



Comparative study of direct and indirect aqueous mineral carbonation of construction and demolition waste fines for CO₂ sequestration

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ABSTRACT

Sustainable development prioritizes waste utilization to mitigate environmental impacts through recycling and reuse. In construction and demolition waste (C&DW) recycling, the recovery of recycled concrete aggregate generates C&DW fines. These fines contain significant calcium (Ca), making them potential feedstock for CO₂ sequestration through accelerated mineral carbonation processes. This approach offers a promising pathway for reducing CO₂ emissions from the cement industry by utilizing alkaline waste to form thermodynamically stable carbonates. This study compares the effectiveness of direct and indirect aqueous mineral carbonation in sequestering CO₂ using classified C&DW fines. Laboratory experiments assessed Ca extraction using borehole water (BHW) and acid mine drainage (AMD) as leaching agents, along with optimization of the solid-to-liquid (S/L) ratio for carbonation. Analysis of raw C&DW fines, leached residue, leachate, and the resulting solid carbonation product was performed using XRD, XRF, TGA, BET/BJH, PSD, and ICP-OES techniques. A 1:10 S/L ratio was optimal for Ca extraction and subsequent carbonation. BHW leached ~328 mg-Ca/L-BHW, significantly higher than AMD (85 mg-Ca/L-AMD). High concentration of SO₄²⁻ in AMD reacted with Ca to form gypsum, limiting Ca leaching into the solution. Thus, BHW was used in subsequent carbonation experiments. Direct aqueous carbonation achieved a total CO₂ uptake of 52.8 g-CO₂/kg-C&DW fines, whereas indirect aqueous carbonation captured 10.5 g-CO₂/kg-C&DW fines. This research highlights the benefit of repurposing C&DW as low-cost material for direct CO₂ capture. Moreover, the potential substitution of BHW with polluted waste streams like AMD underscores an integrated approach to waste valorization and CO₂ mitigation, particularly in water-scarce regions.

1. Introduction

Climate change, driven by greenhouse gas emissions, remains one of the most pressing environmental challenges of the 21st century. Carbon dioxide (CO₂), primarily emitted from anthropogenic activities such as fossil fuel combustion, high-emission industrial operations, and the production of construction materials, is a major contributor to global

warming [1]. Annually, global CO₂ emissions range between 35 and 40 gigatonnes (Gt). Out of that, 2.9 Gt is emitted by the cement and concrete industry [2]. Yet, current carbon capture efforts account for ~0.1 % of global emissions [3]. This remains significantly lower than the estimated 10 Gt per year of CO₂ removal required to keep the global temperature increase below 1.5 °C by 2050 [4]. Thus, the need for effective carbon mitigation strategies has led to increasing interest in

Abbreviations: AMD, acid mine drainage; BET, Brunauer-Emmet-Teller; BHW, borehole water; BJH, Barrett-Joyner-Halenda; CCUS, carbon capture, utilization, and storage; C&DW, construction and demolition waste; ICP-OES, inductively coupled plasma-optical emission spectroscopy; PSD, particle size distribution; SEM, scanning electron microscopy; XRD, X-ray diffraction; XRF, X-ray fluorescence; TGA, thermogravimetric analysis.

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carbon capture, utilization, and storage (CCUS) technologies [5].

CCUS strategies can be broadly categorized into in-situ and ex-situ methods, each with distinct advantages and applications [6,7]. In-situ mineral carbonation involves the geological sequestration of CO_2 in underground rock formations, promoting its reaction with naturally occurring alkaline minerals to form stable carbonates [8]. In contrast, ex-situ mineral carbonation occurs above the ground using reactive materials such as coal fly ash, metallurgical slag, and waste concrete [9]. For instance, Nyambura, et al. [10] demonstrated that two by-products from coal-fired power plants, fly ash and leaching agent brine, capture 71.84 kg of CO_2 per ton of fly ash through aqueous mineral carbonation. Mineral carbonation, an accelerated ex-situ method, has emerged as a promising method for long-term CO_2 sequestration, mimicking the natural weathering process in which alkaline minerals react with CO_2 to form thermodynamically stable carbonate compounds. This approach provides a permanent storage option for medium CO_2 emitters, such as the cement industry, while mitigating the risk of potential CO_2 leakage over time associated with geological sequestration for larger emitters like coal power plants. Aqueous mineral carbonation can be classified into direct and indirect processes [11]. Direct aqueous carbonation is a one-step process in which the feedstock, water, and CO_2 react simultaneously in a single reactor. This approach simplifies the process but results in an integral blend of carbonated and unreacted inert solid residues [12]. Conversely, indirect aqueous carbonation is a two-step process that involves the initial extraction of desired metal ions (Ca^{2+}) from the solid waste, followed by the precipitation of pure carbonate (CaCO_3) from the Ca-rich leachate. Indirect aqueous carbonation is typically conducted in two separate reactors [4,13]. Fig. 1 presents schematic flow diagrams conceptualizing direct and indirect aqueous mineral carbonation processes.

While mineral carbonation has traditionally focused on naturally occurring silicate minerals such as olivine [14], serpentine [15], and wollastonite [16], recent research has highlighted the potential of industrial and construction waste as alternative feedstocks for carbonation due to their omnipresence [12,17,18]. Leventaki, et al. [19] conducted direct aqueous carbonation using industrial steel slags as a feedstock and deionized water as a leaching agent. CO_2 uptake in the range between 81 and 211 g- CO_2 /kg-slag was reported. Lee, et al. [20] investigated carbonation of granulated blast furnace slag via indirect aqueous carbonation. The highest CO_2 uptake of 130 g- CO_2 /kg-slag, was

obtained with an aqueous solution of ammonium salt, NH_4NO_3 . Ho, et al. [21] tested fly ash from coal-fired power in a direct aqueous carbonation reactor employing deionized water as a reaction medium. The maximum CO_2 uptake quantity was determined at 16 g- CO_2 /kg-fly ash. In a separate study, Ho, et al. [22] performed direct aqueous carbonation of concrete fines derived from the aggregate recycling process, using deionized water as a leaching agent. Measured CO_2 uptake was in the range between 40 and 120 g- CO_2 /kg-concrete fines. Indeed, the selection of suitable raw feedstocks is a critical factor in mineral carbonation. Feedstocks rich in alkali (Na) and alkaline earth metals (Mg, Ca) are ideal for mineral carbonation due to their high reactivity with CO_2 . Construction and demolition waste (C&DW) is a significant source of alkaline materials that can be repurposed for CO_2 sequestration [18].

The increasing volume of C&DW is driven by rapid urbanization, infrastructure expansion, and the frequent need for renovations and demolitions due to structural aging, design modifications, and economic development. C&DW derives from construction, renovation, and demolition activities, consists of various materials including concrete, wood, metals, bricks, glass, tiles, plaster, packaging, and excavated materials [23,24]. When recyclable materials such as metals, wood, glass, and tiles are removed, concrete can account for 40–50 % of the total C&DW material, making it the main component [25]. During the aggregate recycling process, fine C&DW particles are generated but are often discarded, as their presence can reduce the strength if reused in newly produced concrete. Currently, the primary disposal methods for C&DW fines include stacking [26,27] and landfills [28,29] but these techniques consume valuable land resources and contribute to environmental degradation. Therefore, finding effective ways to utilize fines from the C&DW recycling process is essential to mitigate the environmental burden derived from the cement and concrete industry. Concrete recycling can be used to reduce CO_2 emissions relative to the emissions associated with the mining and processing of virgin feedstock materials. Replacement of coarse aggregate with a 100 % recycled coarse aggregate reduced carbon emissions by $\sim 10\%$ (CO_2 savings 25–43 kg- CO_2 e/t-concrete) during the concrete life cycle [30]. To reduce concrete-derived CO_2 emissions further, C&DW fines are a good fit. C&DW fines could be suitable for aqueous mineral carbonation treatment due to their high Ca content (20–40 wt%) and alkalinity that enhances the solubility of CO_2 [22]. Along with that, size reduction

