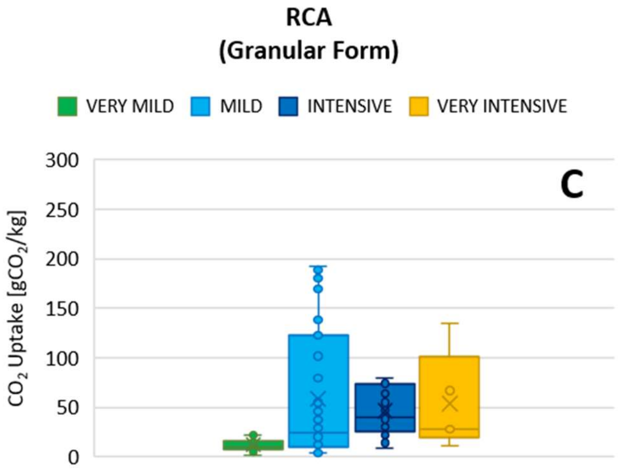
MATERIAL	MASS [g]	OPERATIVE CONDITIONS				PRESSURE [bar]	TEMPERATURE [°C]	TIME [h]	HUMIDITY [%]	CO_2 UPTAKE [g/kg]			GRANULOMETRY [mm]	CO_2 PARAMETERS		REACTOR TYPE	
		VERY MILD	MILD	INTENSIVE	VERY INTENSIVE					PUNCTUAL VALUE	MIN. VALUE	MAX VALUE		FLOW [L/min]	CONC. [%]	DRY	SLURRY_L/S RATIO [l/kg]
SLAGS			x			1	20	96	65	60,40			< 4		100		0,27

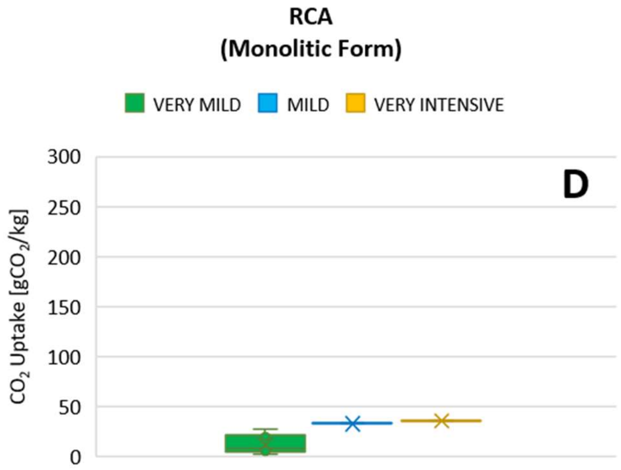
ANNEX III: CRITICAL REVIEW – BOX PLOTS

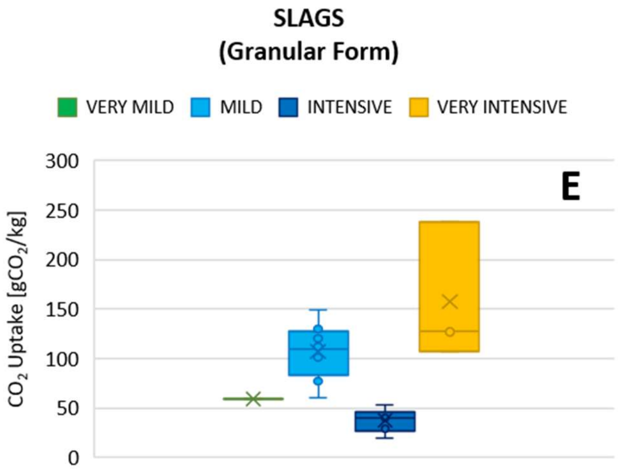
In this annex, the quantities of CO_2 absorbed by each material in different operative conditions are presented through a series of box plots. The operative conditions' categories are: very mild ($T < 35$ °C, $P = 1$ bar, and $C_{CO2} = 0\ \% – 20\ \%$), mild ($T < 35$ °C, $P = 1$ bar, and $C_{CO2} = 20\ \% – 100\ \%$), intensive ($T > 35$ °C or $P > 1$ bar and $C_{CO2} < 20\ \%$), and very intensive ($T > 35$ °C or $P > 1$ bar and $C_{CO2} = 20\ \% - 100\ \%$).

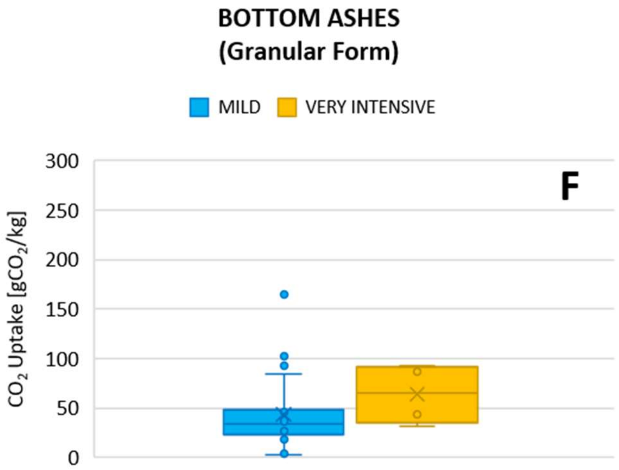

FLY ASHES
(Granular Form)

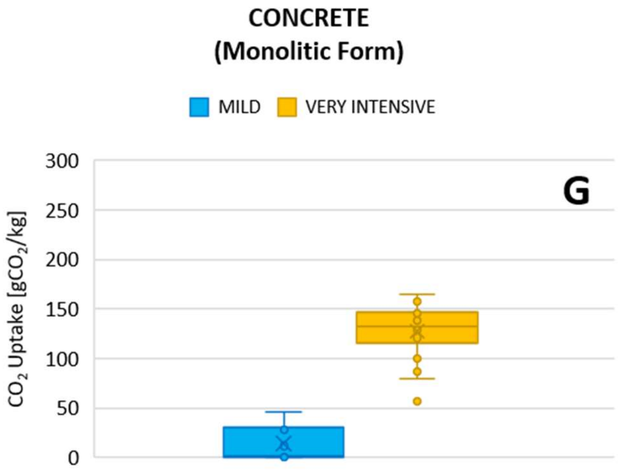

FLY ASHES
(Monolitic Form)

