SLUTRAPPORT

Användning av industriell alkalirik restprodukt för kombinerad CO_2-bindning och produktion av cement med lågt CO_2-innehåll

Utilization of Industrial Alkaline-Rich Residues for Combined CO_2-sequestration and Production of Low Carbon Cement.

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RE:
SOURCE

Slutrapport för projekt:

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Förord

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Innehåll

1. Sammanfattning

Övergången till en cirkulär ekonomi och de höga CO_2-utsläppen är de två stora utmaningar som världen står inför idag. Detta projekt hanterar båda utmaningarna samtidigt genom att undersöka möjligheten att använda industriella alkalirika restprodukter i CO_2-inbindning genom indirekt karbonatiseringsprocess och använda produkten och de fasta restprodukterna från denna process i cement- och betonganvändning som mineraliskt tillsatsmaterial (SCM). Resultatet av detta projekt syftade till att minska CO_2-utsläppen från industriella processer genom accelererad karbonatisering av alkalirika restprodukter och sänka koldioxidavtrycket från cement och betong med SCM utvecklade från accelererad karbonatisering av alkalirester samt öka resurseffektiviteten.

Högvolymindustriella restprodukter från energi- och metallproduktion användes för CO_2-inbindning. De två alkalirika restprodukterna (filteraska från energiproduktion och metallurgisk slagg från metallproduktion) studerades i detta projekt. I processen, genom att optimera CO2-inbindningsreaktionerna, syftade projektet till att maximera mängden CO_2 som binds per enhet industriell restprodukt. Denna process producerade fälld kalciumkarbonat ($CaCO_3$) och fast restprodukt. Baserat på elementanalysen har fälld $CaCO_3$ från denna process liknande CaO-innehåll som indikerar samma renhet som den kommersiella CaCO3-produkten. Enligt renheten av produkten från denna process bör den fällda $CaCO_3$ som har högt värde användas i färg- och beläggningsindustrin och/eller massa- och pappersindustrin, istället för cement- och betonganvändningar. Utfallet av fällningen av Ca som finns i provet till $CaCO_3$ är cirka 70-90% när man jämför mellan den teoretiska beräkningen och experimentet. Processen producerar ~20 - 50 g fast restprodukt från 100 g råmaterial. Dess innehåll beror på källan till råmaterialet. Från beräkningen i detta projekt var den netto CO_2-inbindningen mellan 133 - 194 g CO_2/kg råmaterial i den indirekta karbonatiseringsprocessen.

I den andra delen av projektet studerades de karbonatiserade biprodukterna från CO_2-inbindningsprocessen – de karbonatiserade industriella restprodukterna – för deras potential att användas som mineraliskt tillsatsmaterial (SCM) som delvis kan ersätta vanlig Portlandcement (OPC). Målet var att utvärdera möjligheten att använda den fällda $CaCO_3$ och restprodukterna från den indirekta karbonatiseringsprocessen som SCM vilket kan leda till en minskning av cementrelaterade CO_2-utsläpp tillsammans med minskningen av exploateringen av primära resurser. Utvärderingen utfördes genom reaktivitetstest (R3) av materialen och hydratiseringsstudie av bindemedel (Portlandcement + SCM) jämfört med de klassiska SCM som kolflygaska och kalksten. Resultaten visade att Portlandcement blandat med fälld $CaCO_3$ har låg bearbetbarhet. För de fasta restprodukterna varierar resultaten med ursprunget av råmaterialet. Den kemiska sammansättningen av fast restprodukt från filteraska som producerades i denna studie överskred cementets kemiska sammansättningsstandard, dvs. klorid, sulfat och LOI. Därför kommer det inte att vara en bra kandidat för SCM. Å andra sidan har den fasta restprodukten som produceras från metallurgisk slagg potential att användas i cement- och betonganvändningar som SCM med en 15% ersättningsnivå på grund av reaktiviteten och hydratiseringsreaktionen i cementbindningssystemet. Ytterligare studier kommer att vara nödvändiga för att bekräfta andra egenskaper hos bindemedlet med den fasta restprodukten som produceras från metallurgisk slagg.

Från beräkningen som endast gäller materialen i bindemedlet är de ekvivalenta CO_2-utsläppen från bindemedlen med 15 % ersättning av de fasta restprodukterna från den indirekta karbonatiseringsprocessen lägre än Portlandcement med cirka 25 – 30%, medan bindemedlet med vanliga SCM (kolflygaska och kalksten) minskar endast 15% ekvivalenta CO_2-utsläpp jämfört med Portlandcement.

2. Summary

The transition to a circular economy and the high CO_2 emissions are the two major challenges that the world faces nowadays. This project deals with both challenges simultaneously by investigating the possibility of utilizing industrial alkaline-rich residues in CO_2 sequestration by the indirect carbonation process and using the product and the solid residues from this process in the cement and concrete application as supplementary cementitious materials (SCM). The outcome of this project aimed to reduce CO_2 emissions from industrial processes through accelerated carbonation of alkaline-rich residues and lower the carbon footprint of cement and concrete with SCM developed from accelerated carbonation of alkaline residues as well as increase resource efficiency.

High-volume industrial residues from energy and metal production were used for CO_2 sequestration. The two alkaline-rich residues (filter ash from energy production and metallurgical slag from metal production) were studied in this project. In the process, by optimizing the CO_2 sequestration reactions, the project aimed to maximize the amount of CO_2 sequestrated per unit industrial residue. This process produced precipitated calcium carbonate ($CaCO_3$) and solid residue. Based on the elemental analysis, precipitated $CaCO_3$ from this process has similar CaO content indicating the same purity as the commercial $CaCO_3$ product. According to the purity of the product from this process, the precipitated $CaCO_3$ which has high value should be used in the paint and coatings industry and/or pulp & paper industry, instead of cement and concrete applications. The yield of the precipitation of Ca presenting in the sample into $CaCO_3$ is around 70-90% when compared between the theoretical calculation and experiment. The process produces ~20 - 50g of solid residue from 100 g of feedstock. Its content depends on the source of the feedstock. From the calculation in this project, the net CO_2 capture was between 133 - 194 g CO_2/kg feedstock in the indirect carbonation process.

In the second part of the project, the carbonated by-products of the CO_2 sequestration process – the carbonated industrial residues – were studied for their potential to be used as supplementary cementitious material (SCM) which can partially replace Ordinary Portland

Cement (OPC). The objective was to evaluate the possibility of using the precipitated $CaCO_3$ and the residues from the indirect carbonation process as SCM which can lead to a decrease in cement-related CO_2 emissions along with the reduction in the exploitation of primary resources. The evaluation was carried out by reactivity test (R3) of the materials, and hydration study of binders (Portland cement + SCM) compared to the classical SCM such ash coal fly ash and limestone. The results showed that the Portland cement blended with precipitated $CaCO_3$ has low workability. For the solid residues, the results vary with the origin of the feedstock. The chemical composition of solid residue from filter ash produced in this study exceeded the cement chemical composition standard i.e. chloride, sulfate, and LOI. Thus, it will not be a good candidate for SCM. On the other hand, the solid residue produced from metallurgical slag has the potential to be used in cement and concrete applications as SCM with a 15% replacement level due to the reactivity and hydration reaction in the cementitious system. Further studies will be necessary to confirm other properties of the binder using the solid residue produced from metallurgical slag.

From the calculation concerning only the materials in the binder, the equivalent CO_2 emissions of the binders using 15 % replacement of the solid residues from the indirect carbonation process is lower than Portland cement by approximately 25 – 30%, whereas the binder using common SCMs (coal fly ash and limestone) reduce only 15% equivalent CO_2 emissions compared to Portland cement.

3. Introduction and Background

Introduction: As society aims to become more sustainable, there is a need to identify individual pathways to reach zero net greenhouse gas emissions in 2045 which is the goal for Sweden. One such high-potential pathway is to enhance resource efficiency through processes that enable the use of industrial residues/wastes–and thus address the circularity of resources. At the same time, there is also a need to mitigate the damages that have already occurred due to past human actions, especially the high atmospheric CO_2 concentration resulting from relentless anthropogenic CO_2 emissions. In line with Sweden's goal, and achieve net negative emissions, process industries have developed various roadmaps to reach climate neutrality. For example, the cement industry and steel industry have identified Carbon Capture Utilization and Storage (CCUS) in their respective roadmaps ("Färdplan Cement för ett Klimatneutral Betongbyggande [1]; Klimatfärdplan för en Fossilfri och Konkurrenskraftig Stålindustri Sverige [2]) as an important measure that can contribute to their efforts to reach climate neutrality. This work addresses both the circularity of resources and climate mitigation through the valorization of industrial alkaline residues/wastes to reduce CO_2 emissions and the carbon footprint of concrete.

Background:

Among the different CCUS approaches, ex-situ mineral carbonation is a promising pathway [3,4]. This technology involves the reaction of Ca/Mg with CO_2 to form thermodynamically stable carbonates, thus enabling permanent storage of the captured CO_2 [3]. Alternatively, products from mineral carbonation could be used in various industries such as the construction and the pulp and paper industries. The global CO_2 sequestration potential of mineral carbonation has been estimated to be two orders of magnitude larger than that of geological storage [5]. Examples of potential feedstocks for mineral carbonation are minerals such as natural serpentine, industrial residues such as metallurgical slags, and mine tailings [6]. Most of these feedstocks contain substantial amounts of Ca and/or Mg mainly present in stable silicate mineralogical forms that exhibit relatively low reactivity towards CO_2 even at high temperatures and CO_2 pressures [3,7,8]. Therefore, such feedstocks are subjected to indirect carbonation involving several steps as shown in Figure 1, including [3,6–8]: (a) extraction of Ca and/or Mg from their mineralogical forms with suitable solvents, (b) separation of extracted Ca/Mg from unreacted feedstock, (c) bubbling of a gas containing CO_2 (e.g., flue gas) through the solution enabling CO_2 to react with the Ca and/or Mg ions to form carbonates.