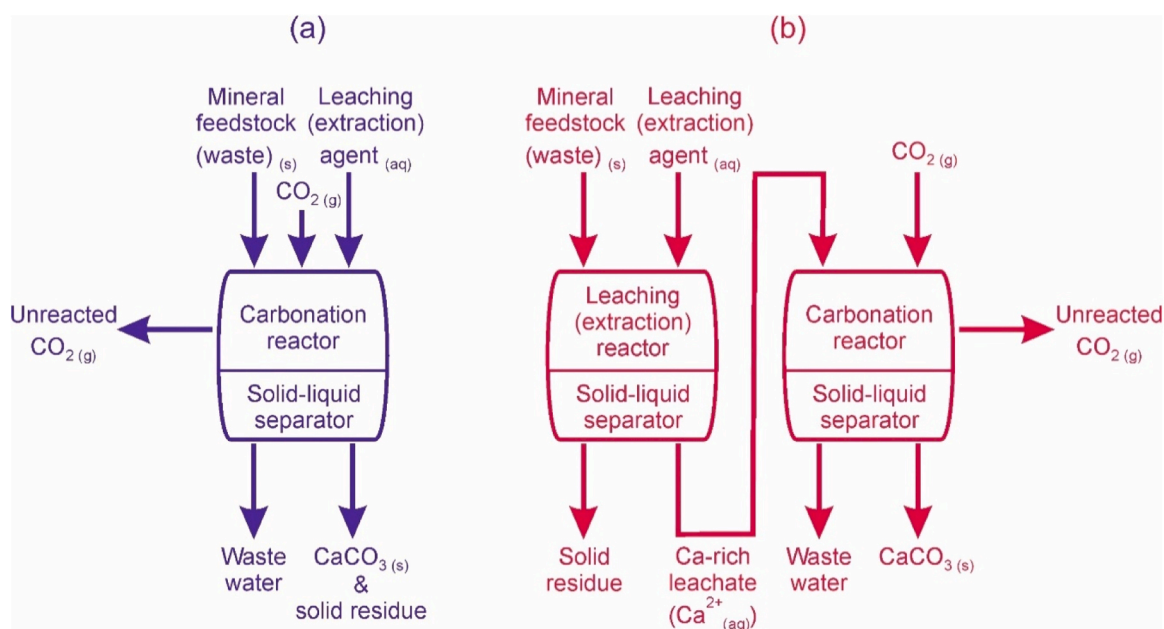


Fig. 1. Schematic flow diagrams depicting (a) direct and (b) indirect aqueous mineral carbonation processes.

(grinding and milling) is avoided, since this step is allocated to the aggregate recycling process [31].

Sorted C&DW (i.e., metals, wood, glass, and tiles are removed) contains crystalline phases of Ca-bearing compounds such as portlandite ($\text{Ca}(\text{OH})_2$), calcium silicate hydrates (C-S-H gel), and other phases that are highly reactive with CO_2 [32]. An opportunity exists for exploiting the potential of C&DW to react with CO_2 under aqueous conditions in both direct and indirect carbonation processes. The effectiveness of the carbonation process depends on several factors, such as reaction pH, temperature, carbonation reaction time, CO_2 pressure, CO_2 concentration, feedstock particle size, and the mineral composition of the waste material [22,33].

In South Africa, the cement and concrete industry is a significant source of CO_2 emissions, generating ~ 671 kg of CO_2 per ton of cement produced, which accounts for 1.7 % of the total emissions in the country [34]. It is estimated that construction and demolition activities in South Africa generate between 5 and 21 million metric tons of C&DW annually [35,36]. However, the lack of a centralized reporting system and inconsistencies in waste quantification complicate efforts to manage C&DW effectively. Given the large volume of C&DW generated, recent studies have emphasized the growing importance of carbon emission assessments and reduction strategies in the construction industry [37, 38], particularly during the materialization stage [39] and through the use of recycled materials [30,40]. These approaches underscore the need for integrating carbon management practices such as CO_2 sequestration into construction waste valorization pathways, supporting the relevance of this study on mineral carbonation of C&DW fines.

Several studies have explored the mineral carbonation of C&DW fines for CO_2 sequestration, with research focusing separately on direct [22,41,42] and indirect [43–45] carbonation processes. Despite these individual studies, comparative studies evaluating these two approaches when applied to C&DW fines remain limited in the literature. Comparing the two techniques, using the same Ca source can help identify the approach that best balances CO_2 sequestration efficiency, process feasibility, and product quality.

Acid mine drainage (AMD) is a highly acidic, metal-laden wastewater generated when sulfide minerals in mine waste react with water and oxygen. In South Africa, abandoned and active gold, coal, and metal mines are the major sources of AMD [46–48]. Approximately 202 million liters of AMD are discharged daily in the Johannesburg region alone [49]. To mitigate its environmental impact, AMD must be effectively managed and utilized [49–51]. Contemporary South African researchers have successfully demonstrated neutralization (sulphate removal) and removal of metallic contaminants present in AMD by precipitation of formed mineral phases. To achieve this, coal fly ash [48, 52] and concrete fines [51] were employed as neutralizers. However, there is currently no reported research on the use of AMD as a leaching agent in aqueous mineral carbonation process. Substituting BHW with AMD is of particular importance in a region like South Africa, where clean water resources are scarce, whereas polluted waste streams from mining activities are abundant. In this context, this study also conducted a comparative assessment of borehole water (BHW) and AMD as leaching agents for C&DW fines. In the recent review by Hanifa, et al. [53], other leaching agents effective in aqueous mineral carbonation process were identified. These are solutions of acids (HCl , HNO_3 , CH_3COOH) [4,53] and ammonium salts (NH_4NO_3 , NH_4Cl , $\text{CH}_3\text{COONH}_4$) [20,53].

The research gaps deduced from the literature review can be summarized in three points: (i) Quantitative data in terms of CO_2 uptake (i.e., CO_2 sequestration) in aqueous mineral carbonation processes utilizing concrete waste materials are scarce. (ii) The waste stream employed as a leaching agent has not been previously tested. (iii) A comparative assessment of direct and indirect aqueous mineral carbonation using C&DW fines as a Ca source has not been done in a single study. Thus, the research questions are: (a) “How do direct and indirect aqueous mineral carbonation processes compare in terms of CO_2 sequestration

effectiveness and product purity, when applied to C&DW fines?” and (b) “Can AMD be employed as an effective leaching agent in aqueous mineral carbonation process?”. To address the research questions, this study focuses on the following objectives: (1) Effectiveness of direct and indirect aqueous mineral carbonation for CO_2 sequestration using C&DW fines as a single feedstock. (2) Evaluate the efficiency of two different leaching agents (BHW and AMD) and examine the influence of varying solid-to-liquid (S/L) ratios on Ca extraction. (3) Quantifies the total CO_2 uptake during the carbonation process. The findings are evaluated with respect to existing literature to provide insights into the optimal carbonation pathway.

2. Materials and methods

Technical roadmap (Fig. 2) illustrates the key experimental stages in this study, and it outlines the systematic workflow from feedstock preparation to the characterization of carbonation products under both direct and indirect aqueous mineral carbonation pathways.

2.1. Materials

2.1.1. Construction and demolition waste (C&DW)

C&DW utilized in this study was sampled from a C&DW recycling facility located in the Western Cape province of South Africa. The C&DW was manually sorted to remove wood, metals, tiles, glass, and red clay bricks to increase the Ca content per unit mass. The sorted C&DW sample, used for experimentation, primarily consisted of concrete, with residual cement bricks and mortar waste.

2.1.2. Leaching agents

In this study, borehole water (BHW) and acid mine drainage (AMD) were tested to identify the most suitable leaching agent for Ca extraction from C&DW fines. BHW was sourced from the premises of the C&DW recycling facility, while the AMD was obtained from an abandoned coal mine in Mpumalanga province, South Africa. The high acidity of AMD makes it a promising candidate for enhancing Ca extraction efficiency, while simultaneously offering an effective approach to repurposing a major environmental pollutant as remediating agent.

2.2. Methods

2.2.1. Solid sample preparation and characterization

The sorted C&DW was milled using a rod mill, followed by sieving to obtain a fine fraction with a particle size of $< 125 \mu\text{m}$ [54,55], mimicking concrete fines typically derived from the aggregate recycling process. The sieved samples were oven-dried at 105°C overnight to remove residual moisture before characterization [56]. The drying process enhanced pore accessibility, prevented dilution effects during Ca extraction, and also facilitated deeper CO_2 penetration, ensuring the carbonation reaction was not limited to the surface of the particle [23, 33]. Identification of the crystalline phase was conducted using an X-ray diffractometer (XRD, Bruker D2 Phaser) equipped with a $\text{Cu K}\alpha$ radiation source (wavelength, $\lambda = 1.5406 \text{ \AA}$) and a scintillation detector. The particle size distribution (PSD) was analyzed using a Microtrac S3500 Laser Diffraction instrument equipped with a dry dispersion unit. Elemental composition analysis of the C&DW fines was performed by X-ray fluorescence (XRF) spectrometer (Panalytical Axios) fitted with a rhodium (Rh) end-window X-ray tube. Initial CaCO_3 content in the raw C&DW fines was determined through thermogravimetric analysis (TGA) [22,41] using a simultaneous thermal analyzer (NETZSCH STA 449 F5 Jupiter®). The surface area of the raw C&DW fines was measured with N_2 physisorption analysis on a Micromeritics Tristar II 3020, applying the Brunauer–Emmett–Teller (BET) method. Barrett–Joyner–Halenda (BJH) model was used to determine the average pore size distribution.