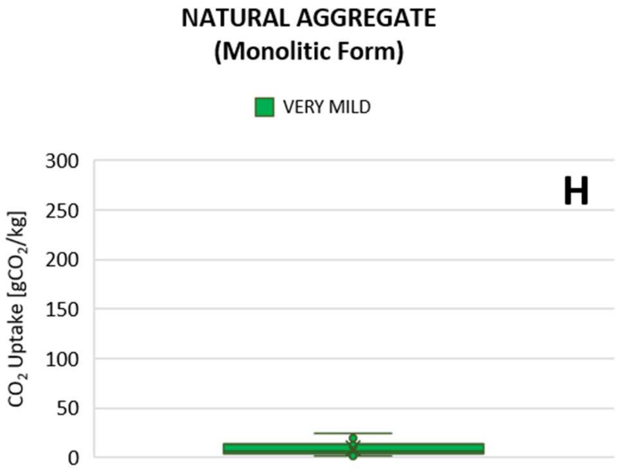

RCA
(Granular Form)


RCA
(Monolitic Form)


SLAGS
(Granular Form)
VERY MILD
MILD
INTENSIVE
VERY INTENSIVE
E
CO₂ Uptake [gCO₂/kg]
300
250
200
150
100
50
0

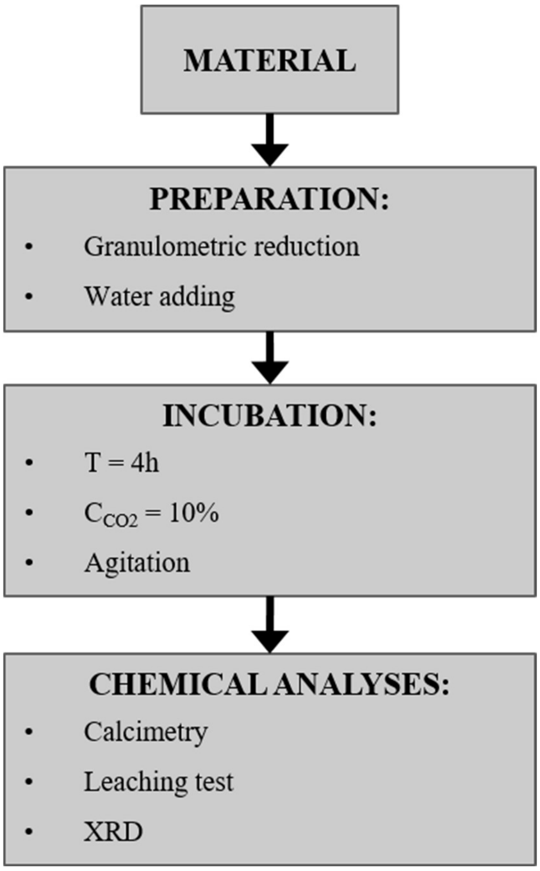

BOTTOM ASHES
(Granular Form)
MILD
VERY INTENSIVE
F
CO₂ Uptake [gCO₂/kg]
300
250
200
150
100
50
0


CONCRETE
(Monolitic Form)
MILD
VERY INTENSIVE
G
CO₂ Uptake [gCO₂/kg]
300
250
200
150
100
50
0


NATURAL AGGREGATE
(Monolitic Form)
VERY MILD
H
CO₂ Uptake [gCO₂/kg]
300
250
200
150
100
50
0

ANNEX IV: OPERATIVE SCHEME

Here the operative scheme used to implement the accelerated carbonation process in the lab is reported. This is the procedure adopted as a reference for each material, thus it’s possible to see the methodology adopted with the pretreatment (granulometric reduction and water adding) and the chemical analysis (calcimetry tests, leaching tests, and X-ray diffraction analysis) that have been performed on the materials used as sorbents in the carbonation reaction.



ANNEX V: MATERIALS CHARACTERISTICS

In the following tables, the humidity values of the materials are reported. The data are referred to both the material non-carbonated and carbonated, and in this last case also to the different liquid to solid rates.

PORTLAND						
CONDITION	**N.**	**P_i**	**P_f**	**UMIDITY**	**AVERAGE UMIDITY**	**SD**
PRE INCUBATION PORT - L/S = 0.0	1	20,423	18,873	0,076	0,074	0,006
	2	20,392	18,765	0,080		
	3	20,261	18,900	0,067		
POST INCUBATION PORT - L/S = 0.0	1	22,569	21,252	0,058	0,061	0,002
	2	22,239	20,853	0,062		
	3	22,426	21,059	0,061		
POST INCUBATION PORT - L/S = 0.2	1	22,561	19,053	0,155	0,160	0,004
	2	22,931	19,230	0,161		
	3	22,750	19,068	0,162		
POST INCUBATION PORT - L/S = 0.3	1	22,407	17,812	0,205	0,205	0,008
	2	22,695	18,239	0,196		
	3	22,487	17,709	0,212		

BFLY						
CONDITION	**N.**	**P_i**	**P_f**	**UMIDITY**	**AVERAGE UMIDITY**	**SD**
PRE INCUBATION BFLY - L/S = 0.4	1	20,052	15,008	0,252	0,280	0,027
	2	20,298	14,110	0,305		
	3	20,498	14,662	0,285		
POST INCUBATION BFLY - L/S = 0.4	1	278,510	223,840	0,196	0,203	0,006
	2	279,760	221,790	0,207		
	3	279,910	222,470	0,205		

SAND

CONDITION	N.	P_i	P_f	UMIDITY	AVERAGE UMIDITY	SD
PRE INCUBATION SAND - L/S = 0.0	1	31,221	29,726	0,048	0,048	0,002
	2	34,355	32,664	0,049		
	3	36,524	34,843	0,046		
POST INCUBATION SAND - L/S = 0.0	1	20,493	19,713	0,038	0,044	0,002
	2	20,327	19,466	0,042		
	3	20,251	19,338	0,045		
POST INCUBATION SAND - L/S = 0.3	1	20,799	16,374	0,213	0,208	0,023
	2	20,251	16,550	0,183		
	3	20,866	16,089	0,229		
POST INCUBATION SAND - L/S = 0.0 - MIX	1	15,743	12,536	0,204	0,202	0,003
	2	14,655	11,680	0,203		
	3	24,039	19,273	0,198		

CLAY

CONDITION	N.	P_i	P_f	UMIDITY	AVERAGE UMIDITY	SD
PRE INCUBATION CLAY - L/S = 0.0	1	207,730	159,640	0,232	0,228	0,005
	2	201,130	155,020	0,229		
	3	213,680	166,110	0,223		
POST INCUBATION CLAY - L/S = 0.0	1	30,772	28,561	0,072	0,183	0,028
	2	30,214	24,096	0,203		
	3	30,548	25,582	0,163		
POST INCUBATION CLAY - L/S = 0.3	1	37,161	24,777	0,333	0,335	0,006
	2	32,505	21,421	0,341		
	3	33,375	22,372	0,330		
POST INCUBATION CLAY - L/S = 0.3 - MIX	1	31,200	20,605	0,340	0,342	0,002
	2	31,468	20,686	0,343		
	3	32,004	20,982	0,344		

ANNEX VI: CALCIMETRY RESULTS

In this annex the calcimetry tests results and the methodology used to measure the quantity of CO_2 absorbed by each material in different operative conditions are reported. Thus it's possible to see the values of carbon uptake obtained by using different materials in different conditions of Liquid-to-Solid ratio.

