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Saeed, M., Stedt, A., Scheer, J. et al (2026). Extraction of $\text{Ca}(\text{OH})_2$ from alkaline industrial wastes for use as absorbent of CO_2 from air in negative emission technologies (NETs). Heliyon, 12(12).
<http://dx.doi.org/10.1016/j.heliyon.2026.e45140>

N.B. When citing this work, cite the original published paper.



Research article

Extraction of $\text{Ca}(\text{OH})_2$ from alkaline industrial wastes for use as absorbent of CO_2 from air in negative emission technologies (NETs)

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ABSTRACT

Calcium hydroxide could play a crucial role in absorbing CO_2 from the atmosphere through various active and passive direct air capture systems, helping achieve negative emissions. Additionally, $\text{Ca}(\text{OH})_2$ can be directly applied to soil and water for enhanced weathering-based CO_2 removal. Significant amounts of calcium exist in industrial by-products and wastes, such as ashes, slags, and mine tailings—materials often landfilled or discarded. However, these materials often contain heavy metals that pose environmental risks due to potential leaching into the biosphere. Efficient extraction of calcium from such wastes would not only provide a useful resource but also reduce hazardous residues requiring disposal. Once extracted, calcium can be precipitated as $\text{Ca}(\text{OH})_2$ through pH adjustment. Despite its potential, research on this process remains limited, as most studies have focused on direct mineral carbonation using concentrated CO_2 streams.

This study investigates calcium extraction and $\text{Ca}(\text{OH})_2$ precipitation from three industrial by-products: steel slag, iron-sponge process waste, and mine tailings. Calcium was leached using either HCl or NH_4Cl , followed by two-stage pH adjustment with NH_4OH and NaOH to separate impurities and precipitate $\text{Ca}(\text{OH})_2$, respectively. ICP-OES and XRD analysis showed that NH_4Cl exhibited higher extraction selectivity, achieving 90–100% $\text{Ca}(\text{OH})_2$ precipitation. In contrast, HCl extracted more calcium but had lower precipitation efficiency (71–83%) due to co-dissolution of impurities, causing some Ca-bearing species to precipitate with the heavy metals during the first precipitation stage. Although HCl produced more slaked lime (wet $\text{Ca}(\text{OH})_2$), NH_4Cl demonstrated strong calcium selectivity and minimal impurity leaching, making it competitive for treating mine tailings and iron sponge slag. Although residues from leaching iron-sponge slag with HCl are safe for disposal, smaller and more manageable solid residues from the intermediate stages may provide a practical alternative for treating harmful elements like sulfur, vanadium, and chromium, as they are not entirely leached in most cases.

1. Introduction

Many nations aim to achieve net-zero carbon dioxide emissions by 2050 to limit global temperature rise to 1.5°C. Negative emission technologies (NETs) that offset hard to abate residual emissions play a key role in achieving this goal [1]. Afforestation, bioenergy with carbon capture and storage (BECCS), direct air capture (DAC), enhanced weathering and direct ocean capture are a few examples of negative emission technologies. Compared to BECCS, DAC does not rely on biomass, a limited resource whose large-scale use can lead to emissions from land-use change and biodiversity loss [2,3]. However, DAC is currently estimated to cost between \$600 and \$1000 for every tonne of CO_2 captured [4]. More affordable and somewhat primitive NETs, such as passive DAC, enhanced weathering, and

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direct ocean capture, could offer solutions at a wider scale and lower cost [5]. A key component in these systems is the use of sorbents like $\text{Ca}(\text{OH})_2$ for capturing CO_2 . Aqueous solution of $\text{Ca}(\text{OH})_2$ has been recognized as an effective solvent for capturing CO_2 from the air and serve as a crucial intermediate in engineering solutions for capturing CO_2 ambiently from air [6–9]. Although calcium hydroxide can be obtained from natural limestone, there are environmental issues associated with increased outtake of limestone in many regions of the world and producing it without CO_2 emissions would entail the use of a carbon capture system [10]. To be able to make a true global impact on current CO_2 emissions by Ca-based CCS processes, we should find sufficiently large industrial waste streams and base our circular and sustainable material value chain on those. For example, given the substantial amounts of calcium in alkaline industrial waste, extracting it with an acidic solvent and then precipitating $\text{Ca}(\text{OH})_2$ could offer environmental, resource, and climate benefits. Additionally, over time, these waste streams can generate acidic leachates that release harmful metals, such as vanadium and chromium, into the environment. This creates environmental hazards, and the high cost of waste storage underscores the need for effective treatment to prevent contamination, safeguard public health, and promote resource recycling [11–13].