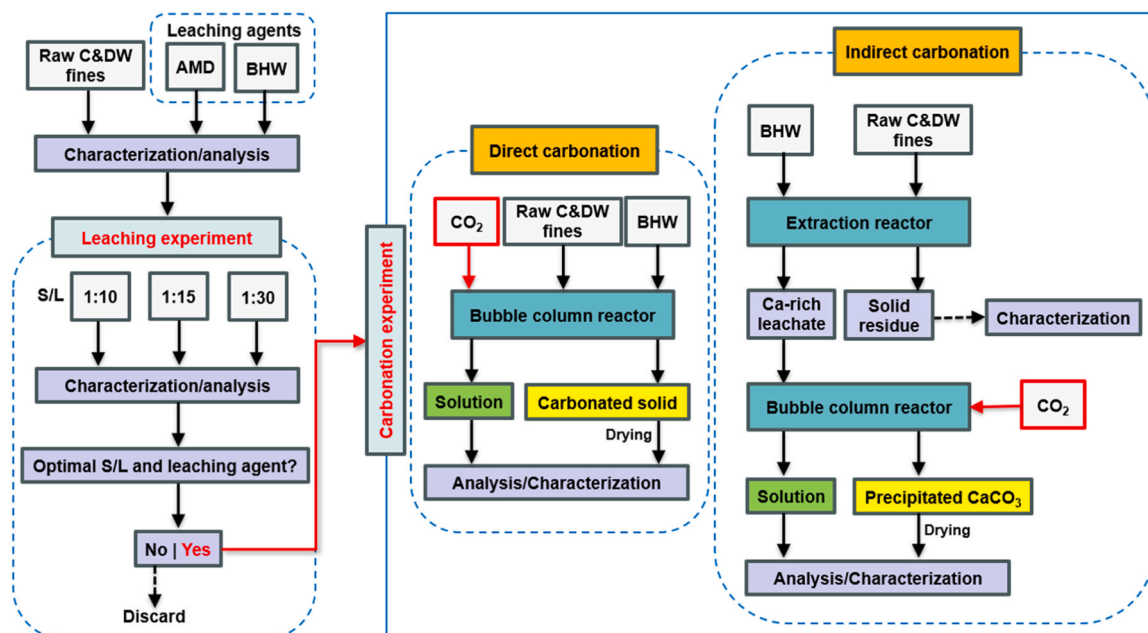


Fig. 2. Technical roadmap of this study.

2.2.2. Liquid sample preparation and analysis

Liquid samples drawn during leaching and/or carbonation were filtered through a 0.45 μm syringe filter before elemental and anion analysis. Samples designated for total elemental analysis were acidified with 3 drops of 3 mol/L HNO_3 and stored at 4 $^\circ\text{C}$ before analysis [56]. This was to ensure sample composition between extraction and analysis remained stable. Elements present in BHW and AMD samples were analyzed pre- and post-experimentation, using an inductively coupled plasma optical emission spectrometry (ICP-OES) instrument (SPECTRO ARCOS FHX3X). Anion (SO_4^{2-} and Cl^-) concentrations were determined using a discrete analyzer (Skalar BlueVisionTM).

2.2.3. Optimization of batch leaching experiments

Batch leaching experiments were conducted to identify the optimal solid-to-liquid (S/L) ratio for Ca extraction. Based on the literature [22, 57], three S/L ratios were investigated: 1:10, 1:15, and 1:30. The S/L ratio of 1:10 is widely used because, across relevant pH ranges, the concentration of dissolved metals is governed by their solubility limits rather than the total amount of solid present [58]. For each experiment, 600 mL of selected leaching agent (BHW or AMD) was transferred into a 1 L beaker. The amount of C&DW fines, corresponding to the designated S/L ratios, was added to the beaker containing the leaching agent. The suspension was continuously stirred at 400 rpm using an overhead stirrer to facilitate the extraction process. Batch leaching experiments were conducted over a 60-min period, with samples collected at the following time intervals: 0, 5, 20, 30, and 60 min. Each collected sample was filtered to separate the solid residue from the leachate. The pH of the leachate for the time-dependent batch leaching process was monitored using a pH meter (pH 50 VioLab).

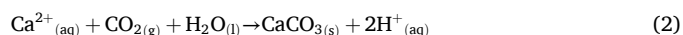
2.2.4. Carbonation experiments

Aqueous carbonation experiments were designed and conducted in a 3 L bubble column reactor designed by Ho, et al. [22]. Typically, the reactor comprised an acrylic column, a glass filter (i.e., perforated plate), and a Teflon tube. The glass filter, with an aperture size of 5–10 μm , was positioned at the base of the column to ensure homogeneous CO_2 dispersion. A Teflon tube was connected at the top of the reactor to take samples. A continuous gas analyzer (EL3020, Uras 26, ABB, Germany) was calibrated by a two-point calibration curve in a range of 0–32 vol% CO_2 . Calibration was carried out by 99.9 N_2 and 32 vol% CO_2

before the commencement of the carbonation experiments. The gas mixture (i.e., CO_2 and N_2) was controlled using a mass flow controller (KOFLOC 8500MC), while the gas analyzer monitored the CO_2 concentration in real-time. To simulate the 20 vol% CO_2 exhaust gas (typically emitted during cement production), the experimental gas mixture was prepared by setting the mass flow control of 99.5 vol% CO_2 to 0.8 L/min and that of 99.9 N_2 to 3.2 L/min, resulting in a total gas flow rate of 4 L/min.

2.3. Indirect aqueous carbonation

Indirect aqueous carbonation is a two-stage process that involves the sequential extraction of Ca from Ca-bearing minerals into solution, followed by its reaction with dissolved CO_2 to form CaCO_3 . The overall reaction mechanisms governing the extraction and carbonation processes are given in Eqs. (1) and (2), respectively:



The indirect aqueous carbonation experiments were conducted as follows. Firstly, Ca extraction from C&DW fines was optimized. The optimal S/L ratio and the most effective leaching agent was determined according to the procedure defined in Section 2.2.3. In the indirect aqueous carbonation experiments, 200 g of C&DW fines was mixed with 2 L of BHW (S/L ratio of 1:10). Following the 60-min extraction stage, the resulting Ca-rich leachate was separated from the solids by sedimentation, and the liquid extract transferred into a bubble column reactor for carbonation. The simulated gas mixture containing 20 vol% CO_2 was continuously bubbled through the reactor for 60 min. Throughout the carbonation process stage, pH and CO_2 concentrations were monitored at predetermined time intervals. The reaction product, which consisted of precipitated CaCO_3 suspended in the slurry, was filtered and oven-dried at 105 $^\circ\text{C}$ overnight. Both the liquid and the dried CaCO_3 were further characterized for their chemical properties.

2.4. Direct aqueous carbonation

Direct aqueous carbonation is a process in which three interdependent stages: Ca extraction, CO_2 dissolution, and CaCO_3 precipitation,

occur simultaneously in a single reactor. The overall reaction mechanism governing this process can be summarised by Eq. (3).



The direct aqueous carbonation was performed as follows: A volume of 2 L of BHW was transferred into the column reactor. The simulated gas mixture containing 20 vol% CO₂ was continuously bubbled through the reactor at a flow rate of 4 L/min until CO₂ saturation equilibrium was reached. The saturation point was confirmed when the CO₂ concentration at the reactor inlet matched the CO₂ concentration at the reactor outlet. Upon achieving the CO₂ saturation point, 200 g of C&DW fines was quickly transferred into the reactor, giving a S/L ratio of 1:10. Carbonation proceeded for 60 min, during which pH and CO₂ concentrations were measured at specific time intervals. At the end of the experiment, the reaction slurry was subjected to filtration to separate the solid product from the carbonated water. The solid product was subsequently oven-dried. The solid product from direct carbonation consisted of a newly precipitated CaCO₃ and C&DW fines. Both the liquid fraction and solid product were further characterized for their chemical properties.

2.5. Calculations

The initial CaCO₃ content in the raw C&DW fines was determined via TGA. Thermal decomposition of CaCO₃ is associated with a distinct weight loss primarily within the 650–850 °C range [22]. Observation from the TGA curve was used to calculate the CaCO₃ content according to Eq. (4):

$$W_{\text{CaCO}_3} (\text{weight } \%) = W_{\text{CO}_2} \times (M_{\text{CaCO}_3} / M_{\text{CO}_2}) \quad (4)$$

where W_{CaCO_3} represents the weight % of CaCO₃ in the raw C&DW fines; W_{CO_2} is the weight loss % of CO₂ between 650 and 850 °C in TGA curve; and M_{CaCO_3} and M_{CO_2} are the molecular weights of CaCO₃ (= 100.09 g/mol) and CO₂ (= 44.01 g/mol), respectively. The amount of CaCO₃ in the raw C&DW fines was subsequently used to determine the Ca content present as CaCO₃ ($W_{\text{Ca,reacted}}$) in the raw sample. This fraction of Ca is considered unavailable for aqueous carbonation because it is already carbonated. The effective Ca content available for carbonation reaction was determined based on Eq. (5):

$$W_{\text{Ca,effective}} (\text{weight } \%) = W_{\text{Ca,total}} - W_{\text{Ca,reacted}} \\ = [(W_{\text{CaO}} \times (M_{\text{Ca}} / M_{\text{CaO}}))] - (W_{\text{CaCO}_3} \times (M_{\text{Ca}} / M_{\text{CaCO}_3})) \quad (5)$$

where $W_{\text{Ca,effective}}$, $W_{\text{Ca,total}}$ and $W_{\text{Ca,reacted}}$ symbolize the weight % of Ca in the raw C&DW fines available for carbonation, total Ca content in the sample, and already carbonated/reacted Ca (present as CaCO₃) within the sample, respectively. M_{CaO} and M_{Ca} are the molecular weights of CaO (=56.08 g/mol) and Ca (=40.08 g/mol), respectively. It is important to note that both $W_{\text{Ca,effective}}$ and $W_{\text{Ca,reacted}}$ are time-dependent, due to the continuous progression of the carbonation process throughout the service life of concrete.