PORTLAND																		
SAMPLE	**SAMPLE MASS [g]**	**H_2O INITIAL [ml]**	**H_20 ADDED [ml]**	**H INITIAL**	**H FINAL**	**TS INITIAL**	**TS FINAL**	**L/S [l/kg]**	**L/S REAL [L/kg]**	**MASS CALCIMETRY [g]**	**VOL. CO_2 CALCIMETRY [L]**	**nCO_2 [mol]**	**mCO_2 [g]**	**mDW [g]**	**gCO2 / kg DW**	**AVERAGE g CO2 / kg DW**	**S.D. g CO2 / kg DW**	**Δ g CO2 / kg DW**
0h PORT - L/S = 0.0				0,074		0,926				0,510	0,045	0,002	0,081	0,472	171,422	186,631	12,000	
				0,074		0,926				0,490	0,045	0,002	0,081	0,454	178,419			
				0,074		0,926				0,510	0,050	0,002	0,090	0,472	190,469			
				0,074		0,926				0,510	0,050	0,002	0,090	0,472	190,469			
				0,074		0,926				0,480	0,050	0,002	0,090	0,444	202,374			
4h PORT - L/S = 0.0	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,500	0,044	0,002	0,079	0,470	168,464	173,046	40,549	-13,585
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,520	0,050	0,002	0,090	0,489	184,074			
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,560	0,055	0,002	0,099	0,526	188,018			
4h PORT - L/S = 0.0	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,520	0,040	0,002	0,072	0,489	147,259			
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,500	0,020	0,001	0,036	0,470	76,575			
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,490	0,050	0,002	0,090	0,460	195,344			
4h PORT - L/S = 0.0	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,510	0,050	0,002	0,090	0,479	187,683			
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,480	0,050	0,002	0,090	0,451	199,413			
	215,054	15,054	0,000	0,074	0,061	0,926	0,939	0.0	0,075	0,500	0,055	0,002	0,099	0,470	210,580			
4h PORT - L/S = 0.2	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,690	0,055	0,002	0,099	0,580	170,575	173,444	14,904	-13,186
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,710	0,055	0,002	0,099	0,597	165,770			
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,730	0,055	0,002	0,099	0,614	161,229			
4h PORT - L/S = 0.2	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,740	0,064	0,003	0,115	0,622	185,076			
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,700	0,047	0,002	0,085	0,588	143,682			
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,710	0,058	0,002	0,104	0,597	174,812			
4h PORT - L/S = 0.2	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,690	0,061	0,002	0,110	0,580	189,184			
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,720	0,063	0,003	0,113	0,605	187,245			
	215,054	15,054	40,000	0,216	0,160	0,784	0,840	0.2	0,275	0,700	0,060	0,002	0,108	0,588	183,424			
4h PORT - L/S = 0.3	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,071	0,003	0,128	0,644	198,197	197,496	6,318	10,866
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,073	0,003	0,131	0,644	203,780			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,072	0,003	0,129	0,644	200,989			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,070	0,003	0,126	0,644	195,406			

PORTLAND																		
SAMPLE	**SAMPLE MASS [g]**	**H_2O INITIAL [ml]**	**H_20 ADDED [ml]**	**H INITIAL**	**H FINAL**	**TS INITIAL**	**TS FINAL**	**L/S [l/kg]**	**L/S REAL [L/kg]**	**MASS CALCIMETRY [g]**	**VOL. CO_2 CALCIMETRY [L]**	**nCO_2 [mol]**	**mCO_2 [g]**	**mDW [g]**	**gCO2 / kg DW**	**AVERAGE g CO2 / kg DW**	**S.D. g CO2 / kg DW**	**Δ g CO2 / kg DW**
4h PORT - L/S = 0.3	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,820	0,073	0,003	0,131	0,652	201,295			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,074	0,003	0,133	0,644	206,572			
4h PORT - L/S = 0.3	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,820	0,068	0,003	0,122	0,652	187,508			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,820	0,070	0,003	0,126	0,652	193,023			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,830	0,070	0,003	0,126	0,660	190,697			
4h PORT - L/S = 0.3 - MIX	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,800	0,072	0,003	0,129	0,636	203,501	198,568	6,012	11,938
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,830	0,074	0,003	0,133	0,660	201,594			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,070	0,003	0,126	0,644	195,406			
4h PORT - L/S = 0.3 - MIX	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,069	0,003	0,124	0,644	192,614			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,810	0,073	0,003	0,131	0,644	203,780			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,820	0,072	0,003	0,129	0,652	198,538			
4h PORT - L/S = 0.3 - MIX	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,800	0,073	0,003	0,131	0,636	206,327			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,820	0,068	0,003	0,122	0,652	187,508			
	215,054	15,054	60,000	0,273	0,205	0,727	0,795	0.3	0,375	0,800	0,070	0,003	0,126	0,636	197,848			

BFLY

SAMPLE	SAMPLE MASS [g]	H_2O INITIAL [ml]	H_20 ADDED [ml]	H INITIAL	H FINAL	TS INITIAL	TS FINAL	L/S [l/kg]	L/S REAL [L/kg]	MASS CALCIMETRY [g]	VOL. CO_2 CALCIMETRY [L]	nCO_2 [mol]	mCO_2 [g]	mDW [g]	gCO2 / kg DW	AVERAGE g CO2 / kg DW	S.D. g CO2 / kg DW	Δ g CO2 / kg DW
0h BFLY - L/S = 0.4				0,280		0,720				1,070	0,075	0,003	0,135	0,770	175,176	53,862	60,926	
				0,280		0,720				1,010	0,015	0,001	0,027	0,727	37,116			
				0,280		0,720				0,990	0,019	0,001	0,034	0,712	47,964			
				0,280		0,720				1,090	0,005	0,000	0,009	0,784	11,464			
				0,280		0,720				1,020	0,007	0,000	0,013	0,734	17,151			
				0,280		0,720				1,020	0,014	0,001	0,025	0,734	34,302			
4h BFLY- L/S = 0.4	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,510	0,030	0,001	0,054	0,407	132,722	139,015	4,827	85,153
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,520	0,031	0,001	0,056	0,414	134,508			
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,510	0,033	0,001	0,059	0,407	145,994			
4h BFLY- L/S = 0.4	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,520	0,031	0,001	0,056	0,414	134,508			
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,500	0,032	0,001	0,058	0,399	144,401			
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,510	0,032	0,001	0,058	0,407	141,570			
4h BFLY- L/S = 0.4	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,500	0,030	0,001	0,054	0,399	135,376			
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,530	0,033	0,001	0,059	0,422	140,485			
	277,778	77,778	0,000	0,280	0,203	0,720	0,797	0.0	0,390	0,510	0,032	0,001	0,058	0,407	141,570			