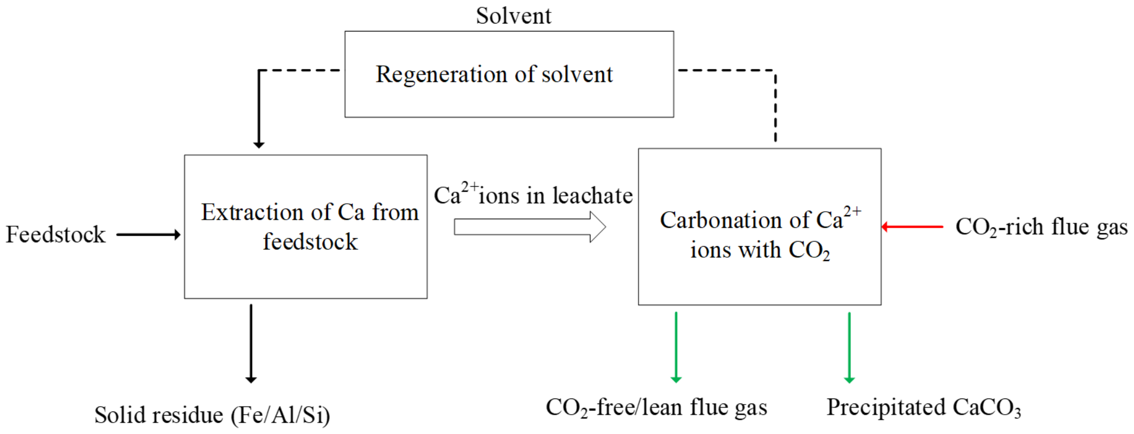

Figure 1 Schematic of indirect carbonation process

In addition to enabling a better optimization of CO_2 sequestration, indirect aqueous carbonation offers the possibility to produce marketable products such as carbonates, metals, and rare earth metals [6]. RISE together with academic and industrial partners are developing carbonation processes wherein industrial alkaline residues/wastes are used for CCUS. Examples of these residues are metallurgical slags, cement kiln dust, boiler ash, and mine tailings. The annual availability of these residues/wastes is > 50 M tons, and their overall CO_2 sequestration potential is in the order of millions of tons. Mineral carbonation may be more economically viable for decentralized CCUS from small (<0.5 M tons of CO_2/year) point source emitters where economics of scale is more favourable. There are 63 points source emitters in Sweden with CO_2

levels in the range 0.1–0.5 M tons/year giving a total CO_2 emission of 14.5 M tons/year (biogenic and abiogenic CO_2) [9]. Examples of point source emitters of CO_2 in this range are Höganäs Sweden AB and Lidköping Energi AB which have CO_2 emission levels in the range 0.1–0.25 M tons/year. These companies are open to alternative CCUS technologies that are decentralized and economically viable than geological-based CCS. While ex-situ mineral carbonation helps to solve the problem of atmospheric CO_2 using secondary resources, it raises the question of the management of carbonated by-products. Their utilization as supplementary cementitious materials (SCM) to replace some of the cement used in concrete can offer multiple advantages [10], including the utilization of carbonated by-products, as well as a reduction in cement production that is known to be responsible for 8% of the total global anthropogenic CO_2-emission. In Sweden, 5–6 million cubic meters of concrete are used annually [11]. Due to rising population, Boverket projects that 600 000 new houses would have to be constructed by the year 2025 [11]. Thus, the demand of concrete and consequently cement is expected to rise as the years go by. To reduce the CO2 emissons linked to cement and concrete, one solution is to replace a part of cement with other materials called SCM. SCMs are often by-products from other industries. In the standard SS-EN 197-1:2011(E), the cement substitution varies from a few percent for CEM I to 95% for CEM III/C. The level of substitution of cement with SCM depends on various factors such as chemistry or particle size that modify the cement properties. For example, limestone ($CaCO_3$) is limited to 35% maximum (CEM IIB-L) while slag can be used up to 95 % (CEM III/C). Nowadays, the availability of SCM is limited in comparison to the demand for cement. Therefore, new types of materials are being studied as potential SCM. RISE is at the forefront studying the potential for utilization of different resources as SCM. The carbonates and the alumino-silicaterich by-products of mineral carbonation could be used as SCM in concrete. This offers three advantages: permanent or long-term storage of CO_2, reduction of the carbon footprint of concrete by efficient use of secondary resources, and valorization of industrial alkaline-rich residues.

In this project, conditions that can enable permanent or long-term storage of CO_2 from industrial processes as carbonates in buildings have been studied. In addition to CO_2 sequestration, the project will contribute to resource efficiency since residues used for carbon capture will produce carbonates and by-products that can be used to replace some of the cement needed for building construction–thereby also reducing the carbon footprint of the concrete industry. Partners in the project include point source emitters of CO_2–Lidköping Energi AB (LE), Höganäs Sweden AB (HB); producers of industrial alkaline residues–LE, and HB; and research institute–RISE Research Institutes of Sweden AB.

Aim:

The aims of the project are:

1. Assess the possibility of using industrial alkaline-rich residues for CO_2 capture via a carbonation process.
2. Assess the possibility of using products from the carbonation of industrial alkaline-rich residues as SCM to reduce the carbon footprint of concrete.
3. Enable a circular use and thus enhance resource efficiency of industrial alkaline-rich residues through CCUS applications.

The knowledge acquired from this pre-study project will pave the way for a more detailed follow-up study.

Project goals

To meet the aim of the work the following objectives were defined:

1. Theoretically and experimentally determine the amount of CO_2 that can be captured and the amount of carbonates that can be produced with various industrial alkaline residues via indirect carbonation.

2. Identify and evaluate carbonated by-products that can be used as SCM to reduce the carbon footprint of cement. This will be done using standard R3 tests for SCM reactivity.

4. Experimental methods

This transdisciplinary project combines expertise from different research streams to simultaneously solve practical problems related to the utilization of industrial alkaline-rich residues as secondary resources, high atmospheric CO_2 concentration, and high rate of exploitation of natural resources. Boiler ash fraction supplied by Lidköping Energi AB, and metallurgical slag supplied by Höganäs Sweden AB were used as feedstocks. The work was carried out in the following work packages (WP):

WP1. CO_2 capture with alkaline residues–carried out by Placid Atongka Tchoffor and Anders Höije: Indirect aqueous carbonation experiments were carried out to determine the amount of CO_2 that can be captured with the applied feedstocks. The feedstocks, solvents and method applied are briefly described below:

Feedstocks: A boiler ash fraction–filter ash (herein denoted as FA) was supplied by Lidköping Energi AB. Metallurgical slag–electric arc furnace slag (referred to as Petrit E at Höganäs Sweden AB, and herein denoted as PE) was supplied by Höganäs Sweden AB. The elemental composition of each of the feedstocks was determined with standard analytical techniques such as inductively coupled plasma optical emission spectroscopy (ICP-OES). The elemental analysis provides the concentration of calcium (Ca) in each feedstock which is a first indication of the CO_2

sequestration potential of the feedstock. Generally, the higher the Ca content of a given feedstock, the higher the CO_2 sequestration potential of the feedstock. However, it is insufficient because the mineralogy, analsed by X-ray diffraction (XRD), can determine whether the Ca is available for carbonation. Ca in the form of CaO and $Ca(OH)_2$ are more readily available to react with CO_2 under dry conditions compared to calcium silicate forms [3,12,13].

Estimation of the maximum amount of CO_2 already stored in the feedstock: From the elemental composition of the feedstock, the maximum amount of CO_2 stored in the feedstock was estimated from its carbon content. Here, it was assumed that if the feedstock is heated in air at temperatures > 800 °C, all the carbon in the feedstock will be converted to CO_2. It was also assumed that carbon in the feedstock is present only as carbonates.

Determination of the theoretical maximum CO_2 capture capacities of the feedstocks: The theoretical CO_2 storage potential (TCO_2) of the feedstock was first determined from the elemental composition (on an oxide basis) of the feedstock using Stenoir's equation below [14].

$$TCO_2 = 0.785\ (\%\ CaO – 0.7\%\ SO_3) + 1.09\%\ MgO + 0.71\%\ Na_2O + 0.468\%\ K_2O \qquad (1)$$

Thereafter, the net theoretical maximum CO_2 capture capacity of the feedstock was determined by subtracting the amount of CO_2 that is already stored in the feedstock T_{CO2}.

Experimental method:

Indirect aqueous carbonation experiments were carried out in two main steps namely the leaching step, and the carbonation step. These steps are briefly described below:

1. Leaching step: In the first step, calcium (Ca) and other leachable elements in the applied feedstocks were leached at room temperature with a 1.5 M solution of hydrochloric (HCl) acid with purities above 99.9 % were purchased from Fisher Scientific.

The obtained leachate was stored for the next step of the indirect aqueous carbonation process. The solid residue obtained from each experiment was properly rinsed with deionized water and dried to constant mass. The concentration of Ca ions (and other ions) in the leachate was determined with ICP-OES. The extraction efficiency (X_i) of element i from a given feedstock during leaching was calculated from the following equation [15].

$$X_{\mathrm{i}} = \frac{V_{\mathrm{l}} \times C_{\mathrm{i-l}}}{W_{feedstock} \times C_{\mathrm{i}-W}} \times 100 \qquad (2)$$

Where V_l is the volume (l) of the leachate, $C_{i\text{-}l}$ is the concentration (g/l) of element *i* (e.g., Ca) in the leachate determined from ICP-OES analysis, $C_{i\text{-}W}$ is the concentration (mass %) of element *i* in the applied feedstock, and $W_{feedstock}$ is the amount (g) of feedstock used in each experiment.