Data recorded by the gas analyzer during the carbonation reaction were used to determine the CO₂ uptake. The ideal gas law was applied to calculate the total quantity of CO₂ absorbed from the gas phase following Eq. (6):

$$\text{Weight (g) of CO}_2 \text{ uptake} = \frac{(\Delta V_{\text{CO}_2} \times P)}{RT} \times M_{\text{CO}_2} \quad (6)$$

where ΔV_{CO_2} is the volume of CO₂ uptake (L), $P = 1$ atm, $R = 0.0821$ L.atm/mol.K, $T = 293$ K, and $M_{\text{CO}_2} = 44.01$ g/mol. The volume of CO₂ uptake was calculated as shown in Eq. (7):

$$\Delta V_{\text{CO}_2} (\text{L}) = \int_0^t F_{\text{total}} \times \frac{(C_{\text{CO}_2,i} - C_{\text{CO}_2,t})}{100} dt \quad (7)$$

where F_{total} is the total flow rate (L/min); while $C_{\text{CO}_2,i}$ and $C_{\text{CO}_2,t}$ denote

the initial and time-dependent CO₂ concentrations (%), respectively, as measured by the continuous gas analyzer. The CO₂ absorptive rate is utilized to determine the volume of CO₂ dissolved in the reaction slurry. The volume of dissolved CO₂ is equal to the integration area under the CO₂ uptake curve. Note that the weight/volume of CO₂ uptake as denoted by Eqs. (6) and (7) consist of the amount of CO₂ captured in solid CaCO₃ and the amount of CO₂ remaining dissolved in water.

To calculate the CO₂ captured in CaCO₃ in the indirect carbonation, the weight of dry solid precipitate (W_{CaCO_3}) was the input in Eq. (8):

$$\text{Weight (g) of CO}_2 \text{ captured in CaCO}_3 \text{ (indirect)} = W_{\text{CaCO}_3} \times \frac{M_{\text{CO}_2}}{M_{\text{CaCO}_3}} \quad (8)$$

To quantify the CO₂ captured in CaCO₃ in the direct carbonation, it is necessary to determine the CaCO₃ content in the carbonated solid product. As mentioned previously, the CO₂ data obtained from the TGA curve can be used to determine the CaCO₃ content according to Eq. (4). Thus, the amount of CO₂ captured in CaCO₃ can be calculated according to Eq. (9):

$$\text{Weight (g) of CO}_2 \text{ captured in CaCO}_3 \text{ (direct)} \\ = \frac{W_{\text{CaCO}_3,f} - W_{\text{CaCO}_3,i}}{100} \times \frac{M_{\text{CO}_2}}{M_{\text{CaCO}_3}} \quad (9)$$

where $W_{\text{CaCO}_3,i}$ is the initial amount of CaCO₃ in the raw C&DW fines and $W_{\text{CaCO}_3,f}$ is the final amount of CaCO₃ in the solid product after carbonation.

3. Results and discussion

3.1. Analysis of leaching agents: BHW and AMD

Analytical results of the leaching agents (BHW and AMD) used for the batch Ca extraction are shown in Table 1. BHW has a higher pH (7.4) than highly acidic AMD (pH 2.6). AMD has significantly higher levels of Ca, Mg, Fe, and Al. The low pH of AMD suggests that Ca extraction from the raw C&DW fines could primarily occur during the neutralization process, where the acidic starting conditions enhance the dissolution of Ca-bearing phases. Additionally, compared to BHW, AMD is characterized by elevated concentrations of Cl[−] and SO₄^{2−}, due to its origin from strong acid-generating reactions in geochemical processes [50]. The chemical composition of the AMD used in this study is consistent with those reported in the literature [59,60].

3.2. Characterization of raw C&DW fines

3.2.1. XRF analysis

XRF analysis of raw C&DW fines is presented in Table 2. SiO₂ is the

Table 1
Elemental and ionic composition of leaching agents (BHW and AMD).

Analyte	Concentration (mg/L)	
	BHW	AMD
Ca	63.1	430
K	4.7	46.7
Mg	3.3	436
Na	9.7	56.1
Si	7.6	50.1
Al	N.D.	430
Fe	N.D.	3201
Mn	N.D.	88.4
Pb	N.D.	0.23
Zn	N.D.	8.1
Cl [−]	20.5	110
SO ₄ ^{2−}	19.1	3561

N.D. – not detected

Table 2

XRF characterization (oxide composition) of raw C&DW fines.

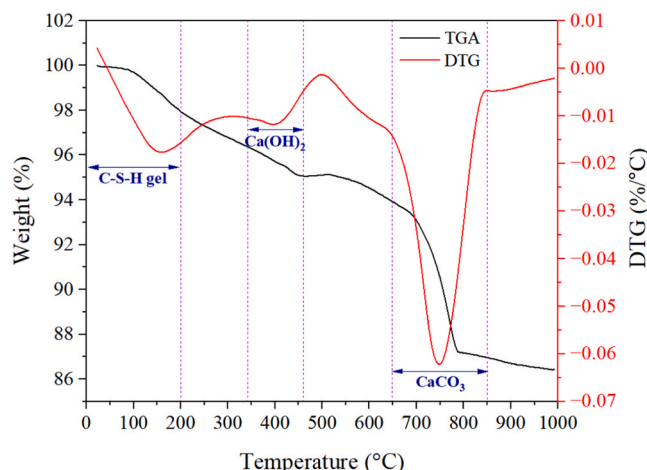
Analyte	Concentration (wt%)*
SiO ₂	46.4
Al ₂ O ₃	4.28
Fe ₂ O ₃	1.97
MnO	0.04
MgO	0.68
CaO	23.5
Na ₂ O	0.33
K ₂ O	0.59
SO ₃	0.56
LOI	14.4
H ₂ O	6.16

* Values expressed on as-received basis

major component with a value of 46.4 wt%, while the content of CaO, the key component for carbonation reaction, is 23.5 wt%. The value of CaO is within the range for building demolition waste reported in the literature: 11.19–16.86 wt% [61], 22–34.9 wt% [58], 26.1 wt% [62], and 27.5 wt% [63]. Variations in CaO content can be attributed to factors such as material source, crushing method, concrete service life, and exposure to climatic conditions [58]. The volatile matter content was estimated by subtracting the weight percent value of H₂O from the loss on ignition (LOI) value obtained from the XRF. The volatile matter content (8.24 wt%) suggests that a portion of the Ca in the raw C&DW fines has already reacted with atmospheric CO₂, forming CaCO₃, which decomposes upon heating. Also, residual water from the concrete curing process and other hydrated phases (Ca(OH)₂, C-S-H gel) likely contributed to the observed weight loss during ignition.

3.2.2. TGA analysis

The TGA and derivative TGA (DTG) analysis (Fig. 3) revealed that the raw C&DW fines released 7.93 wt% CO₂, corresponding to a CaCO₃ content of 18.03 wt% calculated by Eq. (4). This value is lower than the 22.9–44.2 wt% of CaCO₃ in raw concrete fines, reported by Ho, et al. [22]. This suggests that in this study, less of the Ca present in C&DW had naturally reacted with CO₂. As a result, a greater proportion of reactive Ca (i.e., effective Ca) species remained available for further carbonation. Typically, the effective Ca comprises Ca(OH)₂ and C-S-H gel, which are both components of hydrated cement. Comparison between the volatile matter content (8.24 wt%) and the CO₂ loss (7.93 wt%) suggests that CO₂ release accounts for the majority of the volatile matter in the raw C&DW fines. However, a small fraction of other volatiles, such as structural water from hydrated cementitious phases, may also contribute to the remaining weight loss.

**Fig. 3.** TGA and DTG of raw C&DW fines.

The DTG curve inflection between 650 and 850 °C is associated with CO₂ released due to CaCO₃ thermal decomposition. Dehydroxylation of portlandite (Ca(OH)₂) occurred between 340 and 460 °C wt%. The section from room temperature to 250 °C is indicative of water loss deriving from surface water and structural water embedded in hydrated cementitious components, such as ettringite and C-S-H gel [45,64]. The combined mass loss due to moisture release and dehydroxylation corresponds closely to the H₂O content value of 6.16 wt% obtained by XRF. Decomposition of CaCO₃ occurs between 650 and 850 °C, accounts for ~8 wt% mass loss, representing about half of the total LOI value of 14.4 wt% obtained from XRF.