SAND

SAMPLE	SAMPLE MASS [g]	H_2O INITIAL [ml]	H_20 ADDED [ml]	H INITIAL	H FINAL	TS INITIAL	TS FINAL	L/S [l/kg]	L/S REAL [L/kg]	MASS CALCIMETRY [g]	VOL. CO_2 CALCIMETRY [L]	nCO_2 [mol]	mCO_2 [g]	mDW [g]	gCO2 / kg DW	AVERAGE g CO2 / kg DW	S.D. g CO2 / kg DW	Δ g CO2 / kg DW
0h SAND - L/S = 0.0				0,048		0,952				5,040	0,041	0,002	0,074	4,800	15,363	16,947	4,076	
				0,048		0,952				5,050	0,034	0,001	0,061	4,809	12,715			
				0,048		0,952				5,140	0,030	0,001	0,054	4,895	11,023			
				0,048		0,952				5,180	0,056	0,002	0,101	4,933	20,417			
				0,048		0,952				5,080	0,046	0,002	0,083	4,838	17,101			
				0,048		0,952				5,300	0,046	0,002	0,083	5,047	16,391			
				0,048		0,952				4,980	0,050	0,002	0,090	4,742	18,961			

SAND																		
SAMPLE	**SAMPLE MASS [g]**	**H_2O INITIAL [ml]**	**H_20 ADDED [ml]**	**H INITIAL**	**H FINAL**	**TS INITIAL**	**TS FINAL**	**L/S [l/kg]**	**L/S REAL [L/kg]**	**MASS CALCIMETRY [g]**	**VOL. CO_2 CALCIMETRY [L]**	**nCO_2 [mol]**	**mCO_2 [g]**	**mDW [g]**	**gCO2 / kg DW**	**AVERAGE g CO2 / kg DW**	**S.D. g CO2 / kg DW**	**Δ g CO2 / kg DW**
				0,048		0,952				5,200	0,065	0,003	0,117	4,952	23,607			
4h SAND - L/S = 0.0	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	4,830	0,040	0,002	0,072	4,589	15,678	12,263	2,922	-4,684
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,320	0,040	0,002	0,072	5,054	14,234			
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,120	0,022	0,001	0,040	4,864	8,134			
4h SAND - L/S = 0.0	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	4,900	0,037	0,002	0,067	4,655	14,295			
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,050	0,028	0,001	0,050	4,798	10,496			
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,220	0,028	0,001	0,050	4,959	10,155			
4h SAND - L/S = 0.0	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,170	0,044	0,002	0,079	4,912	16,112			
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	4,720	0,030	0,001	0,054	4,484	12,033			
	210,000	10,000	0,000	0,050	0,044	0,950	0,956	0.0	0,050	5,330	0,026	0,001	0,047	5,064	9,235			
4h SAND - L/S = 0.3	210,019	10,019	60,000	0,260	0,208	0,740	0,792	0.3	0,350	5,760	0,020	0,001	0,036	4,561	7,886	25,221	11,116	8,274
	210,019	10,019	60,000	0,260	0,208	0,740	0,792	0.3	0,350	6,030	0,058	0,002	0,104	4,775	21,846			
	210,019	10,019	60,000	0,260	0,208	0,740	0,792	0.3	0,350	5,960	0,078	0,003	0,140	4,719	29,724			
4h SAND - L/S = 0.3	210,019	10,019	60,000	0,260	0,208	0,740	0,792	0.3	0,350	5,870	0,076	0,003	0,137	4,648	29,405			
	210,019	10,019	60,000	0,260	0,208	0,740	0,792	0.3	0,350	5,610	0,092	0,004	0,165	4,442	37,246			
4h SAND - L/S = 0.3 - MIX	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,770	0,045	0,002	0,081	4,606	17,569	24,779	3,624	7,832
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	6,340	0,066	0,003	0,119	5,062	23,451			
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,750	0,065	0,003	0,117	4,591	25,466			
4h SAND - L/S = 0.3 - MIX	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,790	0,080	0,003	0,144	4,622	31,126			
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	6,030	0,064	0,003	0,115	4,814	23,909			
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,740	0,068	0,003	0,122	4,583	26,687			
4h SAND - L/S = 0.3 - MIX	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,730	0,065	0,003	0,117	4,575	25,554			
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,690	0,058	0,002	0,104	4,543	22,963			
	210,019	10,019	60,000	0,260	0,202	0,740	0,798	0.3	0,350	5,570	0,065	0,003	0,117	4,447	26,289			

CLAY																		
SAMPLE	**SAMPLE MASS [g]**	**H_2O INITIAL [ml]**	**H_20 ADDED [ml]**	**H INITIAL**	**H FINAL**	**TS INITIAL**	**TS FINAL**	**L/S [l/kg]**	**L/S REAL [L/kg]**	**MASS CALCIMETRY [g]**	**VOL. CO_2 CALCIMETRY [L]**	**nCO_2 [mol]**	**mCO_2 [g]**	**mDW [g]**	**gCO2 / kg DW**	**AVERAGE g CO2 / kg DW**	**S.D. g CO2 / kg DW**	**Δ g CO2 / kg DW**
0h CLAY - L/S = 0.3				0,228		0,772				0,490	0,068	0,003	0,122	0,378	323,206	280,993	44,980	
				0,228		0,772				0,500	0,060	0,002	0,108	0,386	279,478			
				0,228		0,772				0,480	0,059	0,002	0,106	0,371	286,271			
				0,228		0,772				0,490	0,068	0,003	0,122	0,378	323,206			
				0,228		0,772				0,510	0,064	0,003	0,115	0,394	292,265			
				0,228		0,772				0,480	0,070	0,003	0,126	0,371	339,644			
				0,228		0,772				0,520	0,056	0,002	0,101	0,402	250,814			
				0,228		0,772				0,510	0,040	0,002	0,072	0,394	182,666			
				0,228		0,772				0,500	0,049	0,002	0,088	0,386	228,241			
				0,228		0,772				0,480	0,064	0,003	0,115	0,371	310,531			
				0,228		0,772				0,480	0,061	0,002	0,110	0,371	295,975			
				0,228		0,772				0,500	0,058	0,002	0,104	0,386	270,162			
				0,228		0,772				0,500	0,049	0,002	0,088	0,386	228,241			
				0,228		0,772				0,490	0,068	0,003	0,122	0,378	323,206			
4h CLAY - L/S = 0.3	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,490	0,070	0,003	0,126	0,401	314,295	296,163	55,115	15,169
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,520	0,059	0,002	0,106	0,425	249,622			
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,500	0,082	0,003	0,147	0,409	360,810			
4h CLAY - L/S = 0.3	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,520	0,060	0,002	0,108	0,425	253,853			
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,500	0,067	0,003	0,120	0,409	294,808			
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,480	0,078	0,003	0,140	0,392	357,510			
4h CLAY - L/S = 0.3	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,480	0,042	0,002	0,076	0,392	192,505			
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,490	0,073	0,003	0,131	0,401	327,764			
	259,740	59,740	0,000	0,228	0,183	0,772	0,817	0.0	0,299	0,490	0,070	0,003	0,126	0,401	314,295			
4h CLAY - L/S = 0.6	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,560	0,052	0,002	0,094	0,373	250,991	255,269	30,889	-25,724
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,520	0,057	0,002	0,103	0,346	296,288			
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,490	0,046	0,002	0,083	0,326	253,749			
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,540	0,038	0,002	0,068	0,359	190,209			