2. Carbonation: The leachate containing Ca ions was carbonated in step 2 of the indirect aqueous carbonation process. Since the pH of each of the leachates was < 3 while the optimal pH for carbonation is in the range 9–11 [16], the pH of each of the leachates was lifted to pH 10 by adding a 10 M solution of NaOH to the leachate. Sodium hydroxide (NaOH) with purities above 99.9 % was purchased from Fisher Scientific. Lifting of the pH was carried out in 2 steps–namely pH 5, and pH10 to determine substances in the leachate that can precipitate (and be recovered) from the leachate at these pH levels. The pH was monitored continuously while adding NaOH dropwise. When pH 5 was reached, the amount of added NaOH was noted and the formed precipitate was removed by centrifugation (5 min at 8000 rpm) followed by filtration and saved for later analysis. The liquid was returned to the beaker and additional NaOH was added until pH 10 was reached. Again, the amount of added NaOH was registered and the precipitate was removed in the same way as previously. The liquid was then returned to the beaker for carbonation. Carbonation of the leachate at pH 10 was carried out with simulated flue gas consisting of 15 % CO_2 in nitrogen gas–which reflects the concentration of CO_2 in the flue gas generally obtained from heat and power plants (7%–15 %) CO_2 in nitrogen gas). The gas was bubbled through the leachate for 90–120 minutes. The leachate was filtered, and the solids obtained were dried in an oven at 70 °C for 48 hours. The concentrations of Ca and other substances in the leachate were determined with ICP-OES. The carbonation efficiency (CE) was determined from equation 3 below:

$$CE = \frac{C_{ca-before} - C_{Ca-after}}{C_{Ca-before}} \times 100 \tag{3}$$

Where $C_{Ca\text{-}before}$ and $C_{Ca\text{-}after}$ are the concentrations of Ca in the leachate before and after carbonation respectively determined with ICP-OES.

The amount of CO_2 captured (in g CO_2/kg feedstock) during the experiment was calculated from equation 4 below:

$$CO_2\ captured\ = \frac{W_{Ca} \times M_{CO2}}{M_{Ca} \times W_{feedstock}} \times 1000 \tag{4}$$

Where W_{Ca} is the mass of Ca in the leachate that has been carbonated, M_{Ca} and M_{CO2} are the molar masses of Ca and CO_2 respectively, $W_{feedstock}$ is the mass of the feedstock used in the experiment.

WP2. CO_2 storage as carbonates in concrete by Natechanok Chitvoranund:

In this WP, the possibilities, and limitations of using carbonates as well as by-products from the carbonation process as SCM in cement for building construction were assessed at RISE.

The carbonates and by-products (or solid residues) from the indirect carbonation process obtained in WP1 were characterized to evaluate their compatibilities with standards regarding the use of SCM to replace cement in concrete. Different techniques were used to evaluate their performance as SCM.

Materials characterization: The oxide compositions of carbonates and by-products were characterized by micro X-ray fluorescence (µ-XRF) using the M4 Tornado from Brucker equipped with an Ag tube. The sample powder was placed in a plastic holder 1mm deep and 4 cm in diameter. The sample was analyzed under a vacuum with a scanning speed of 10 ms/pixel and a pixel size of 50 µm to ensure a good statistic of the result and to decrease the deviation.

The Loss on Ignition (LOI) was measured by burning 1 g of the sample in an alumina crucible at 1000 ˚C for two hours in a furnace from Carbolite. The samples were weighed precisely before and after the calcination. The LOI value was calculated as the weight loss (wt.%) from the initial weight, as shown in equation 5:

$$LOI = \frac{m_1 - m_2}{m_1} \times 100 \tag{5}$$

where m_1 is the initial mass of the test portion, expressed in grams and m_2 is the mass of the ignited test portion, expressed in grams.

Reactivity test (R3): The carbonate and by-product were studied by the R3 ("Rapid, Relevant, and Reliable") RILEM test [17]. This test involves isothermal calorimetry on model mixes at 40°C. The mixes consist of studied material, portlandite, and gypsum, with a portlandite-to-SCM ratio of 3:1 and an SO_3/Al_2O_3 molar ratio of 1, adjusted by adding gypsum. The powder is mixed with a 0.5 mol/L KOH solution, achieving a water-to-solid ratio of 1. Heat flow was recorded over 7 days at 40°C.

Hydration study: The cement hydration study was carried out with isothermal calorimetry, and the development of hydrated phases by Thermogravimetric Analysis (TGA) on cement pastes. Initially, the hydration study was planned to use X-ray diffraction (XRD) for the analysis, but the XRD machine had a problem during the project. Therefore, the samples were characterized by TGA instead of XRD in agreement with the working group.

The materials from the indirect carbonation process in WP1 are unprocessed combustion ash - filter ash (Ref FA), unprocessed metallurgical slag - Petrit E (Ref PE), solid residue – filter ash (Res

FA), solid residues – Petrie E (Res PE), and precipitated calcium carbonate (Cc), as shown in Figure 2. Coal fly ash (F) and limestone (L) are common supplementary cementitious materials (SCM) which were used to compare the properties with the studied materials. CEM I 52.5R was used as a reference for Portland cement (PC). The powders were homogenized before blending. The samples for the hydration study with a fixed water-to-binder ratio (w/b) of 0.45 using deionized water were studied are listed below:

- 100% PC (Ref)
- 85%PC + 15% Ref FA
- 85% PC + 15% Res FA
- 85% PC + 15% Ref PE
- 85% PC + 15% Res PE
- 85% PC + 15% Cc
- 85% PC + 15% L
- 85% PC + 15% F

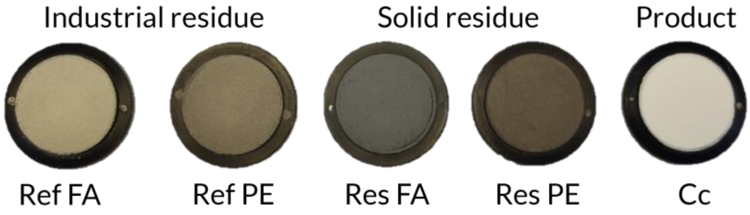

Figure 2 The materials from the indirect carbonation process in WP1

The heat development of binders indicates the hydration kinetics from the heat flow and the extent of the reaction from the cumulative heat. The heat flow of cement (CEM I) is presented in Figure 3 indicating the reactions in the cement hydration. For the heat evolution experiments using isothermal calorimetry (TAM Air, TA instrument), the dry binder was weighed carefully before mixing by hand. The cement paste was prepared with 10 g of dry binder and mixed by hand for 2 minutes with a spatula. Around 5g of the sample was transferred to a glass vial, sealed, and placed in the calorimeter directly after mixing. Deionized water with an amount that has a similar heat capacity to the binders was used as a reference sample. The heat release was measured for up to 7 days of hydration at 20 ˚C. The cumulative heat data per gram of total powder were collected after 45 min to let the sample equilibrate within the calorimeter.

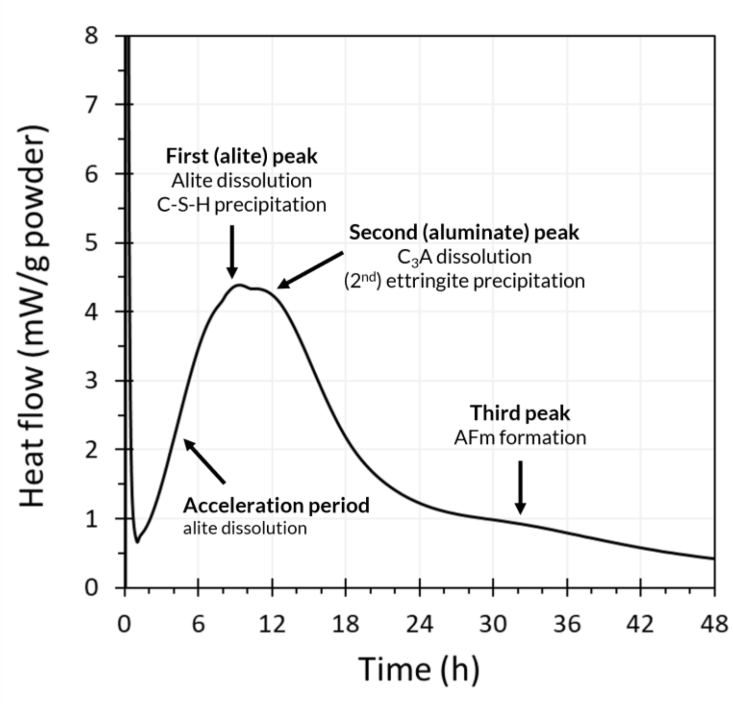

Figure 3 The heat flow of the hydration kinetics of cement (CEM I).

Each binder was blended by hand before adding deionized water and mixed at 500 rpm for 1 minute, then let to rest for 30 seconds to scrape the paste off the wall of the container. Then, the cement paste was mixed at 1600 rpm for 2 minutes and transferred into a 50 ml plastic container in two layers. The container was tapped 60 times after each layer to remove air bubbles. The sample container was sealed with the cap and paraffin film and then stored in the controlled chamber at 20 ˚C with 85% RH for 28 days.

The hydration was stopped by the solvent exchange method as prescribed in [19]. The cement pastes of 3 mm thickness were cut by a circular diamond saw with water-cooling. The sample discs were immersed in isopropanol for a week which was regularly changed after 1, 3, and 7 days. After 8 days in isopropanol, the samples were dried in a desiccator under a light vacuum at least 7 days before the TGA analysis. Before analyses, a small piece in the middle of the sample was polished to remove the potentially carbonated area and gently ground to fine powder by hand with agate mortar. About 35 mg of powder was placed in a 70 µL alumina crucible, then was analyzed by TGA (Mettler Toledo TGA-DSC 3+) from 20 – 1000 ˚C with the rate of 10 K/min and under 50 ml/min N_2 gas flow. Only one sample of each binder was measured. The hydrate phase identification was analyzed from the derivative TGA data. The weight loss from TGA analysis was used to calculate the amount of chemically bound water (BW) in equation 6 and the amount of portlandite (CH_{dry}) in equation 7.

$$BW = \frac{W_{30} - W_{520}}{W_{520}} \times 100 \qquad (6)$$

where W_t is the sample weight in grams at the temperature t (˚C) – the low index.

$$CH_{dry} = \frac{WL_{CH}}{W_{600}} \times \frac{M_{CH}}{M_{water}} = \frac{WL_{CH}}{W_{600}} \times \frac{74}{18} \qquad (7)$$

Where WL = weight loss due to the evaporation of water at the portlandite peak, M_{CH} = molecular weight of portlandite ($Ca(OH)_2$), and M_{water} = molecular weight of water (H_2O)

Mortar compressive strength: The materials produced from WP1 were insufficient to cast mortar bars. Therefore, it is not possible to test the compressive strength.