In this paper, we will analyze a novel method for extracting Ca from waste-based materials using solvents such as HCl and NH_4Cl and producing $\text{Ca}(\text{OH})_2$ which can be used in cost effective negative emission technologies (NETs), as shown in Fig. 1. The process also aims at simultaneously treating industrial alkaline waste for hazardous elements that present risks to land, water bodies and natural habitats.

1.1. Carbonation of slaked lime

Many of the suggested processes for utilization of Ca as a sorbent raw material for carbonation utilize solid $\text{Ca}(\text{OH})_2$, which reacts effectively with CO_2 in the air. For instance, in the passive DAC process, gaseous CO_2 in the atmosphere dissolves in the water layer formed around the particles due to moisture, forming H_2CO_3 , and subsequently calcium hydroxide from the surface of the sorbent also dissolves. Here, $\text{Ca}(\text{OH})_2$ dissociates into Ca^{2+} and OH^- ions in solution, while H_2CO_3 dissociates into H^+ , HCO_3^- , and CO_3^{2-} . Finally, the calcium and carbonate ions react to form calcium carbonate [14].

Enhanced weathering involves a similar process, but the natural process of chemical weathering is accelerated by finely grinding rocks and spreading them over large areas of land [15]. Carbonate mineralization-induced removal of CO_2 from ocean water involves three separate processes: aqueous dissolution of CO_2 , calcium leaching, and precipitation of CaCO_3 through the reaction between calcium and bicarbonate ions. Overall CO_2 solubility in ocean water rises because of increased precipitation of CaCO_3 [16–18].

1.2. Extraction of Ca and treatment of industrial waste

The feasibility of extracting calcium from industrial byproducts for CO_2 sequestration through carbonation has been widely investigated. The core concept involves dissolving calcium ions in an aqueous solution, and if a concentrated CO_2 source—such as emissions from fuel combustion—is available, it can be directly utilized to precipitate CaCO_3 in situ from the solution [19–22]. However, a significant limitation of this process is the requirement for a highly concentrated CO_2 stream. While industries such as power generation, steelmaking, and cement production often offer suitable point sources for post-combustion carbon capture, the locations of the metal oxide sources and CO_2 emissions are not always aligned, potentially necessitating additional transport infrastructure. In this paper, the idea is to precipitate out calcium hydroxide for later ex-situ carbonation to be more independent from a high concentrated CO_2 stream.

With respect to the first Ca-extraction step, with the main goal of dissolving Ca-ions from the solid material, the efficiency and selectivity for Ca are crucial for the process to maximize its economic value [23]. According to Said et al., several factors, including particle size, temperature, solvent choice, solvent concentration, solid to liquid ratio (SLR), and agitation, are important in this process [24]. Many extraction agents have been used in literature at different concentrations. However, the most frequently used are acetic acid (CH_3COOH) [21,22,25], ammonium chloride (NH_4Cl) and other ammonium salts [26], and hydrochloric acid [22,23,27]. It is

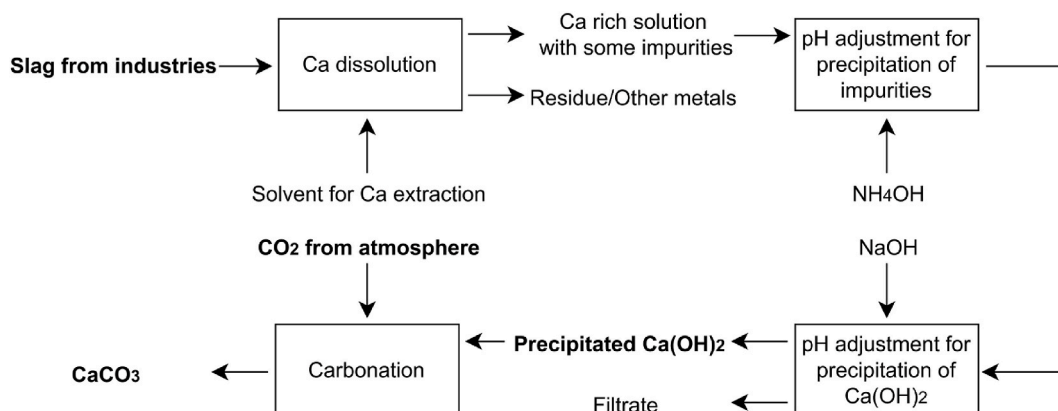
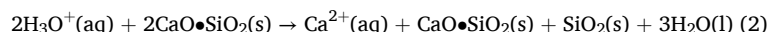
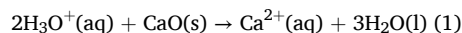


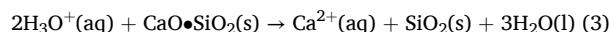
Fig. 1. Flowchart representing the process of producing $\text{Ca}(\text{OH})_2$ from alkaline by-products for use in affordable NETs.

claimed that the widely used solvent NH_4Cl is extremely selective in terms of calcium extraction, with values up to 99.6% [22]. HCl is an extractant that, at normal temperature, also extracts other impurities including iron and silica [27], but Lee et al. preferred it because of its high calcium extraction efficiency though less than NH_4Cl [23].