CLAY																		
SAMPLE	**SAMPLE MASS [g]**	**H_2O INITIAL [ml]**	**H_20 ADDED [ml]**	**H INITIAL**	**H FINAL**	**TS INITIAL**	**TS FINAL**	**L/S [l/kg]**	**L/S REAL [L/kg]**	**MASS CALCIMETRY [g]**	**VOL. CO_2 CALCIMETRY [L]**	**nCO_2 [mol]**	**mCO_2 [g]**	**mDW [g]**	**gCO2 / kg DW**	**AVERAGE g CO2 / kg DW**	**S.D. g CO2 / kg DW**	**Δ g CO2 / kg DW**
4h CLAY - L/S = 0.6	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,500	0,048	0,002	0,086	0,333	259,486			
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,490	0,047	0,002	0,085	0,326	259,265			
4h CLAY - L/S = 0.6	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,560	0,057	0,002	0,103	0,373	275,124			
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,550	0,047	0,002	0,085	0,366	230,981			
	259,740	59,740	60,000	0,375	0,335	0,625	0,665	0.3	0,599	0,490	0,051	0,002	0,092	0,326	281,330			
4h CLAY - L/S = 0.6 - MIX	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,490	0,048	0,002	0,086	0,322	267,830	265,026	12,645	-15,967
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,530	0,051	0,002	0,092	0,349	263,093			
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,530	0,050	0,002	0,090	0,349	257,934			
4h CLAY - L/S = 0.6 - MIX	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,550	0,055	0,002	0,099	0,362	273,410			
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,560	0,052	0,002	0,094	0,368	253,881			
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,530	0,051	0,002	0,092	0,349	263,093			
4h CLAY - L/S = 0.6 - MIX	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,510	0,046	0,002	0,083	0,335	246,605			
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,560	0,055	0,002	0,099	0,368	268,528			
	259,740	59,740	60,000	0,375	0,342	0,625	0,658	0.3	0,599	0,470	0,050	0,002	0,090	0,309	290,862			

ANNEX VII: PORTLAND SAMPLES DATA

In this section, the characteristics of 5 samples of Portland material are communicated by a table. The concerns their physical-chemical properties (pH, color, dry fraction), concentrations of metals and hydrocarbons, and the limit values established by the Italian law (Ministerial Decree about the individuation of non-hazardous wastes subjected to simplified procedures for their recovery).

	Unit of measure	Limits	Limits reference	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Test number				20LA15591	20LA15564	20LA15566	20LA17554	20LA15561
CER Code				17 01 01 Cement	17 01 01 Cement	17 01 01 Cement	17 01 01 Cement	17 01 01 Cement
Sample description				Concrete residue	Concrete residue	Concrete residue	Concrete residue	Concrete residue
Sampled by				ECAMRICERT technician	ECAMRICERT technician	ECAMRICERT technician	ECAMRICERT technician	ECAMRICERT technician
Sampling location				Via Dante Alighieri, Loc. Ponte del Ghelpack, Fraz. Canove, Roana, VI	Via Cinquevie 32, Zugliano, VI	Via Ponte degli Alpini, Marano Vicentino, VI	Via J. F. Kennedy 4/A, Riese Pio X, TV	Via Bojadori, Cogollo del Cengio, VI
Sampling date				28/10/2020	28/10/2020	28/10/2020	26/11/2020	28/10/2020
Analysis start date				28/10/2020	28/10/2020	28/10/2020	26/11/2020	28/10/2020
Analysis end date				10/11/2020	10/11/2020	10/11/2020	07/12/2020	10/11/2020
Cgemical-Physical analysis								
Physical aspect				Non-powdery solid	Non-powdery solid	Non-powdery solid	Non-powdery solid	Non-powdery solid
Color				varied	varied	varied	Grey	varied
Dry Fraction at 105°C	%			67,3	61,1	90,1	87,7	51,7
pH	Unità di pH			12	12	8	10	12
Antimony	mg/kg			< 10,00	< 10,00	< 10,00	< 10,00	< 10,00
Arsenic	mg/kg			< 5,00	< 5,00	< 5,00	< 5,00	< 5,00
Cadmium	mg/kg			< 10,00	< 10,00	< 10,00	< 10,00	< 10,00
Cobalt	mg/kg			< 10,00	< 10,00	< 10,00	< 10,00	< 10,00
Chrome	mg/kg			< 10,00	13,32	10,73	< 10,00	15,8
Chrome VI	mg/kg			< 1,00	< 1,00	< 1,00	2,6	8,3
Mercury	mg/kg			< 5,00	< 5,00	< 5,00	< 5,00	< 5,00