WP3. Life Cycle Assessment by Natechanok Chitvoranund and Viktoria Kozaric: It is not possible to calculate the full LCA due to the decrease in the RISE budget and lack of expertise. Therefore, a simplified potential environmental benefit gained was calculated by comparing the equivalent CO_2 emissions per cubic meter ($CO_{2\text{-ekv}}/m^3$) of different binders instead. The CO_2 emissions of binders with 15% replacement with different SCMs were compared to the reference Portland cement. The SCM used for a comparison in this part are solid residue from filter ash, solid residue from Petrit E, limestone, and coal fly ash. The calculation for CO_2 emissions of solid residues assumes that the indirect carbonation process uses green energy. Based on the data from WP1, the process consumes CO_2 to produce the solid residues. Therefore, the CO_2 emission for the two solid residues in this study is negative. The CO_2 emissions of limestone and coal fly ash were calculated based on the Embodied Carbon Footprint Databased (limestone = 0.01577 kg CO_2/kg and coal fly ash = 0.0416 kg CO_2/kg) from ICE database [20].

5. Result and discussion

5.1. CO_2 capture with alkaline residues

5.1.1. Elemental compositions of feedstocks:

The elemental compositions of the filter ash (FA) and electric arc furnace slag (PE) applied in this project are shown in Table 1 below. The feedstocks contain substantial amounts of Ca and some Mg providing a first indication that they could be used for CO_2 capture. FA contains substantial amounts of some trace elements e.g. S, Cl, alkalis, etc. indicating that that it may not be directly used as a construction material.

Element	Filter ash (FA)	Electrical arc furnace slag (PE)	Analysis method
Al, (mass %)	3.0	3.4	ASTM D 3682 (ICP-OES)
Si, (mass %)	4.9	6.6	
Fe, (mass %)	1.1	11	
Ti, (mass %)	1.7	0.48	
Mn, (mass %)	0.06	1.8	
Mg, (mass %)	1.4	11	
Ca, (mass %)	24.8	26	
Ba, (mass %)	0.44	0.07	
Na, (mass %)	1.9	0.10	
K, (mass %)	0.66	0.11	
P, (mass %)	0.23	0.10	
S, (mass %)	2.6	<0.05	SS 187186
Cl, (mass %)	14.6	1.7	Ion chromatography
C, (mass %)	1.8	1.7	SP method 0658
As, (mg/kg)	52	<20	ASTM D 3683 (ICP-OES)
Cd, (mg/kg)	56	<1	
Co, (mg/kg)	50	9	
Cr, (mg/kg)	260	1950	
Cu, (mg/kg)	7600	19	
Mo, (mg/kg)	38	22	
Ni, (mg/kg)	89	18	
Pb, (mg/kg)	4300	12	
V, (mg/kg)	53	590	
Zn, (mg/kg)	6500	1400	

Table 1. Elemental composition of the feedstocks.

5.1.2. Net theoretical CO_2 capture capacities of the feedstocks:

The theoretical CO_2 capture capacities of the feedstocks (~284 g CO_2/kg FA, and ~485 g CO_2/kg PE) estimated from equation 1 are shown in Figure 4 below. To obtain the net theoretical CO_2 capture capacity for each feedstock (that is, ~218 g CO_2/kg FA, and ~423 g CO_2/kg PE), the amount of CO_2 stored in each feedstock (that is, ~66 g CO_2/kg FA, and ~62 g CO_2/kg PE) was deducted from the theoretical capture capacity for each.

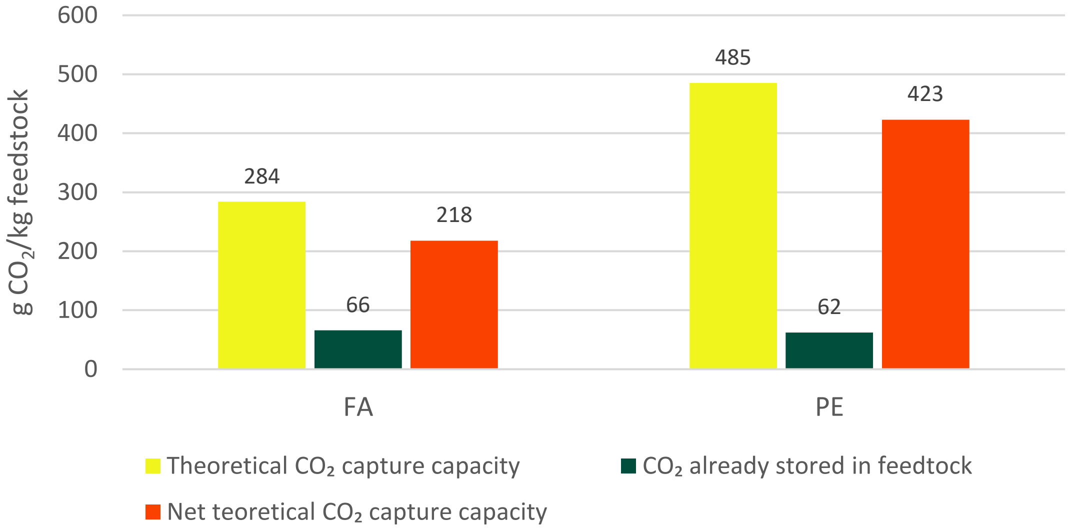

Figure 4. Net theoretical CO_2 capture capacities of the feedstocks.

5.1.3. Extraction of Ca and other elements from the leachate:

Figure 5 shows the extraction levels (mass %) of Ca, Mg, Al, Si, and Fe from the feedstocks during leaching. Over 50 % of Ca in both feedstocks were extracted during leaching. Some of the Ca that was extracted from the FA precipitated in the leachate as gypsum prior to the ICP-OES analysis of the leachate. Since only the Ca ions in the leachate was analyzed with ICP-OES, the actual extraction level of Ca from FA is higher than what is reported in Figure 5. In addition to Ca, the extraction levels of Mg, Al, Si, and Fe were significant from FA. However, except for Si and Al, the contents of these elements are very low in FA as shown in Table 1.

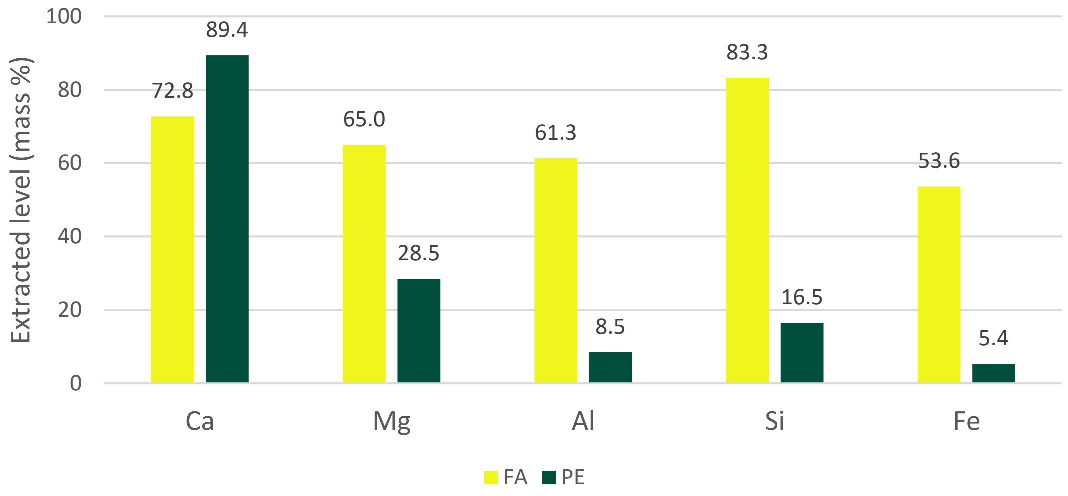

Figure 5. Extraction levels of Ca, Mg, Al, Si, and Fe from the different leachates: filter ash (FA), and petrit E (PE).

5.1.4. Carbonation of calcium ions in the leachates:

Prior to carbonation of calcium ions in the leachates, the pH of each of the leachates was lifted to 5 and 10 with a solution of NaOH as described in the method section. Figure 6 shows the precipitation of metals during the pH lifting process.

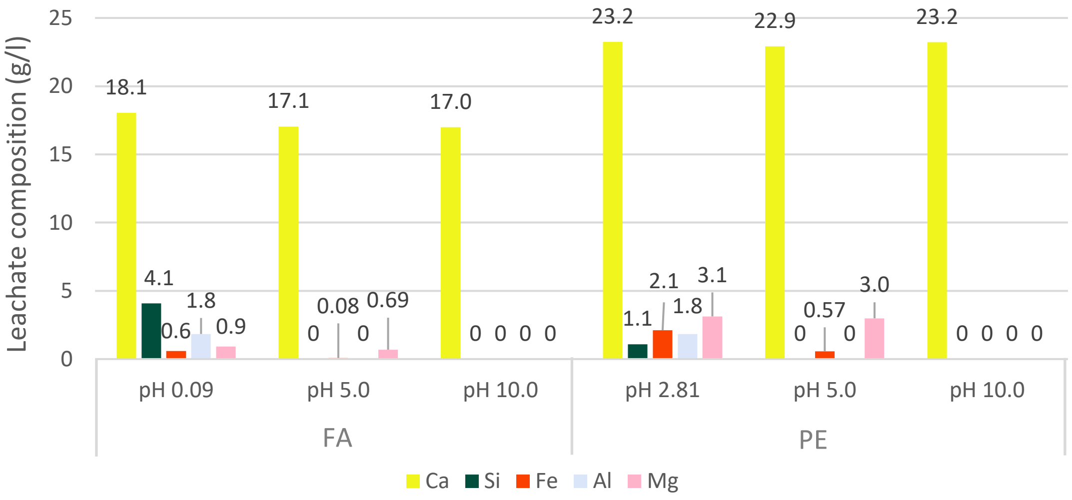

Figure 6. Concentrations of Ca, Si, Al, and Fe in the leachate at various pHs during pH lifting.

5.1.5. Carbonation efficiency of leachates obtained from leaching of the feedstocks:

ICP-OES analysis of each pH-adjusted leachate revealed that neglectable or no Ca ions were present in the leachate after carbonation. This implies that all Ca ions in each leachate were precipitated–as $CaCO_3$. Thus, the carbonation efficiency for the carbonation step of the indirect carbonation process (equation 3) of each leachate is ~100%.