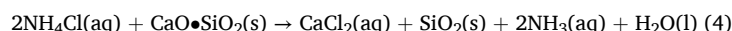
The Ca-extraction from alkaline waste materials, applying such solvents, only occurs for certain calcium bearing phases. In the case of steelmaking slags, Mattila et al. mentioned that the dissolution of calcium occurs according to the following reactions [20]:



One may determine the stoichiometric amount of slag required to be fully dissolved at a given solvent concentration using reactions 1 and 2 and the known Ca content of the slag material. However, Owais et al. was able to obtain a larger yield from these reactions than anticipated, therefore he added a third reaction, 3, which dissolves calcium from calcium silicate when H_3O^+ is in access [28]:



Reactions 4 and 5 below can be used to determine the stoichiometric amount of extracted calcium given that 1 mol of, say, NH_4Cl or HCl yields 1 mol of H_3O^+ .



Another parameter affecting the extraction process is the particle size. The surface area of the solid increases and the diffusion path length shortens by decreasing the particle size, hence improving the extraction efficiency. Additionally, the calcium content in finer particles has been found to be greater which would be beneficial for the extraction process [20]. For this reason, most studies in the literature have been found to employ particle sizes below 250 μm .

Acid leaching extracts calcium as well as removes harmful metals like vanadium, chromium, and sulfur from the waste, allowing safe disposal without costly post-processing [11]. HCl is effective for metals in oxide or sulfide forms but may not leach metals in complex minerals [22,23,27,29]. NH_4Cl , though less efficient than HCl, can leach calcium and other metals forming soluble ammonium complexes [21,22,25]. This would make NH_4Cl easier to regenerate, similar to the regeneration of CH_3COOH [30–32]. In future renewable-based power systems, the cost of electricity could be as low as zero, making electrolysis a feasible option to regenerate almost any solvent [33].

1.3. Precipitation of $\text{Ca}(\text{OH})_2$

There has been very little focus on the extraction of Ca to be precipitated as $\text{Ca}(\text{OH})_2$ using the pH swing method, with most studies focusing on direct precipitation of CaCO_3 (PCC) [34]. This means, however, that a concentrated CO_2 stream is available, which may not always be the case. In this paper, the $\text{Ca}(\text{OH})_2$ precipitation is investigated which is the crucial component that connects the atmospheric CO_2 capture technologies to the efficient calcium dissolution from alkaline by-products.

In every study that has been done on this subject, pH regulation has been employed to achieve precipitation of $\text{Ca}(\text{OH})_2$ as solid phase. Um et al. demonstrated that calcium hydroxide must precipitate out from CaCl_2 in NaOH - H_2O solutions at a high alkaline condition. A pH of 12.5 was necessary for the precipitate to form at 25°C–90°C using NaOH as a control variable [35].

Lee et al. used 2M HCl for Ca extraction followed by the precipitation of $\text{Ca}(\text{OH})_2$ at pH 13 before the impurities like Fe, Al, and Mg hydroxides were removed at pH 9.5 with NH_4OH , achieving a 99.9% Ca yield and pure portlandite. $\text{Ca}(\text{OH})_2$ precipitates out from the solution using NaOH as per Reaction 6.



1.4. Aim and scope

In this paper, a process for calcium extraction and $\text{Ca}(\text{OH})_2$ precipitation while simultaneously treating industrial alkaline waste has been studied by applying two different extraction agents/solvents (HCl and NH_4Cl) using three different byproducts from the mining and steelmaking industry. The validity of the process is investigated by a comprehensive quantitative assessment of Ca leaching and $\text{Ca}(\text{OH})_2$ precipitation behavior between the investigated byproducts and solvents, applying the selected methods and varying certain parameters. It should be emphasized that there have been a significant number of studies on direct mineral carbonation for capturing CO_2 from highly concentrated CO_2 streams, but very little research has been done with respect to producing $\text{Ca}(\text{OH})_2$ from alkaline wastes and low CO_2 concentrations. A qualitative evaluation is also conducted to examine the phase changes and the leaching of heavy metals following extraction with solvents. Considering the significant need for cheap and environmental materials for negative emission technologies, and the enormous amounts of alkaline waste products produced yearly, this study could provide insights on how these by-products can be utilized in an effective way for maximum climate benefit.