	Unit of measure	Limits	Limits reference	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Molybdenum	mg/kg			< 5,00	< 5,00	< 5,00	< 5,00	< 5,00
Nickel	mg/kg			< 10,00	< 10,00	< 10,00	< 10,00	11,35
Lead	mg/kg			< 10,00	< 10,00	< 10,00	< 10,00	< 10,00
Copper	mg/kg			< 10,00	13,03	< 10,00	< 10,00	15,71
Selenium	mg/kg			< 5,00	< 5,00	< 5,00	< 5,00	< 5,00
Vanadium	mg/kg			< 10,00	13,6	14,27	14,02	14,66
Zinc	mg/kg			67,07	133,49	43,52	51,49	125,73
Total hydrocarbons	mg/kg			< 200	< 200	< 200	< 200	< 200
Analysis performed on the eluate								
COD	mg/L	30	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.3	13	15	< 10,00	24	20
pH	Unità di pH	5,5 + 12	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.4	> 12,0	> 12,0	11	12	> 12,0
Arsenic	µg/L	50	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.5	< 0,10	< 0,10	0,47 ± 0,09	0,48 ± 0,10	< 0,10
Barium	mg/L	1	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.6	1,53 ± 0,28	2,70 ± 0,50	0,026 ± 0,005	1,00 ± 0,19	1,82 ± 0,34
Beryllium	µg/L	10	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.7	< 0,20	< 0,20	< 0,20	< 0,20	< 0,20
Cadmium	µg/L	5	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.8	< 0,10	< 0,10	< 0,10	< 0,10	< 0,10
Cobalt	µg/L	250	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.9	1,33 ± 0,26	3,42 ± 0,66	0,44 ± 0,09	1,71 ± 0,33	2,63 ± 0,51
Total Chrome	µg/L	50	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.10	92,00 ± 17,00	63,00 ± 11,00	34,5 ± 6,2	44,0 ± 7,9	66,0 ± 12
Mercury	µg/L	1	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.11	< 0,10	< 0,10	< 0,10	< 0,10	< 0,10
Nickel	µg/L	10	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.12	1,19 ± 0,26	1,20 ± 0,26	0,48 ± 0,10	0,78 ± 0,17	0,87 ± 0,19
Lead	µg/L	50	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.13	0,85 ± 0,19	1,99 ± 0,45	< 0,70	1,16 ± 0,26	1,49 ± 0,33
Copper	mg/L	0,05	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.14	0,003 ± 0,001	0,016 ± 0,003	0,007 ± 0,001	0,005 ± 0,001	< 0,003
Selenium	µg/L	10	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.15	< 0,30	< 0,30	< 0,30	< 0,30	< 0,30
Vanadium	µg/L	250	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.16	1,46 ± 0,31	1,49 ± 0,31	30,5 ± 5,5	3,24 ± 0,68	1,51 ± 0,32
Zinc	mg/L	3	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.17	< 0,005	0,007 ± 0,001	< 0,005	0,009 ± 0,002	< 0,005
Chlorides	mg/L	100	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.18	< 5	< 5	< 5	< 5	< 5
Fluorides	mg/L	1,5	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.19	0,28 ± 0,04	0,23 ± 0,04	< 0,1	0,32 ± 0,05	0,28 ± 0,04
Nitrates	mg/L	50	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.20	1,7 ± 0,3	0,53 ± 0,08	0,74 ± 0,12	1,1 ± 0,2	0,67 ± 0,10
Solfates	mg/L	250	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.21	2,3 ± 0,4	1,8 ± 0,3	35 ± 3	1,3 ± 0,2	2,1 ± 0,4

	Unit of measure	Limits	Limits reference	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Cyanides	µg/L	50	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.22	< 30	< 30	< 30	< 30	< 30
Asbestos	mg/L	30	D.M. 05/02/1998 (coord. D.M. n. 186/2006) - All.23	< 1	< 1	< 1	< 1	< 1
Hazardous class				None	None	None	None	None
Sample preparation				UNI EN 15002:2015	UNI EN 15002:2015	UNI EN 15002:2015	UNI EN 15002:2015	UNI EN 15002:2015
Classification				NON HAZARDOUS	NON HAZARDOUS	NON HAZARDOUS	NON HAZARDOUS	NON HAZARDOUS

ANNEX VIII: LEACHING TESTS RESULTS

In this annex the results obtained through the leaching tests, according the UNI EN 12457-2004 standard, are reported. These analyses have been performed on the carbonated sample, exhibiting the maximum absorption of CO_2, and the initial sample, non-carbonated, to examine the variation on the release of metals due to the accelerated carbonation.

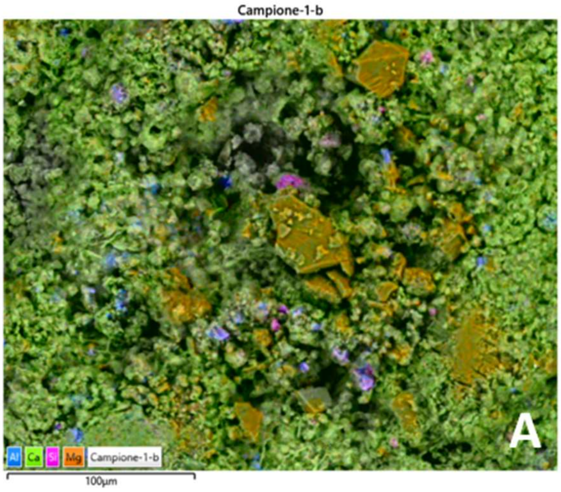
	ID	Ba (µgBa/l)	Be (µgBe/l)	Cd (µgCd/l)	Co (µgCo/l)	Cr (µgCr/l)	Cu (µgCu/l)	Mo (µgMo/l)	Ni (µgNi/l)	Pb (µgPb/l)	V (µgV/l)	Zn (µgZn/l)
PRE CARBONATION	PORT - L/S = 0.0	1645	10	10	10	68	73,8	10	10	10	10	10
	PORT - L/S = 0.0	1500	10	10	10	62	31,5	10	31,2	10	10	10
	PORT - L/S = 0.0	1405	10	10	10	38,6	21,5	10	10	10	10	10
POST CARBONATION	PORT - L/S = 0.3 - MIX	40,8	10	10	10	24,8	38,4	13,4	53	10	10	10
	PORT - L/S = 0.3 - MIX	41,9	10	10	10	10	31,2	12,7	37,4	10	10	10
	PORT - L/S = 0.3 - MIX	35,1	10	10	10	10	74,3	12	24,6	10	10	10

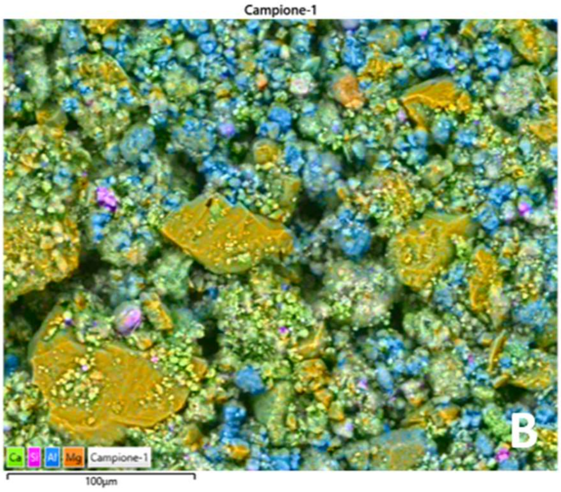
	ID	Ba (µgBa/l)	Be (µgBe/l)	Cd (µgCd/l)	Co (µgCo/l)	Cr (µgCr/l)	Cu (µgCu/l)	Mo (µgMo/l)	Ni (µgNi/l)	Pb (µgPb/l)	V (µgV/l)	Zn (µgZn/l)
PRE CARBONATION	SAND - L/S = 0.0	16,5	10	10	10	10	14,6	41,1	10	10	10	12,6
	SAND - L/S = 0.0	14,2	10	10	10	10	10	34,3	10	10	10	10
	SAND - L/S = 0.0	19,1	10	10	10	10	23,6	30,1	10	10	10	11,3
POST CARBONATION	SAND - L/S = 0.3 - MIX	18,2	10	10	10	10	10	23,5	10	10	10	28,6
	SAND - L/S = 0.3 - MIX	18,7	10	10	10	10	10	25,3	10	10	10	10
	SAND - L/S = 0.3 - MIX	29,4	10	10	10	10	10	22,4	10	10	10	11