5.1.6. Amount of CO_2 captured:

The overall amount of CO_2 captured during the indirect carbonation of each feedstock is shown in Figure 7. For PE, ~256 g of CO_2/kg of PE was captured while for FA, 199 g of CO_2/kg of FA was captured. As earlier mentioned, the feedstocks had some amount (~ 66 g CO_2/kg FA, and ~62 g CO_2/kg PE) prior to the experiments–which were likely released during the leaching step of the experiments where Ca was extracted from the feedstock. The amount of CO_2 that was stored in each feedstock was subtracted from the overall amount of CO_2 captured during the indirect carbonation experiment to give the net amount (~194 g of CO_2/kg PE, and ~133 g CO_2/kg FA) of CO_2 captured with each feedstock.

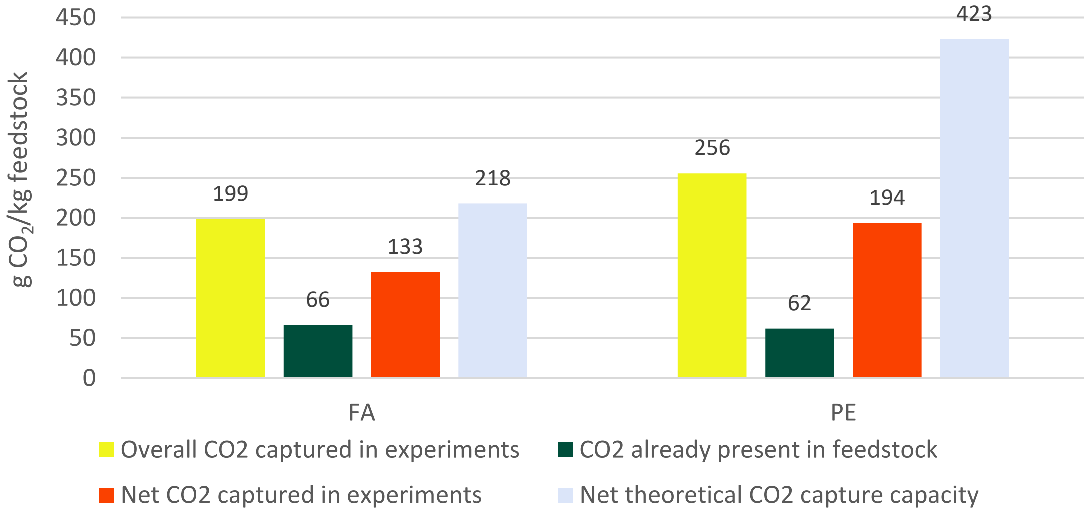

Figure 7. Net amount of CO_2 captured with each feedstock during the experiments. The net theoretical CO_2 capture capacity as well as the amount of CO_2 stored in each of the feedstock prior to the experiments are also included in the figure.

5.1.7. Compositions and yields of carbonation products:

The elemental composition (on oxide basis except for Cl) of the carbonation products obtained from the indirect carbonation of PE and FA are shown in Figure 8. The elemental composition of ACS grade $CaCO_3$ with purity 99.95 % purchased from Fisher Scientific is also shown in Figure 8. The elemental composition of the products obtained from the indirect carbonation of PE and FA are similar to that of the ACS grade $CaCO_3$–which suggests that the products obtained from PE and FA are $CaCO_3$ of high purity.

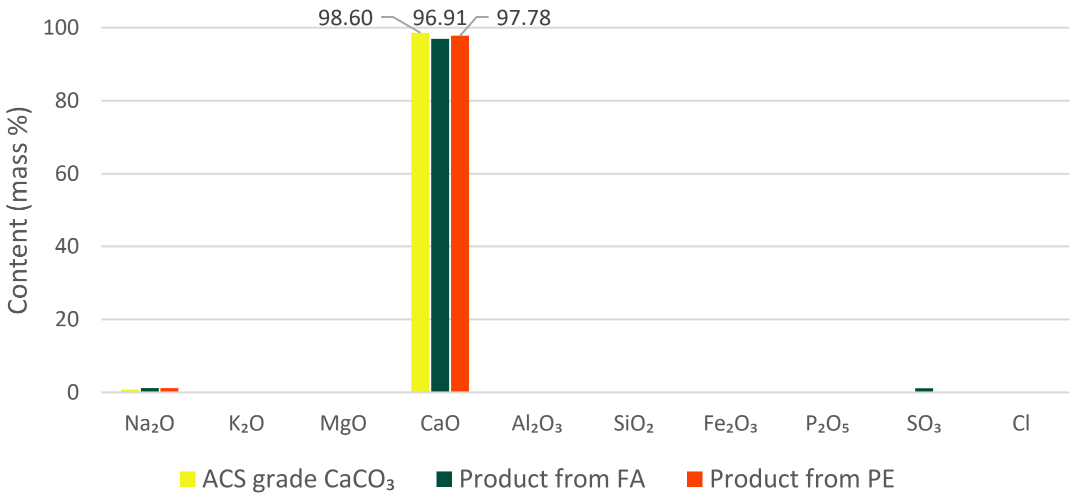

Figure 8. Elemental composition (on oxide basis, except for Cl) of the carbonation products obtained from PE and FA. The elemental composition of an ACS grade CaCO3 (purity 9.995 %) purchased from Fisher Scientific is also presented.

The elemental compositions (by XRF analysis) of the carbonation products obtained from the indirect carbonation of PE and FA are shown in Table 2. The elemental composition of ACS grade $CaCO_3$ with purity 99.95 % purchased from Fisher Scientific is also shown in the table. It shows that the compositions of the carbonation products are similar to that of the ACS grade $CaCO_3$–thus suggesting that their purities are comparable.

Oxides (mass %)	Analytical grade PCC (purity: 99.99 %)	$CaCO_3$ produced from FA	$CaCO_3$ produced from PE
Na_2O	0.77	1.19	1.20
K_2O	0.02	0.03	0.02
MgO	0.16	0.10	0.21
CaO	**98.60**	**96.91**	**97.78**
Al_2O_3	0.20	0.21	0.20
SiO_2	0.08	0.10	0.11
Fe_2O_3	0.00	0.00	0.00
P_2O_5	0.10	0.05	0.08
MnO	0.01	0.01	0.01
SO_3	0.03	1.11	0.07

Cl	0.00	0.11	0.10
Br	0.00	0.00	0.00
TiO_2	0.00	0.01	0.04
Cr_2O_3	0.00	0.00	0.00
Cu	0.00	0.00	0.00
ZnO	0.01	0.01	0.01
SrO	0.00	0.16	0.15
As	0.00	0.00	0.00
Mo	0.00	0.00	0.00
Cd	0.00	0.00	0.00
NiO	0.00	0.00	0.00
Pb	0.01	0.01	0.01
Se	0.01	0.00	0.00
Ba	0.00	0.00	0.00

Table 2. Composition of the carbonation products compared to analytical grade precipitated calcium carbonate

5.2. CO_2 storage in cement

The evaluation of potential use as supplementary cementitious materials (SCM) was carried out in 5 materials (see Figure 2) by their compositions, their reactivity, and the hydration study at 28 days. Unfortunately, the solid residue materials, as by-products of the indirect carbonation process, were produced in a small quantity. Therefore, the materials were not enough to evaluate the compressive strength.

5.2.1. Standard compatibilities

The chemical requirements of several standards were used as criteria for the materials to compare with in this study (see Table 3). The standard SS-EN 197-1:2011 is a European Standard defining the specifications of different types of cement and their constituents [21]. The chemical composition of the binder should meet this standard. Apart from the cement standard, there are some materials standards used as SCM such as silica fume for concrete (SS-EN 13263-1:2009), fly ash for concrete (SS-EN 450-1:2012), and GGBS for concrete (EN15167-1: 2006). SCMs used in concrete also have chemical requirements. The common chemical requirements in every standard are loss on ignition (LOI), chloride, and sulfate (SO_3). However, no heavy metal content is specified in the standards, but only regulations on these substances in materials when they shall be deposited as waste due to the risk of contamination of the ground and the groundwater.

Standard	Property	Requirements
SS-EN 197-1:2011 (cement composition)	Loss on ignition	≤ 5.0 %
	Insoluble residue	≤ 5.0 %
	SO_3	≤ 3.5 %for CEM I, II, IV, and V ≤ 4.0 % for CEM III
	Chloride content	≤ 0.10 %
SS-EN 13263-1:2009 (silica fume for concrete)	Element silicon	≤ 0.40 %
	Free CaO	≤ 1.0 %
	SO_3	≤ 2.0 %
	Chloride content	≤ 0.10 %
	Loss on Ignition	≤ 4.0 %
SS-EN 450-1:2012 (fly ash for concrete)	Free CaO	≤ 1.5 %
	SO_3	≤ 3.0 %
	Chloride content	≤ 0.10 %
	Loss on Ignition	≤ 9.0 %
	Reactive calcium oxide	≤ 10.0 %
	Na_2O equivalent	≤ 5.0 %
	Magnesium oxide	≤ 4.0 %
	Phosphate	≤ 5.0 %
EN 15167-1: 2006 (GGBS for concrete)	SO_3	≤ 2.5 %
	Chloride content	≤ 0.10 %
	Loss on Ignition	≤ 3.0 %
	Sulfite	≤ 2.0 %
	Magnesium oxide	≤ 18 %
	Moisture content	≤ 1.0 %

Table 3. The restriction of the substance specified in the standards.

The oxide compositions of the materials from the indirect carbonation process are given in Table 4. The reference filter ash (Ref FA) contains high sulfate, chloride, and LOI which exceeds all the standard requirements in Table 3. On the contrary, reference Petrit E (Ref PE) most of the oxide components of metallurgical slag are below the restriction in the standards, except LOI content when compared to the silica fume and GGBS standards.

The solid residue from filter ash (Res FA) has high sulfate and LOI content, while the chloride content slightly exceeds the standard limit. The oxide compositions of the solid residue from metallurgical slag Petrit E (Res PE) meets the restriction in the standards, except chloride and LOI.