Table 3 presents the total Ca, reacted Ca, and effective Ca content in the raw C&DW fines (Eq. 5). It is estimated that ~57.25 wt% of the total Ca in the raw C&DW fines can be theoretically carbonated. The effective Ca (9.67 wt%) obtained in this study is lower than the value (12.4–21 wt%) reported by Ho, et al. [22]. This observation may be attributed to variations in the mineral composition and degree of hydration of the raw C&DW fines, which can affect Ca availability for carbonation.

3.2.3. XRD analysis

Phase identification of the raw C&DW fines that are depicted in Fig. 4, revealed the presence of minerals such as portlandite (Ca(OH)₂), ettringite (Ca₆Al₂(SO₄)₃(OH)₁₂·26 H₂O), albite (NaAlSi₃O₈), illite ((KH₃O)(Al,Mg,Fe)₂(Si,Al)₄O₁₀·[(OH)₂(H₂O)]), quartz (SiO₂), and calcite (CaCO₃). Peaks corresponding to quartz and albite observed in the raw C&DW fines indicate that quartz sand and granite were likely the natural aggregates used in the parent concrete. Notably, no distinct diffraction peak corresponding to C-S-H gel was observed, which is consistent with its largely amorphous nature and poor crystallinity [45]. However, the presence of CaCO₃ peaks suggests that some degree of carbonation may have occurred over time. The identified mineral phases are typical of aged C&DW and align well with findings from existing literature [22,65]. However, the presence of unreacted Ca(OH)₂ plays a key role in the carbonation process, as it readily reacts with CO₂, facilitating the formation of CaCO₃.

3.2.4. PSD and BET/BJH analysis

The evolution of Ca extraction into solution and subsequent carbonation of the Ca-rich leachate is closely related to the particle size distribution (PSD) in the raw C&DW fines. The PSD (Fig. 5) indicates that the volumetric median particle size, D50 value of 68.13 μm, presents a relatively fine fraction with sufficient surface area available for reaction, which enhances the dissolution of Ca-bearing phases. The predominant fraction in the fine particle range enhances nucleation sites for CaCO₃ precipitation and prevents agglomeration in bubbling suspension systems, thus improving mass transfer and reaction kinetics during carbonation. In crushed materials, finer particles frequently occlude within the interstices and pore spaces between larger particles, a characteristic that reduces reactive surface area in mineral carbonation [66].

The raw C&DW fines had a pore volume of 0.038 cm³/g and a BET surface area of 22.7 m²/g, which is notably higher than the values reported for similar C&DW materials, 11.09 m²/g [67] and 9.12 m²/g [68]. This implies that in this study, more sites are available for leaching agents to access and dissolve Ca-bearing minerals. The physisorption isotherm and hysteresis loop provide insights into the pore structure and interactive nature of a material. As shown in Fig. 6a, the N₂

Table 3

Total Ca, reacted Ca, and effective Ca content in the raw C&DW fines.

Content	Weight (%)
W _{Ca total}	16.89
W _{Ca reacted}	7.22
W _{Ca effective}	9.67

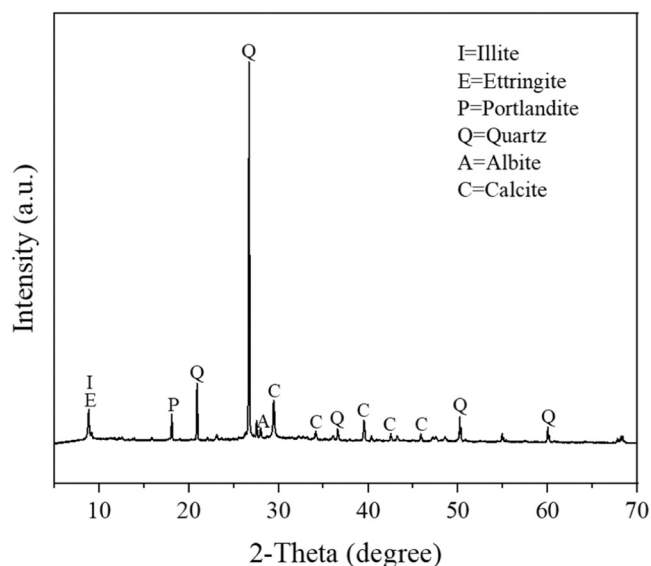


Fig. 4. XRD pattern of raw C&DW fines.

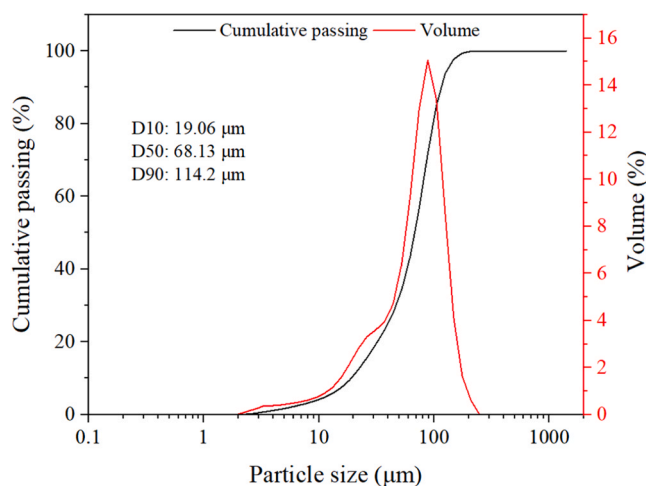


Fig. 5. Particle size distribution (PSD) in raw C&DW fines.

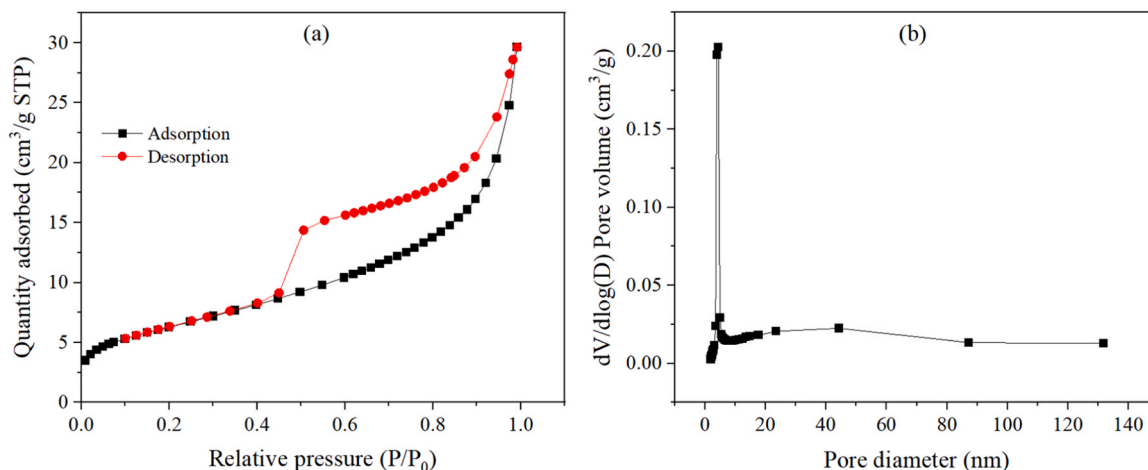


Fig. 6. (a) Physiosorption isotherm and (b) BJH pore-size distribution in raw C&DW fines.

physiosorption isotherm of the raw C&DW fines exhibits a Type IV shape and an H3 hysteresis loop, which is typical of mesoporous materials [69]. This indicates that capillary condensation occurs within the pore network, confirming the mesoporous nature of the fines. During leaching, this pore structure can enhance Ca extraction process by retaining leaching agents within the pores, thereby prolonging leaching or reaction time. The pore size distribution, depicted in Fig. 6b, further supports this observation. The measured average pore size of 7.8 nm falls within the mesoporous range (2–50 nm) [69], which is known to promote both leaching and carbonation reactions.

3.3. Influence of leaching agent and S/L ratios on pH evolution and Ca extraction

The evolution of suspension pH during Ca extraction from C&DW fines using BHW and AMD under different S/L ratios is illustrated in Figs. 7a and 7b, respectively. The results indicate that leaching with BHW leads to a rapid increase in pH, reaching values well above the recommended pH range (9–10) for aqueous mineral carbonation [70]. The rapid equilibration observed within the first 10 min across all BHW experiments indicates that Ca extraction occurs swiftly irrespective of the different S/L ratios applied, due to the near-neutral initial pH (7.4) and relatively low buffering capacity of BHW. This implies that a high S/L ratio is not beneficial for BHW. In contrast, AMD leaching exhibits a different pH evolution. The pH increases gradually, reaching pH stabilization within 30 min. This observation can be attributed to the initial acidity of AMD (pH 2.6) and its high sulfate and metal concentrations, which likely consume alkalinity generated during the leaching of C&DW fines. The buffering effect of AMD suggests that significant alkalinity is required before the system overcomes the acidic conditions and reaches pH levels suitable for CaCO_3 precipitation. Nonetheless, this was dependent upon the S/L ratio applied. Madzivire, et al. [48] thoroughly studied the neutralization of AMD by coal fly ash. The buffering effect in the pH range 4–7 was attributed to hydrolysis induced by Fe^{3+} and Al^{3+} ions, followed by the precipitation of Fe and Al hydroxides. These findings suggest that BHW promotes more effective Ca extraction than AMD across all applied S/L ratios, as evidenced by the higher pH values and greater calcium availability in solution. This enhanced Ca release has important implications for the subsequent carbonation stage, where dissolved Ca serves as the key reactant for CO_2 mineralization.