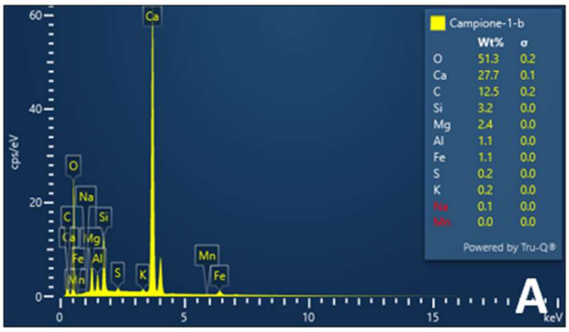
	ID	**Ba** **(µgBa/l)**	**Be** **(µgBe/l)**	**Cd** **(µgCd/l)**	**Co** **(µgCo/l)**	**Cr** **(µgCr/l)**	**Cu** **(µgCu/l)**	**Mo** **(µgMo/l)**	**Ni** **(µgNi/l)**	**Pb** **(µgPb/l)**	**V** **(µgV/l)**	**Zn** **(µgZn/l)**
PRE CARBONATION	CLAY - L/S = 0.0	10	10	10	10	10	10	10	10	10	14,8	10
	CLAY - L/S = 0.0	10	10	10	10	10	10	10	10	10	13,4	10
	CLAY - L/S = 0.0	10	10	10	10	10	10	10	10	10	13,7	10
POST CARBONATION	CLAY - L/S = 0.3	10	10	10	10	10	10	10	10	10	11,8	11,9
	CLAY - L/S = 0.3	10	10	10	10	10	10	10	10	10	11,3	10
	CLAY - L/S = 0.3	10	10	10	10	10	10	10	10	10	11,3	10

ANNEX IX: XRD TESTS RESULTS

This annex presents all the information obtained with the X-Ray Diffraction method; the figures signed with "A" represent the non-carbonated samples with a L/S = 0.0, while the figures signed with "B" concern the carbonated samples with a L/S = 0.3 mixed every hour inside the incubator.







Campione-1-b

	Wt%	σ
O	51.3	0.2
Ca	27.7	0.1
C	12.5	0.2
Si	3.2	0.0
Mg	2.4	0.0
Al	1.1	0.0
Fe	1.1	0.0
S	0.2	0.0
K	0.2	0.0
Na	0.1	0.0
Mn	0.0	0.0

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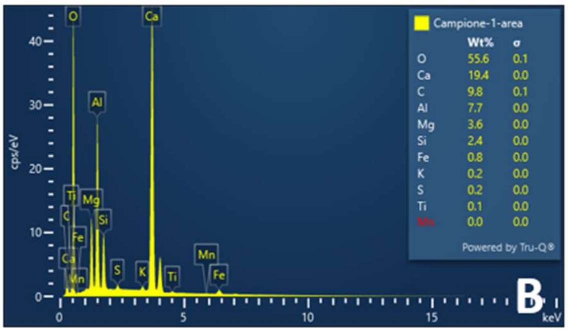

Campione-1-area

	Wt%	σ
O	55.6	0.1
Ca	19.4	0.0
C	9.8	0.1
Al	7.7	0.0
Mg	3.6	0.0
Si	2.4	0.0
Fe	0.8	0.0
K	0.2	0.0
S	0.2	0.0
Ti	0.1	0.0
Mn	0.0	0.0

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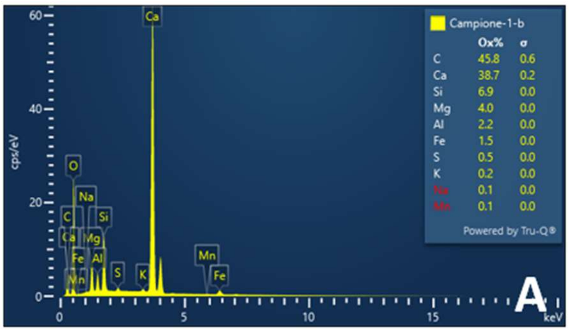
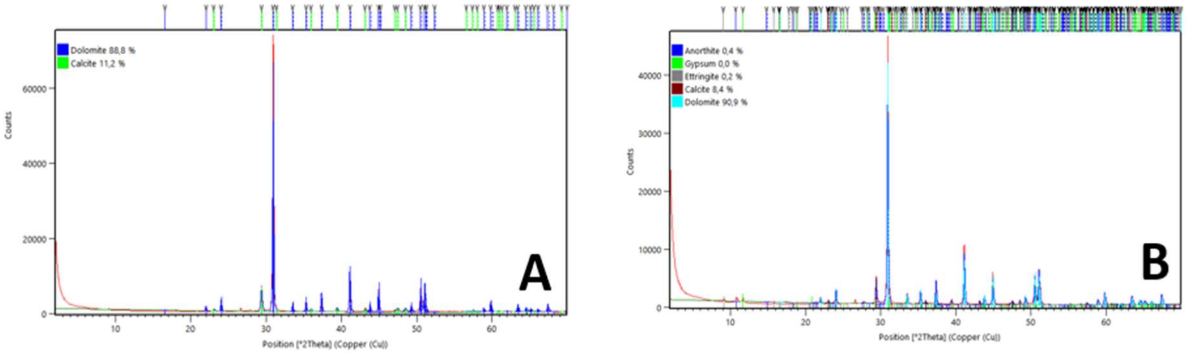


Campione-1-b

	Ox%	σ
C	45.8	0.6
Ca	38.7	0.2
Si	6.9	0.0
Mg	4.0	0.0
Al	2.2	0.0
Fe	1.5	0.0
S	0.5	0.0
K	0.2	0.0
Na	0.1	0.0
Mn	0.1	0.0