Oxides (mass %)	**Reference FA (Ref FA)**	**Reference PE (Ref PE)**	**Solid residue FA (Res FA)**	**Solid residue PE (Res PE)**
Na_2O	3.87	0.00	0.00	0.00
K_2O	1.15	0.17	0.6	0.18
MgO	1.25	7.69	2.45	5.68
CaO	26.1	39.0	13.4	10.2
Al_2O_3	3.11	7.81	6.64	12.2
SiO_2	4.94	16.7	18.5	31.0
Fe_2O_3	1.18	19.1	1.99	27.3
P_2O_5	0.20	0.18	0.95	0.48
SO_3	**6.09**	**0.13**	**11.8**	**0.05**
Cl	**14.8**	**0.00**	**0.13**	**0.35**
TiO_2	2.39	0.99	17.0	1.67
Cr_2O_3	0.04	0.55	0.19	0.93
MnO	0.07	2.91	0.00	3.34
Cu	0.70	0.00	0.67	0.00
ZnO	0.85	0.08	1.31	0.05
SrO	0.00	0.00	0.12	0.00
NiO	0.00	0.00	0.02	0.00
Br	0.23	0.00	0.00	0.00
Pb	0.47	0.00	0.63	0.00
LOI*	**32.6**	**4.68**	**23.7**	**6.38**
SUM	100.0	100.0	100.0	99.7
Na_2O equivalent	**5.02**	**0.17**	**0.60**	**0.18**

*LOI = Loss on Ignition

Table 4. The chemical composition of the materials from the indirect carbonation process in WP1: unprocessed combustion ash - filter ash (Ref FA), unprocessed metallurgical slag - Petrit E (Ref PE), solid residue – filter ash (Res FA), solid residue – Petrie E (Res PE), and precipitated calcium carbonate (Cc).

5.2.2. Reactivity

The reactivity test is the material property testing (without cement). The cumulative heat per gram of SCM over time up to 7 days (168h) is shown in Figure 9. The reactivity of the studied materials (Ref FA, Ref PE, Res FA, Res PE, and Cc) was compared to the reactivity of the common

SCM (calcined clay, GGBS, and coal fly ash) from [22]. The studied materials show lower reactivity than calcined clay and GGBS but are comparable to coal fly ash, except Cc showing very low heat. The reactivity of industrial residue materials (Ref FA and Ref PE) is lower than solid residues (Res FA and Res PE) and coal fly ash. The solid residue PE shows a higher reactivity than the solid residue FA. Therefore, the two solid residues have the potential to be used as SCM in cement.

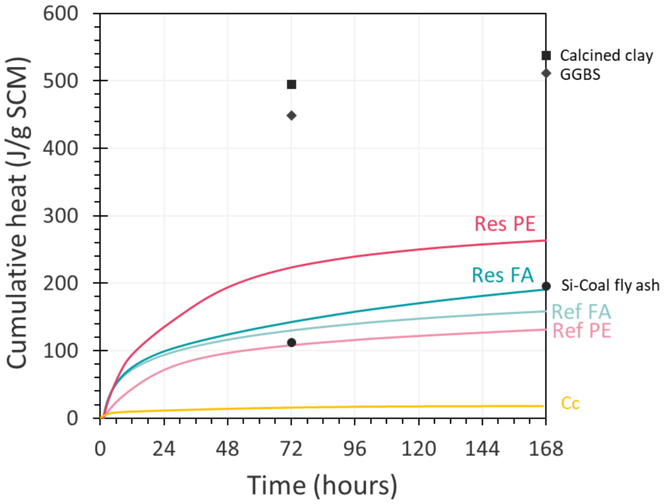

Figure 9. The cumulative heat up to 7 days from the R3 test of unprocessed combustion ash - filter ash (Ref FA), unprocessed metallurgical slag - Petrit E (Ref PE), solid residue – filter ash (Res FA), and solid residues – Petrie E (Res PE) compared to the reactivity of calcined clay, GGBS, and siliceous coal fly ash from literature [22].

5.2.3. Hydration study

The hydration study of different binders (Portland cement blended with 15% of the different material tested in this project) was investigated by the heat of hydration, hydrated phases composition, bound water content, and portlandite content at 28 days of hydration. Some small remarks were observed while mixing. The cement pastes of Portland cement blended with reference filter ash (PC + Ref FA) released a smell of ammonia due to the trace of ammonia in the source of filter ash. The paste of Portland cement with solid residue FA (PC + Res FA) was quite dry while mixing. The workability of Portland cement blended with precipitated $CaCO_3$ from WP1 (PC + Cc) is poor.

Heat of hydration: The heat flows of the investigated binders are shown in Figure 10. The hydration kinetics of the binder with feedstock FA differs from the others: the silicate and aluminate peaks are combined. This could lead to flash setting of the paste. To avoid this situation gypsum could be added to postpone the aluminium reaction. It is a well-known procedure and could be studied in the following project. The binder with solid residue FA demonstrates a slower reaction than Portland cement, probably due to the high sulfate content in solid residue FA. Portland cement with reference Petrit E binder (PC + Ref PE) reacts slightly slower than Portland cement blended with the solid residue PE (PC + Res PE). The binder incorporated with solid residue PE reacts similarly to Portland cement with coal fly ash (PC + F). However, the second aluminate peak of binder with solid residue PE is lower compared to the binder with reference PE and coal fly ash which an explanation is still unclear. The reaction of the binder with Cc is accelerated and shows a higher aluminate peak, probably due to a fine particle size which increases nucleation sites for hydrate formation. Moreover, the paste with Cc showed poor workability while mixing which is not a good property for cement and concrete applications.

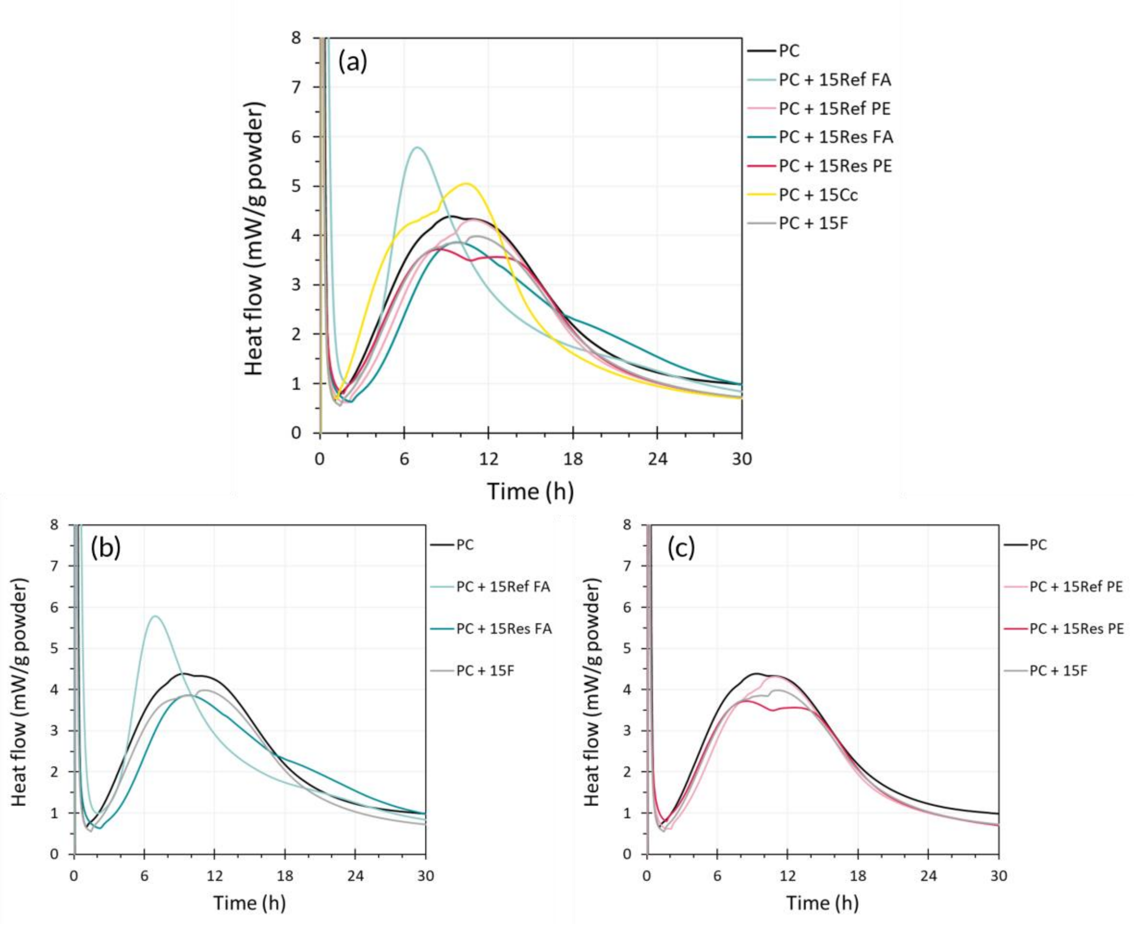

Figure 10. Heat flow curves of the investigated binders measured at 20 ˚C: PC – Portland cement, Ref FA – feedstock from filter ash, Ref PE – feedstock from metallurgical slag Petrit E, Res FA – solid residue from filter ash, Res PE – solid residue from Petrit E, Cc – Precipitated $CaCO_3$ from WP1, and F – coal fly ash. (a) all systems, (b) group of filter ash, and (c) group of metallurgical slag Petrit E.

The cumulative heat of the blended systems in Figure 11 shows lower heat release at 7 days than PC due to the dilution effect of 15% replacement. The binder with reference PE has low heat development similar to the fly ash system, while the other blended systems have higher heat than these two systems and are in the same range at 7 days. The reaction after 24 hours of the binder with solid residue PE is the lowest development, but it can reach 330 J/g powder at 7 days which is a similar range as the other blended systems. PC + Cc shows higher heat development than the other blended systems but slows down after 96 hours.