Since leaching with AMD at an S/L ratio of 1:10 resulted in a final pH of 11.6, which exceeds the recommended range of 9–10 [70], its performance was compared with all tested ratios using BHW, as summarized in Table 4. Notably, all BHW leaching conditions yielded a final pH of ~12.8, indicating BHW as a favorable leaching agent for Ca extraction and subsequent carbonate formation. Subtraction of the values from

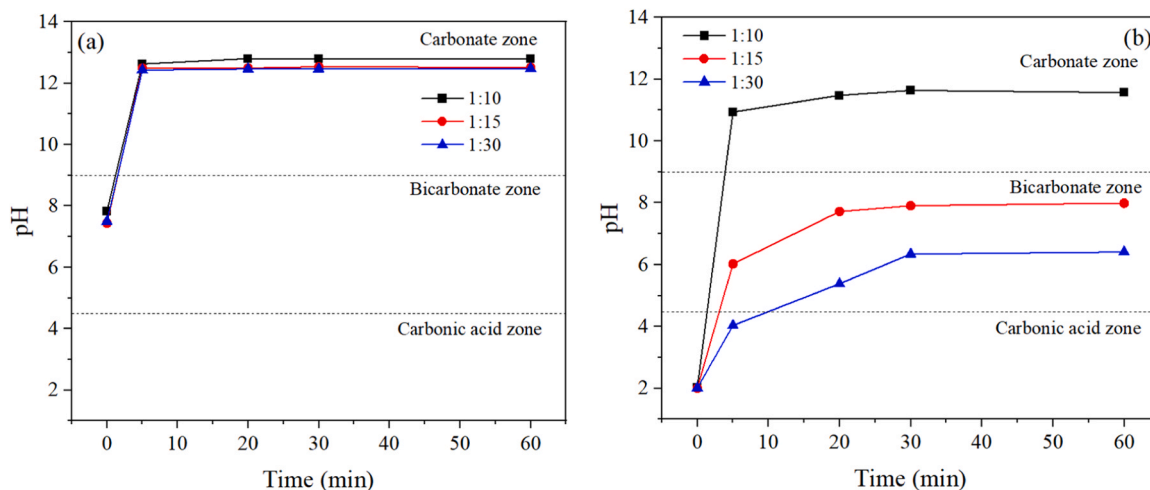


Fig. 7. pH evolution during leaching under different S/L ratios. Leaching agent (a) BHW and (b) AMD.

Table 4

Elemental and ionic composition of BHW and AMD leachate after 60 min leaching of C&DW.

Analyte	BHW (mg/L)			AMD (mg/L)
	1:10	1:15	1:30	1:10
Ca	391	378	211	515
K	19.7	14	9.9	35.4
Mg	N.D.	N.D.	N.D.	66
Na	31.7	26.3	17.8	84
Si	1.1	N.D.	1.8	1.1
Al	N.D.	0.08	0.59	0.7
Cl ⁻	42.7	31.2	26	59
SO ₄ ²⁻	18.3	19.2	27.3	2459

N.D. – not detected

Tables 4 and 1 (i.e., 391 minus 63.1 mg/L) reveals that using BHW as a leaching agent at S/L ratio of 1:10 resulted in the highest Ca extraction (~328 mg-Ca/L-BHW) from the C&DW fines, outperforming other ratios. Subtraction of the values referring to AMD (i.e., 515 minus 430 mg/L) under the same S/L ratio resulted in a leached concentration of 85 mg-Ca/L-AMD. Although Table 4 shows that the apparent Ca content in AMD is higher compared to BHW, this is due to the higher initial content of Ca in AMD (Table 1). During leaching, the quantities of SO₄²⁻ in AMD diminished (from 3561 to 2459 mg/L). As the pH of AMD

gradually increases into the alkaline zone, the SO₄²⁻ is removed by precipitation of gypsum (CaSO₄·2H₂O) and Fe and Al oxyhydroxysulphates [48]. In particular SO₄²⁻ in AMD, may compete with CO₃²⁻ for available Ca precipitating gypsum instead of calcite. To conclude, this section highlights the superior effectiveness of BHW in promoting Ca extraction.

3.4. XRD analysis of C&DW residue after leaching

Figs. 8a and 8b show the XRD diffractograms of the solid C&DW residue after leaching with BHW and AMD, together with the raw C&DW fines. Notably, the ettringite, portlandite, and illite phases present in the raw C&DW fines disappeared following leaching with either BHW or AMD at all the S/L ratios investigated. This disappearance aligns with the higher pH values observed in Figs. 7a and 7b, indicating the effective dissolution of Ca-bearing phases, which makes them available for carbonation. This observation corroborates the findings reported by Ho, et al. [22]. After leaching with AMD (Fig. 8b), new peaks corresponding to gypsum and starkeyite (MgSO₄·4H₂O) emerged, resulting from reactions between dissolved Ca²⁺ and Mg²⁺ with SO₄²⁻, respectively. The peak of gypsum was more pronounced at a S/L ratio of 1:30 compared to 1:15 and 1:10, suggesting that increased dilution altered the chemical environment, particularly pH and ion mobility, in a manner that favoured the formation or stabilization of the sulfate mineral. While the Ca

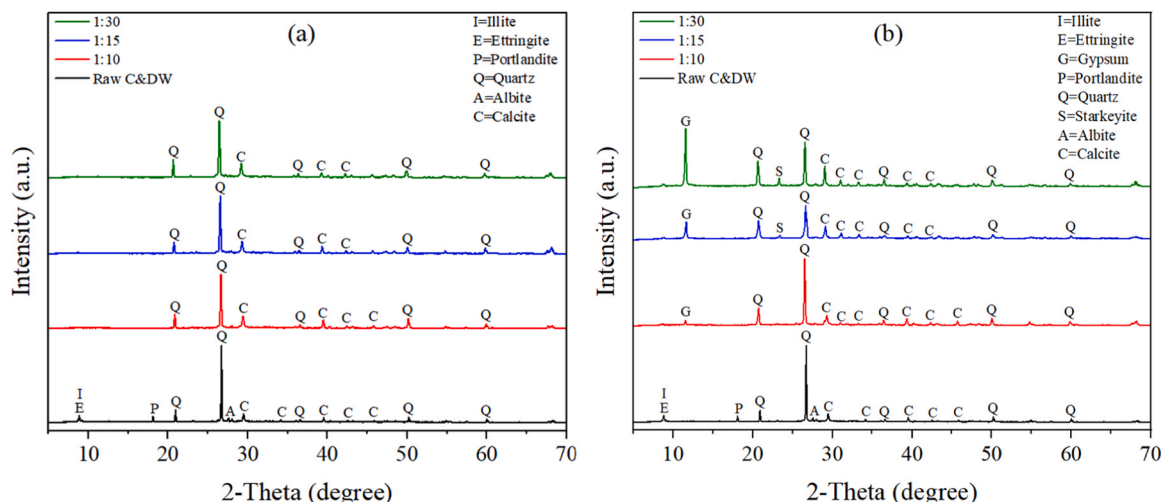


Fig. 8. XRD patterns of the solid C&DW residue after leaching with (a) BHW and (b) AMD at the different S/L ratios, and the pattern of the raw C&DW fines.

in C&DW promoted the removal of sulphates from AMD via gypsum formation, this precipitation simultaneously reduced the availability of dissolved Ca for subsequent carbonation reactions. Based on these findings, BHW at S/L ratio of 1:10 was selected for further carbonation experiments.

3.5. Direct and indirect aqueous carbonation reaction

Fig. 9 illustrates the variation in pH and CO₂ uptake during both direct and indirect carbonation experiments employing BHW leaching agent. It should be noted that the leaching agent, AMD, has not been tested for carbonation reaction due to its poor extraction of Ca from C&DW fines. As shown in Fig. 9a, the indirect carbonation process begins with a high initial pH within the carbonate zone. As CO₂ is introduced to the Ca-rich supernatant, the pH rapidly declines below 8 (bicarbonate zone), resulting in the formation of a mixture of calcium bicarbonate (Ca(HCO₃)₂) and calcium carbonate (CaCO₃), before stabilizing just above pH 6. This decline is driven by CO₂ dissolution and conversion to bicarbonate, which promotes the continued presence of calcium bicarbonate in solution.