Campione-1-area

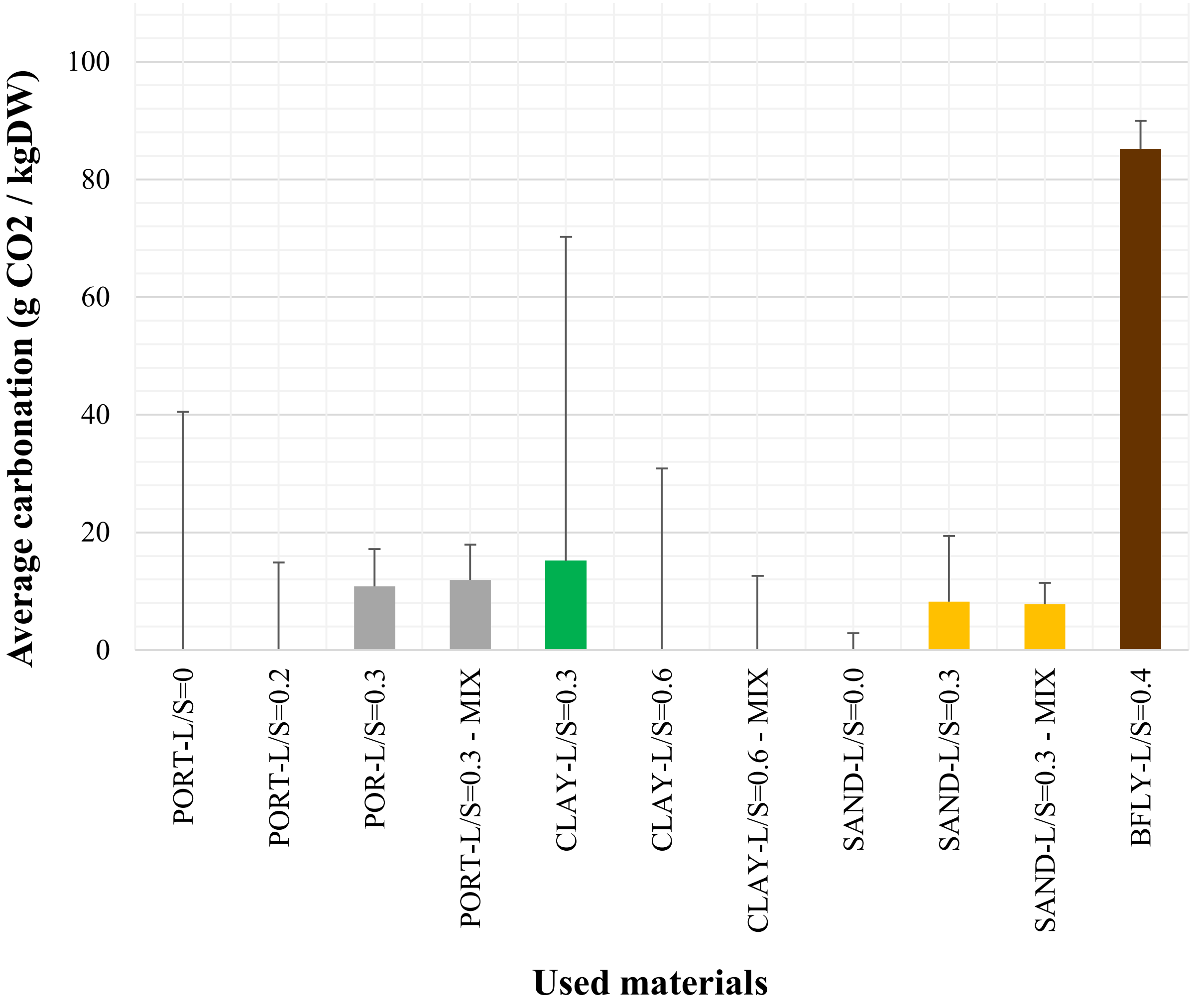
	Ox%	σ
C	40.3	0.6
Ca	29.5	0.2
Al	15.8	0.1
Mg	6.4	0.0
Si	5.7	0.0
Fe	1.2	0.0
S	0.5	0.0
K	0.3	0.0
Ti	0.3	0.0
Mn	0.1	0.0



ANNEX X: GRAPH OF THE EXPERIMENTAL RESULTS

In this annex, the average quantity of CO_2 absorbed by each material is shown by a graph in which the values are divided in each different L/S condition. While in the following table, the data used to build the graph are shown.

ID	CONDITION	H INITIAL	L/S EFFECTIVE [l/kg]	N. TESTS	EXPERIMENTAL		ASSUMED	
					AVERAGE ABSORPTION g CO2 / kg DW	STANDARD DEVIATION g CO2 / kg DW	AVERAGE ABSORPTION g CO2 / kg DW	STANDARD DEVIATION g CO2 / kg DW
PORT-L/S=0.0	Time: 4h L/S=0.0 Water adding: NO Mix: NO	0,074	0,075	9	-13,585	40,549	0,000	40,549
PORT-L/S=0.2	Time: 4h Water adding: YES L/S=0.2 Mix: NO	0,216	0,275	9	-13,186	14,904	0,000	14,904
POR-L/S=0.3	Time: 4h L/S=0.3 Water adding: YES Mix: NO	0,273	0,375	9	10,866	6,318	10,866	6,318
PORT-L/S=0.3 - MIX	Time: 4h L/S=0.3 Water adding: YES Mix: YES	0,273	0,375	9	11,938	6,012	11,938	6,012
CLAY-L/S=0.3	Time: 4h L/S=0 Water adding: NO Mix: NO	0,228	0,299	9	15,169	55,115	15,169	55,115
CLAY-L/S=0.6	Time: 4h L/S=0.3 Water adding: YES Mix: NO	0,375	0,599	9	-25,724	30,889	0,000	30,889
CLAY-L/S=0.6 - MIX	Time: 4h L/S=0.3 Water adding: YES Mix: YES	0,375	0,599	9	-15,967	12,645	0,000	12,645
SAND-L/S=0.0	Time: 4h L/S=0 Water adding: NO Mix: NO	0,050	0,050	9	-4,684	2,922	0,000	2,922
SAND-L/S=0.3	Time: 4h L/S=0.3 Water adding: YES Mix: NO	0,260	0,350	5	8,274	11,116	8,274	11,116
SAND-L/S=0.3 - MIX	Time: 4h L/S=0.3 Water adding: YES Mix: YES	0,260	0,350	9	7,832	3,624	7,832	3,624
BFLY-L/S=0.4	Time: 4h L/S=0.4 Water adding: NO Mix: NO	0,280	0,390	9	85,153	4,827	85,153	4,827


Average carbonation (g CO2 / kgDW)
0
20
40
60
80
100
PORT-L/S=0
PORT-L/S=0.2
POR-L/S=0.3
PORT-L/S=0.3 - MIX
CLAY-L/S=0.3
CLAY-L/S=0.6
CLAY-L/S=0.6 - MIX
SAND-L/S=0.0
SAND-L/S=0.3
SAND-L/S=0.3 - MIX
BFLY-L/S=0.4
Used materials

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