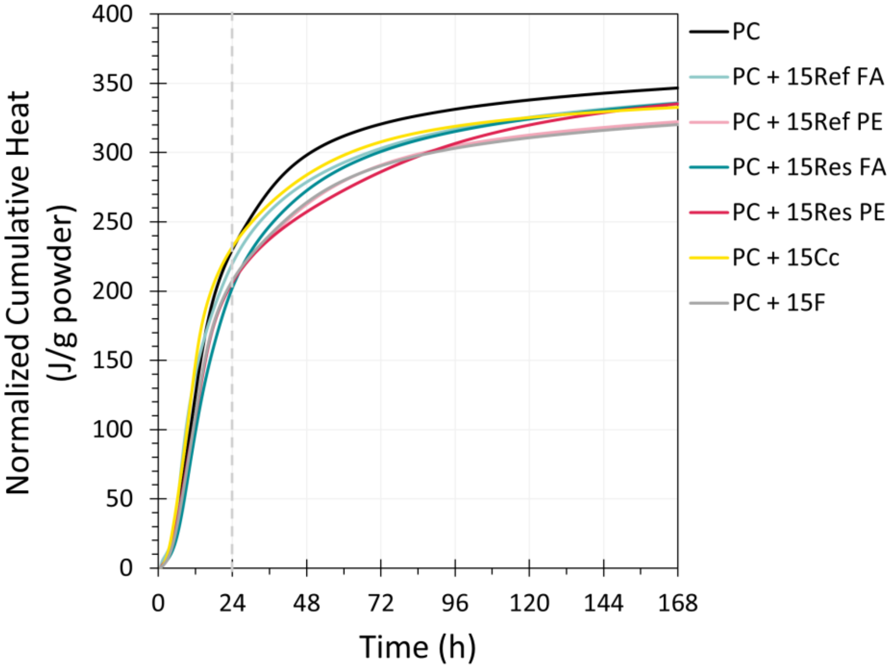

Figure 11. Cumulative heat release of the investigated binders at 20 days up to 7 days of hydration: PC – Portland cement, Ref FA – unprocessed filter ash, Ref PE – unprocessed Petrit E, Res FA – solid residue of filter ash, Res PE – solid residue of Petrit E, Cc – Precipitated $CaCO_3$ from WP1, and F – coal fly ash.

Hydrated phase identification: At 28 days, the cement pastes of different binders were investigated. The paste of the binder with solid residue FA had poor workability but could be able to cast in a cylinder, while the binder with Cc was not possible to cast due to a very dry mix. Therefore, only the binders with reference FA, solid residue FA, reference PE, and solid residue PE were investigated and compared to Portland cement, the systems with limestone, and the binder with coal fly ash. Note that the cracks were observed on top of the sample of PC + Ref FA after curing for 28 days in sealed condition, as shown in Figure 12.

Figure 12. The cracks in the hydrated paste of Portland cement blended with 15% of unprocessed filter ash after curing for 28 days.

From Figure 13, the hydrated phases formation was identified with TGA. In every binder, the usual cement hydrate: C-S-H (CaO_x-$(SiO_2)_y$-$H2O_z$), ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}.26H_2O$), and portlandite ($Ca(OH)_2$) are observed. All systems produce hemicarbonate ($Ca_4Al_2(OH)_{13}(CO_3)_{0.5}(H_2O)_5$) and monocarbonate ($Ca_4Al_2(OH)_{13}(CO_3)(H_2O)_5$), except the binder with reference FA which forms Friedel's salt ($Ca_2Al(OH)_6Cl(H_2O)_2$)due to the high chloride content in reference FA. However, the data from TGA cannot confirm which forms of carbonate-AFm (hemicarbonate (Hc) or monocarbonate (Mc)) are present in the binder. This would need further analysis using XRD. The blended cement with limestone shows a strong intensity of carbonates because limestone is composed of carbonate ($CaCO_3$).

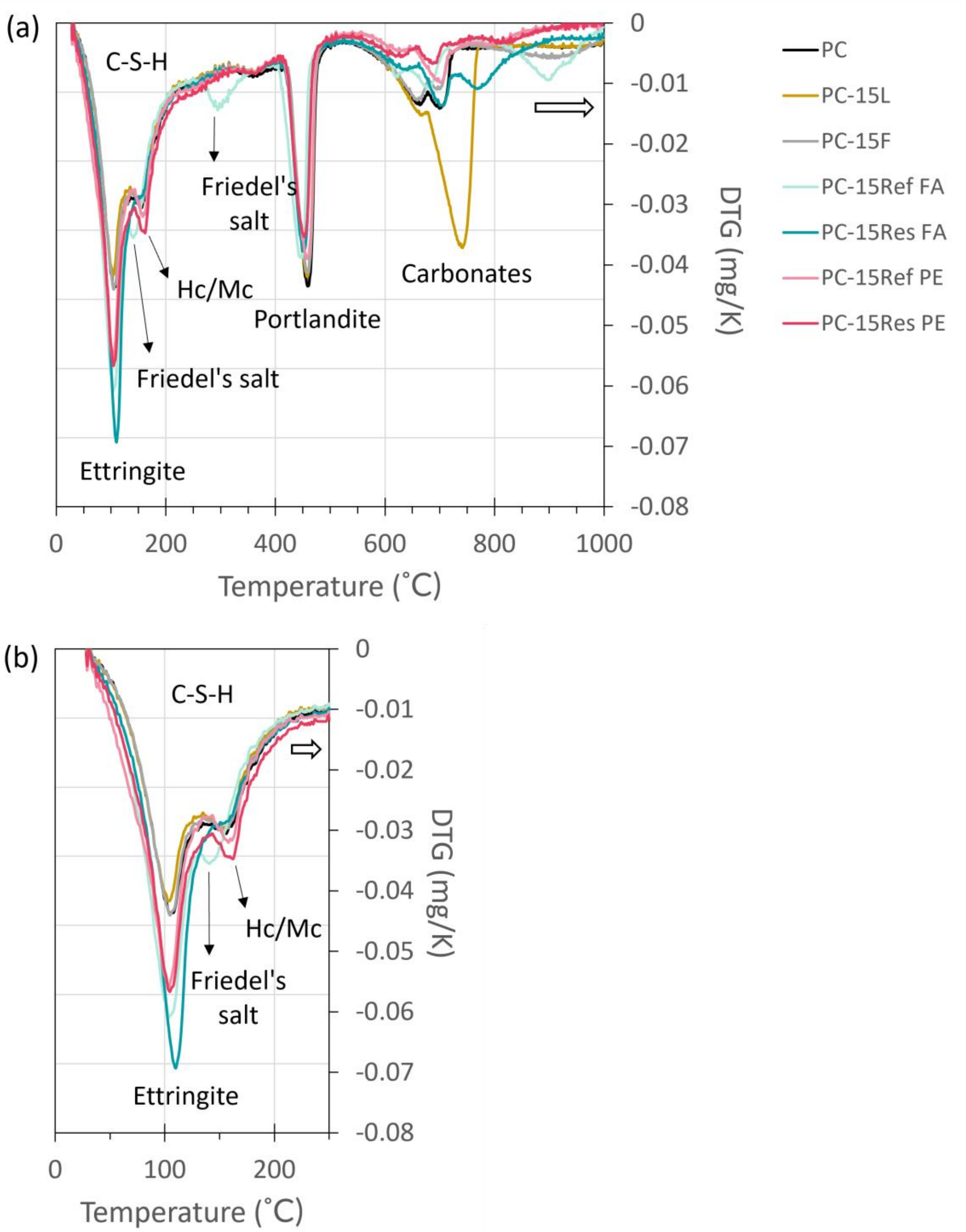

Figure 13. Hydrate phases of the investigated binders at 28d of hydration (a) temperature between 0 – 1000 ˚C and (b) zoom in between 0 – 250 ˚C: C-S-H = calcium silicate hydrates, Hc = hemicarbonate, and Mc = monocarbonate.

The bound water content of the investigated binders at 28 days is shown in Figure 14. The binders with common SCM (limestone and coal fly ash) have slightly lower bound water than Portland cement. On the contrary, the binders with the materials from WP1 have higher bound water content than Portland cement indicating that more hydrates are formed in the system. The binder with solid residue FA has the highest bound water probably due to a larger precipitation ettringite and Friedel's salt. The other binders with the studied materials have similar bound water content. The high bound water content indicates that the materials can react in the cementitious system.

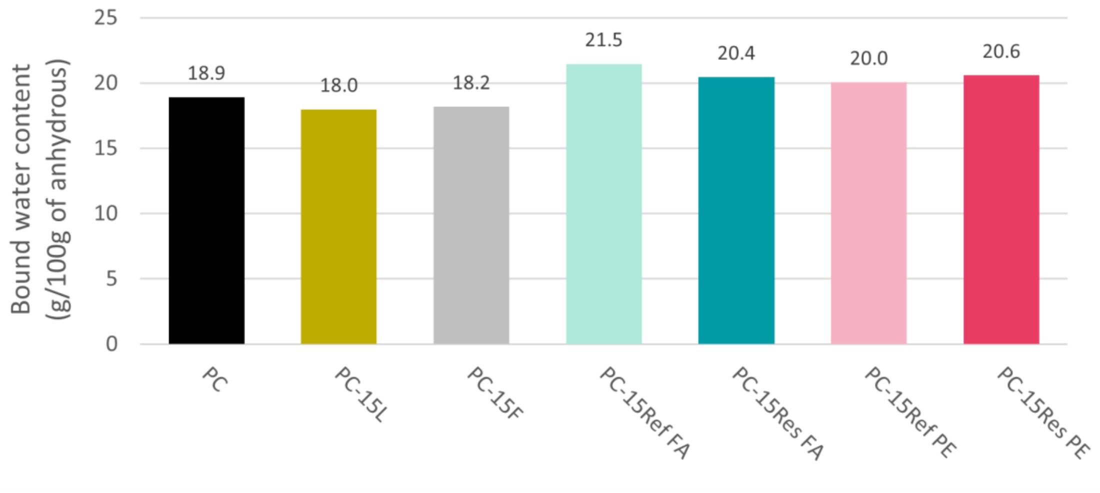

Figure 14. The bound water content of the investigated binders at 28 days.

Portlandite content, presented in Figure 15, is the hydration product from the reaction of water and calcium silicate phases in clinker. When SCM has a pozzolanic reaction, portlandite is consumed by amorphous SiO_2 and Al_2O_3 to form more C-S-H or C-A-S-H. Therefore, it possibly refers to the reactivity of SCM.