The initial rapid uptake of CO₂ corresponds to the precipitation of CaCO₃, which continues until the pH drops below 9, where carbonate precipitation ceases. Below this pH, the continuous introduction of CO₂ may promote the formation of bicarbonate species in solution, depending on the equilibrium between CO₂ dissolution and Ca availability in solution. The available Ca in the leachate is the rate-limiting step, directly influencing the extent of CO₂ dissolution in the leachate and subsequent carbonation (i.e., CO₂ sequestration). In the study of Madzivire, et al. [59], aluminium hydroxide was used to buffer the Ca-rich supernatant derived from leaching fly ash, maintaining its pH in the carbonate zone to enhance CaCO₃ precipitation. The direct carbonation follows a different pH evolution pattern. Initially, the solution maintains a near-neutral pH due to the equilibrium between CO₂ and Ca present in the fresh BHW. Upon introducing raw C&DW fines, the pH rises to 11.67 as alkaline components are not buffered. At this high pH, CO₃²⁻ dominate, and conditions are favorable for CaCO₃ precipitation rather than Ca(HCO₃)₂ formation. Unlike indirect carbonation, where Ca is readily available for reaction, direct carbonation involves simultaneous Ca extraction and CO₂ dissolution, leading to a slower, gradual decline in pH over time. After 10 min, the pH reached the bicarbonate zone thereafter, suggesting that CO₃²⁻ species remained dissolved in solution and were prevented from immediately forming solid carbonate precipitates.

Fig. 9b presents the CO₂ uptake rate, highlighting distinct CO₂ uptake trends for both carbonation pathways. It is important to note that

Fig. 9b measures the CO₂ exiting the system and thus encompasses the amount of CO₂ captured in solid CaCO₃ and the amount of CO₂ remaining dissolved in water. Ghacham, et al. [71] and Ho, et al. [22] also acknowledged that a notable portion of CO₂ remains dissolved in water and unavailable for carbonate formation. During indirect carbonation, CO₂ dissolution occurs rapidly within the first ~5 min, and CO₂ uptake is driven by the immediate reaction with available Ca, but is terminated by the pH decreasing to below the carbonate zone. Conversely, direct carbonation exhibits a prolonged CO₂ uptake phase, as the pH remains in the carbonate zone for ~10 min, due to the concurrent Ca extraction, CO₂ dissolution, and carbonation reaction, which maintains favorable pH conditions for CaCO₃ precipitation. The slower CO₂ uptake in direct carbonation suggests that the rate of Ca extraction from C&DW fines governs the overall reaction rate, maintaining the favorable pH for at least 10 min, which is finally limited by the lack of immediately available reactive Ca species. The reaction pH continues to decrease after 10 min until 30 min. In this period, CO₂ keeps on dissolving, which is indicated by the tailing in Fig. 9b. CO₂ forms bicarbonate ions until the suspension system gets saturated with CO₂. Once equilibrium is established between CO₂ uptake and Ca release, both pH and CO₂ uptake stabilize, indicating that further carbonation is constrained by both the unavailability of dissolved Ca and the unfavorable pH regime in the bicarbonate zone.

3.6. CO₂ accounting in the suspension system

Given that CO₂ was continuously supplied at a flow rate of 0.8 L/min for 60 min, the total CO₂ volume introduced into the reactor amounted to 48 L per experiment. Considering the density relation, the weight of CO₂ supplied to the suspension system was 95.0 g for both carbonation approaches. During the indirect carbonation experiments, a total of 10.5 g-CO₂/kg-C&DW fines was taken up by the suspension system, whereas the direct carbonation process resulted in a significantly higher CO₂ uptake of 52.8 g-CO₂/kg-C&DW fines. Table 5 allocates captured CO₂ between solid CaCO₃ and the amount remaining dissolved in water.

Table 5
Allocation of captured CO₂ during direct and indirect carbonation reaction.

Carbonation experiment	Total CO ₂ uptake by suspension system (g-CO ₂ /kg-C&DW fines)	CO ₂ captured in solid phase as CaCO ₃ (g-CO ₂ /kg-C&DW fines)	CO ₂ remaining dissolved in water (g-CO ₂ /kg-C&DW fines)
Direct	52.8	39.9	12.9
Indirect	10.5	0.95	9.2

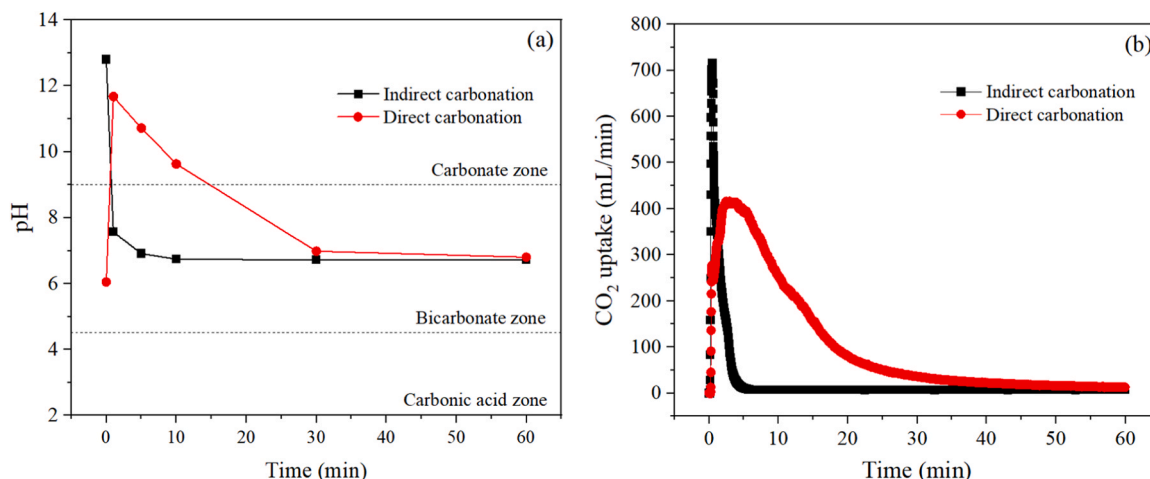


Fig. 9. (a) pH variation and (b) CO₂ uptake rate during direct and indirect carbonation experiments employing BHW leaching agent at S/L of 1:10.

In the direct carbonation experiment, 84.5 g of CO₂ (out of a total of 95.0 g of CO₂ input) exits the reactor as unreacted gas. Notably, from the 52.8 g-CO₂/kg-C&DW fines of total CO₂ uptake, 39.9 g-CO₂/kg-C&DW fines was captured in the solid phase as CaCO₃, while 12.9 g-CO₂/kg-C&DW remained dissolved in water. CO₂ that remained dissolved in water existed in two forms: either as a carbonate ion (CO₃²⁻) or as bicarbonate ion (HCO₃⁻). In contrast, indirect carbonation exhibited lower CO₂ uptake. Out of the 10.5 g-CO₂/kg-C&DW fines of total CO₂ taken up by the suspension system, only 0.95 g-CO₂/kg-C&DW fines was captured as a solid carbonate, while 9.2 g-CO₂/kg-C&DW fines persisted in solution. These results highlight the substantial disparity in CO₂ sequestration efficiency between direct and indirect carbonation and the large role that pH declines have in the process efficiency. The superior CO₂ uptake in direct carbonation suggests that simultaneous Ca extraction and carbonation enhance the reaction kinetics and overall sequestration efficiency. To increase CO₂ uptake in the indirect carbonation process, it is essential to maintain an alkaline pH (carbonate zone). Zhang and Moment [43] and Lee, et al. [20] achieved that by the addition of NaOH solution into the precipitation reactor.

3.7. Comparative analysis of CO₂ uptake with published studies

Table 6 provides a summary of CO₂ uptake yields reported in previous studies for both direct and indirect aqueous carbonation of concrete waste materials. The CO₂ uptake from direct carbonation in this study was 52.8 g-CO₂/kg-C&DW fines, close to the result of Ghacham, et al. [71] (42–78 g-CO₂/kg-waste concrete) but lower than the values reported by Ho, et al. [22] and Pasquier, et al. [72]. This reduced uptake is mainly due to the low effective Ca content in C&DW (9.67 wt%), which limits CaCO₃ formation. Although the studied C&DW fines had a fine particle size (<125 µm), which typically enhances reactivity by increasing surface area, the limited availability of reactive Ca phases remained the primary bottleneck. In contrast, the indirect carbonation route achieved a much lower CO₂ uptake of 10.5 g-CO₂/kg-C&DW fines. This result cannot be related to literature data. Up to date CO₂ uptake through indirect carbonation route has not been quantified (Table 6). The direct carbonation process in this study sequestered nearly five times more CO₂ than the indirect route under the conditions applied. This superior performance is attributed to the direct exposure of reactive phases to CO₂, bypassing the need for prior Ca extraction and a separate precipitation step that characterizes indirect carbonation. The indirect method, although theoretically more controlled, is more sensitive to kinetic and solubility constraints, especially when dealing with low-Ca and partially carbonated waste materials. Future improvements should focus on enhancing reactive Ca content and optimizing process

parameters, especially for indirect systems, as there are limited studies available on this.