The Portlandite content of the PC at 28 days is higher than the other systems, as expected. Considering 15% replacement with SCM, the portlandite content in binders should be about 16 g/100 g of anhydrous. However, the blended systems show a similar or higher amount of portlandite than expected. When the amount of portlandite is higher than 16 g/100 g anhydrous, it is probably due to the filler effect and late reaction of SCM. With the filler effect, the SCM provides nucleation sites for hydrates accelerating the hydration of clinker. As a result, portlandite which is a main hydration product is formed. When the portlandite content is lower than 16 g/100 g of anhydrous, it could imply the pozzolanic reaction of SCM. Further study on hydration at later age is needed to confirm the reaction of SCM.

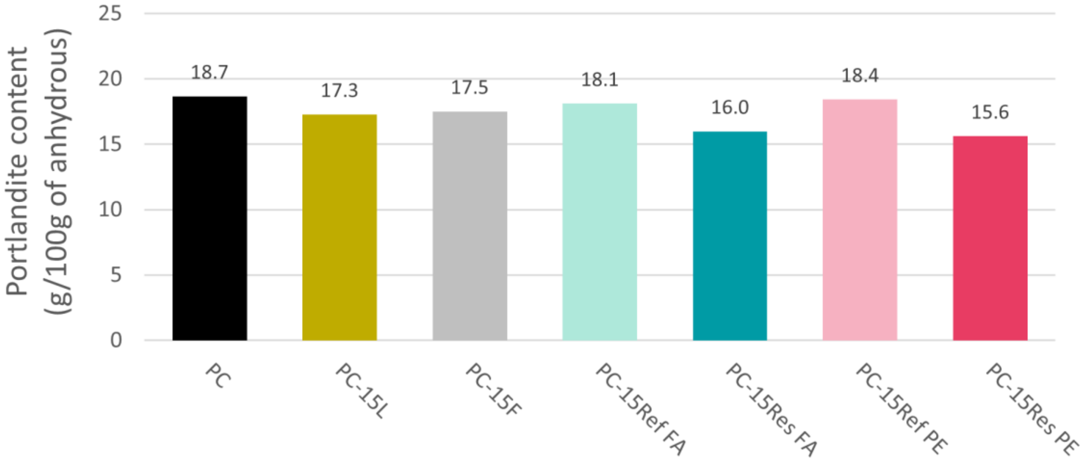

Figure 15. The portlandite content of the investigated binder at 28 days.

In summary, based on the data in a preliminary study, the solid residue Petrit E from the indirect carbonation process has potential use as SCM in cement and concrete applications. The chemical composition is under the requirement of cement composition. However, the high Fe_2O_3 content in solid residue PE should be further investigated. Even though the solid residue FA seems to be reactive in the cementitious system, the constraint is the chemical compositions that exceed the limit of the standard, such as high SO_3, TiO_2, and LOI. The precipitated calcium carbonate from the indirect carbonation process is not realistic to be used in cement and concrete applications.

5.3. CO_2 emissions

CO_2 emissions of different binders used in concrete were calculated and compared. The calculation assumes the energy used in the indirect carbonation process is from the green energy source meaning that it is equal to zero CO_2 emission from the process. About 90% of CO_2 emissions in concrete come from cement clinker production [23]. The common strength class for blended cement in ready-mix concrete is C30/37. According to the report from Svensk Betong in 2022 [23], the fresh concrete with this strength class using the w/b of 0.55 and a cement content in concrete of 400 kg/m^3 of concrete emits CO_2-equivalent approximately 255 kg $CO_{2\text{-ekv}}$/m^3 when using Portland cement. The calculation of the CO_2 emissions assumes that the cement content in concrete is 400 kg/m^3. Then, regular Portland cement generates about 230 kg $CO_{2\text{-ekv}}$/m^3. The calculation of CO_2 emissions of different binders, referring to Portland cement blended with 15% replacement of SCM, was based on this number. Limestone and coal fly ash are used as reference SCM for this calculation using the emission factor from the ICE database [20]. Note that the assumption of CO_2 emission from the ICE database includes the process and combustion in the calculation. From Figure 16, the blended system with the solid residue FA reduces approximately 33% $CO_{2\text{-ekv}}$, and the binder with the solid residue PE reduces approximately 26% $CO_{2\text{-ekv}}$ compared to Portland cement. As the indirect carbonation process produces less amount of the solid residue FA than the solid residue PE, the requirement of the FA feedstock is higher than the PE feedstock to produce the same amount of residue materials. It benefits for the CO_2 emission calculation of the binder with solid residue FA because the production can capture more CO_2 in the process. The binders with the common SCMs show roughly 15% CO_2 reduction compared to the Portland cement mix [20]. The CO_2 emissions of the binders containing the solid residues from the indirect carbonation process are less than the common SCMs thanks to the consumption of CO_2.

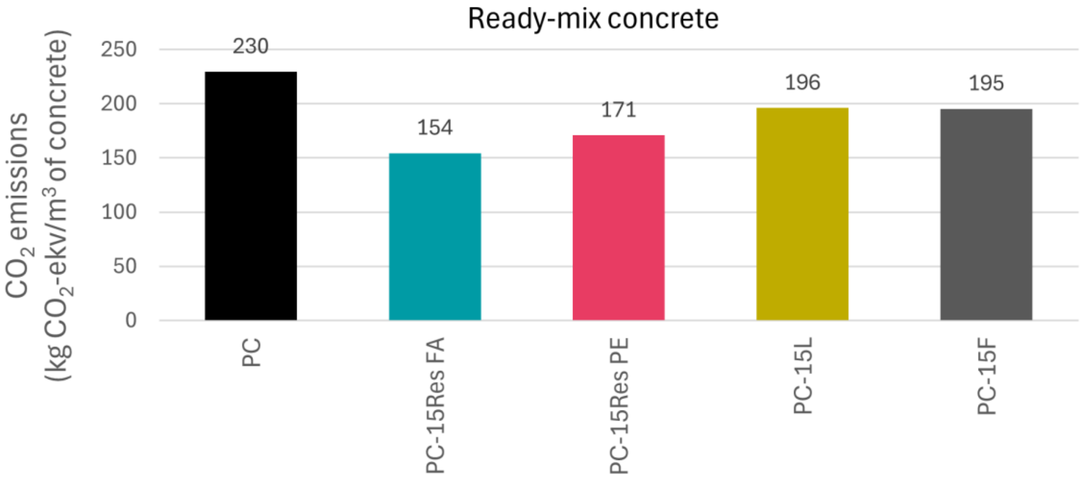

Figure 16. CO_2 emissions of different binders with 15% replacement of SCM of ready-mixed concrete exposed in a dry environment with the strength class of C30/37.

Based on the assumption of this calculation, the advantage of using the solid residue materials from the indirect carbonation process is a significant reduction of CO_2 emissions compared to common SCM. The indirect carbonation process produces ~19% of solid residue FA and ~48% of solid residue PE. Therefore, concrete with 15% replacement requires a lot of industrial residue materials (feedstock) to produce solid residues from indirect carbonation process used in concrete. As a result, the solid residue mixes can capture more CO_2.

6. Conclusions, utilization, and next steps

This study shows the possibility of producing high-purity calcium carbonate from industrial alkaline residues containing substantial calcium for CO_2 capture by indirect carbonation process and the initial assessment of using solid residues of the process in cement applications. The two sources of industrial alkaline residues for this study are filter ash from Lidköping Energi and metallurgical slag – Petrit E from Höganäs. The net theoretical maximum CO_2 capture capacity is approximately 260 – 400 g CO_2/kg feedstock. The extraction from feedstock can extract Ca out from the feedstock more than 90%. The indirect carbonation process generates 20 - 50% solid residue content depending on the types of feedstocks. The precipitated $CaCO_3$ product yield is between 70 - 90% with high purity (more than 95% CaO content in the product) comparable to commercial precipitated $CaCO_3$ grade ACS. As expected, the overall CO_2 captured in the experiments is slightly less than the net theoretical CO_2 capture capacity for filter ash, whereas the discrepancy is higher for metallurgical slag - Petrit E. However, the net CO_2 capture of metallurgical slag - Petrit E is higher than the filter ash.

The reactivity of the solid residue materials produced from the indirect carbonation process is comparable to siliceous coal fly ash, but still significantly lower than GGBS and calcined clay. The precipitated $CaCO_3$ is unsuitable in cement and concrete applications due to the price and low flowability when mixing with cement. However, the potential applications for precipitated $CaCO_3$ can be used in paint & coating, and pulp & paper industries. The solid residue FA has a low potential to be used as SCM in cementitious systems. The hydration of the cement blended with the solid residue FA is retarded due to high sulfate. Therefore, the solid residue PE from the indirect carbonation process shows a potential to be used as SCM with 15% replacement, but further study is needed to confirm other properties.

CO_2 emission calculation was simplified based on the assumption of using green energy for the indirect carbonation. In ready-mixed concrete, the CO_2 emission of the binder with 15% solid residue can be reduced by approximately 25 – 35% compared to Portland cement.

This study shows a positive assessment of continuing to work on using the solid residue from metallurgical slag - Petrit E as SCM in cement and concrete applications. However, other important properties, such as compressive strength, fresh properties of concrete, durability, etc., should be verified. The full life cycle assessment should be calculated to confirm the results of CO_2 capture of the product.

7. Publication list

No publication.

8. Project communication

The project communication was carried out through two different platforms:

1. RISE webpage – https://www.ri.se/en/what-we-do/expertises/carbonation-based-ccs

   Target group: Alkaline-rich materials producers, Waste management, Academic research

2. Linkedin of Placid Atongka Tchoffor - https://www.linkedin.com/posts/placid-atongka-tchoffor-phd-55729326_karbonatisering-ccus-ccs-activity-6951163942261948416-ur3j?utm_source=social_share_sheet&utm_medium=member_desktop_web

   Target group: Alkaline-rich materials producers, Waste management, Academic research

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10. Appendices

- Appendix 1: Budget report

RE:Source är ett strategiskt innovationsprogram som fokuserar på att utveckla cirkulära, resurseffektiva materialflöden. Vårt mål är att uppnå en hållbar materialanvändning där vi håller oss inom planetens gränser.

RE:
SOURCE

resource-sip.se