3.8. Analysis and application of carbonation solid product

The carbonation solid products obtained after 60 min of reaction time from both direct and indirect carbonation processes were analyzed and compared with high-purity CaCO₃ (Analytical Grade, 95.5 %). As depicted in Fig. 10a, the crystalline phase of the precipitate formed during indirect carbonation was primarily composed of calcite, confirming the successful formation of CaCO₃. In contrast, the product from direct carbonation exhibited a mixed crystalline composition, predominantly consisting of quartz and calcite. TGA (Fig. 10b) was employed to quantify the weight % of CaCO₃ in the carbonated solid product. Leveraging on the differences in thermal stability of CaCO₃ and other components in the product, the weight % of CaCO₃ in the product was also taken as the percentage purity of CaCO₃. This was determined based on the CO₂ weight loss observed from TGA, using the relationship provided in Eq. (4). The results indicate ~87 % purity of the carbonated product from indirect carbonation, comparable to analytical-grade CaCO₃. Comparing TGA curves with those presented by Zhang and Moment [43] implies that indirect carbonation produces high-purity CaCO₃. This high-purity product is suitable for use in several applications, such as a functional filler in polymer, plastic [73], and paper [74] industries due to its rheological and visual properties. Relative to the cement and concrete industry, Yang, et al. [75] tested the addition of nano-sized CaCO₃ to 3D printing cementitious materials. The addition of 1 wt% nano CaCO₃ as a binder material accelerated the hydration of Portland cement and improved mechanical properties such as yield stress and green strength. Zhao and Zou [76] investigated flue gas desulfurization employing micro- and nano-CaCO₃ fluids. The authors achieved absorption capacity between 47.7 and 54.4 mg-SO₂/mL-fluid while nano CaCO₃ fluids demonstrated favorable rheology properties. Moreover, the review published by Jin, et al. [77] identified research gaps that need to be addressed to reduce CO₂ emissions via large-scale utilization of calcium carbonate products derived from alkaline wastes. Direct carbonation product had a significantly lower purity of ~27 % due to the integral blend of produced carbonates together with silicates and other minerals present in the C&DW fines. The direct carbonation product can be used as a backfilling material in construction operations [12], supplementary cementitious materials or aggregates in concretes [78], component in sub-base layers for road construction, or as a stabilizing agent in geotechnical applications [79,80], particularly in areas where mechanical strength is more critical than chemical purity.

Although a detailed quantitative economic assessment is beyond the current scope of this study, we recognize the importance of evaluating

Table 6

Literature overview of the key features and research gaps associated with direct and indirect aqueous carbonation of various concrete waste materials.

Carbonation experiment	Feedstock	Particle size (µm)	Effective Ca (wt%)	Leaching agent used	CO ₂ uptake (g-CO ₂ /kg-feedstock)	Reference
Direct	Synthesized concrete fines	212	36.4	Water	190	Ho, et al. [41]
Direct	Concrete fines	16.8–96.2	21	Water	40–120	Ho, et al. [22]
Direct	Waste concrete	< 500	-	Water	42–78	Ghacham, et al. [71]
Direct	Waste concrete	< 500	-	Water	110	Pasquier, et al. [72]
Direct	Concrete fines	< 2360	-	Water	Only qualitative analysis of carbonation products	Jiang, et al. [42]
Direct	Construction and demolition waste fines	< 125	9.67	Borehole water	52.8	This study
Indirect	Recycled concrete aggregate fines	< 4000	-	Ammonium nitrate	Only leaching tests	Gasós, et al. [44]
Indirect	Waste concrete	< 425	13.4	HCl, NaOH	Only qualitative analysis of carbonation products	Vanderzee and Zeman [45]
Indirect	Construction and demolition waste fines	50–350	-	HCl	Only leaching tests and qualitative analysis of carbonation products	Zhang and Moment [43]
Indirect	Construction and demolition waste fines	< 125	9.67	Borehole water	10.5	This study

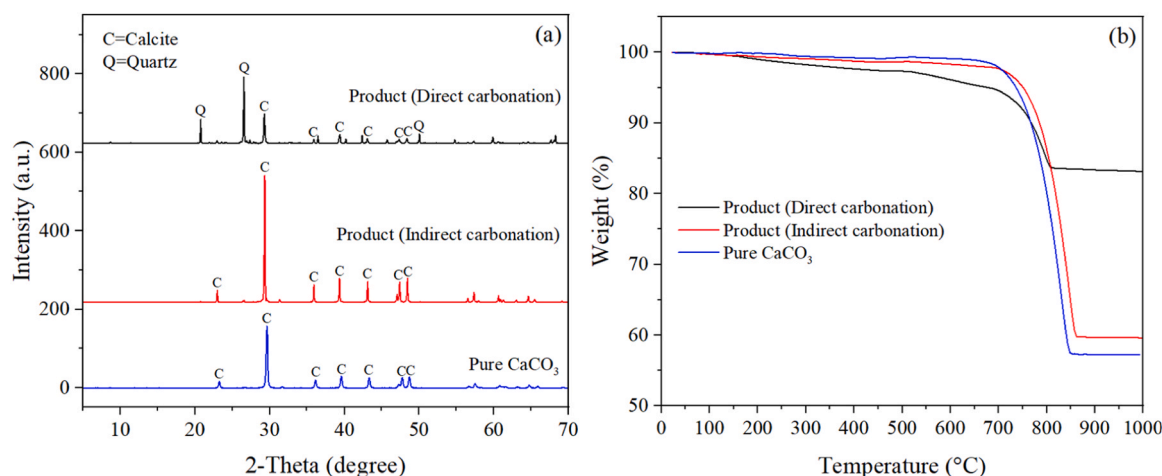


Fig. 10. Direct and indirect carbonation solid product (a) XRD pattern and (b) TG curve.

the economic implications to support the practical implementation of these technologies. From a qualitative perspective, direct aqueous mineral carbonation tends to offer cost advantages due to its relatively simpler process configuration, lower chemical and energy input requirements, as demonstrated in this study. Along with that, an unambiguous carbon-negative balance of the process would accelerate the market uptake. On the other hand, indirect carbonation generally involves multiple steps and the use of chemical reagents such as acids and bases for extraction and precipitation. These added steps can increase both capital and operational expenditures. Nevertheless, the potential for by-product recovery and more efficient CO₂ utilization could offer economic benefits in specific contexts. The selection between the two approaches should therefore consider the trade-offs between process complexity, conversion efficiency, reagent costs, and potential for material valorization.

4. Conclusion

This study investigated two aqueous mineral carbonation methods using construction and demolition waste (C&DW) fines as a calcium (Ca) source. The effectiveness of borehole water (BHW) and acid mine drainage (AMD) as leaching agents was assessed, along with the influence of solid-to-liquid (S/L) ratios of 1:10, 1:15, and 1:30. For the water-scarce regions like South Africa, the utilization of abundant but polluting waste stream such as AMD is of particular importance. The results demonstrated that:

- BHW exhibited superior Ca leachability compared to AMD, with the optimal Ca extraction occurring at an S/L of 1:10. Under the same conditions, BHW leached ~328 mg-Ca/L-BHW, while AMD leached 85 mg-Ca/L-AMD. High initial acidity, sulfate, and metal concentrations in AMD consume the alkalinity required for CaCO₃ precipitation. Along with that, sulfates and metals in AMD compete for the reactants in the carbonation reaction.
- Direct aqueous carbonation showed greater potential for CO₂ sequestration. Continuous leaching of Ca from the C&DW fines maintained favorable pH conditions for CaCO₃ precipitation. Direct aqueous carbonation achieved a significantly higher CO₂ uptake of 52.8 g-CO₂/kg-C&DW fines compared to 10.5 g-CO₂/kg-C&DW fines for indirect aqueous carbonation.
- The solid product from indirect aqueous carbonation was primarily composed of ~87 % pure calcite (i.e., CaCO₃), whereas the solid product from direct aqueous carbonation predominantly consisted of quartz and calcite (~27 % purity).

- These findings highlight C&DW as a promising feedstock for Ca-based aqueous mineral carbonation and support the adoption of direct aqueous carbonation as a more effective CO₂ sequestration strategy over indirect aqueous carbonation.
- To improve the effectiveness of CO₂ capture in indirect aqueous carbonation, further studies, including pH control, have to be conducted.
- Leaving out the energy consumption in the process is a limitation of this study. Future work should quantify the energy consumption and present the ratio of CO₂ capture versus CO₂ emissions associated with the carbonation process.

CRediT Authorship Contribution Statement

K. Ramasenya, B. Oladipo, V. N. Katambwe: Investigation, Methodology, Data curation, Formal analysis, Validation, Writing – Original draft. S. Gcasamba, A. Horvat: Visualization, Writing – Review & editing. V. R. K. Vadapalli, J. Petersen, H. Coetzee, H.-J. Ho, A. Iizuka, L. F. Petrik, T. V. Ojumu: Conceptualization, Funding acquisition, Project administration, Software, Resources, Supervision, Visualization, Writing – Review & editing.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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