2. Methodology

Three very different but widely available alkaline waste materials have been used in this paper. These materials have a higher concentration of calcium-bearing phases. The suggested experimental approach is therefore enhanced for extracting Ca and precipitating $\text{Ca}(\text{OH})_2$.

2.1. Materials and sample preparation

In this paper, three different industrial byproducts have been used:

1. Steel converter slag, also known as Linz-Donawitz slag (LD-Slag), is a by-product of the steelmaking process that results from the Linz-Donawitz process, which is used to cure pig iron before it enters the Basic Oxygen Furnace (BOF) [36]. The material was provided by SSAB Merox (Sweden).
2. Petrit T, a byproduct from iron sponge process, resembles slag generated in a blast furnace. It was supplied by Höganäs AB, a Swedish business that makes metal powders for the powder metallurgy sector. Iron and aluminum are present in large amounts in addition to calcium and silicon [37].
3. One of the biggest zinc mines in the world, Tara Mining, which is run by the new Boliden AB in Ireland, produced the third industrial by-product tested in this work. Due to the limestone ore body, the underground Tara Mine generates a sizable volume of mine tailings with a high calcium and silicon content [38].

These materials were received in a fine powder form with a large weight fraction between a particle size of 90 and 250 μm . For the extraction tests, a particle size distribution of 90 – 250 μm is chosen to guarantee a large surface area and to assure that a considerable portion of the industrial byproduct could be utilized. Particle sizes as small as 90 μm have been used in earlier research, but investigations using particles as large as 250 μm have also shown successful extraction characteristics [20].

2.2. Experimental setup

The experimental process consists of three stages. Stage 1 involves performing the Ca-extraction from the solid material in an acidic solvent. The precipitation of impurities (stage 2) and the final precipitation of portlandite (step 3) would subsequently be made possible by a pH swing approach.

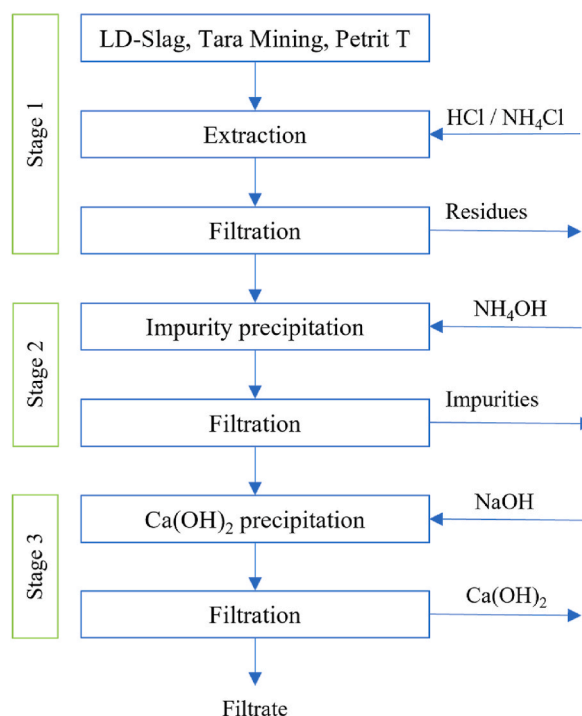


Fig. 2. Flow diagram of the experimental method used for each industrial alkaline byproduct.

2.2.1. Stage 1: Calcium extraction

Two well-known calcium extraction agents have been used in this work, i.e., ammonium-chloride (NH_4Cl) and hydrochloric acid (HCl). The rationale for using HCl is that it is known to remove calcium more effectively than NH_4Cl . Although it is anticipated that NH_4Cl would extract less calcium at the same concentration, it has been found that the solvent has a higher selectivity for Ca meaning that it mostly only dissolves the Ca bearing phases. Also, in terms of regeneration, it has been found that NH_4Cl is easier to regenerate [32]. The two solvents have been employed at the same concentration of 2 mol/L for better comparison.

It is also necessary to establish the extraction time and solid to liquid ratio, SLR. This study uses an SLR of 100 g/L, which is within the range used in numerous previous investigations [23]. The extraction period has been set at 20 min, which is rather short compared to other experiments but is thought to be enough given the choice of particle sizes ($<250\ \mu\text{m}$). The entire procedure was carried out at ambient temperature and pressure.

A batch reactor with an 800-rpm magnetic stirrer is used to mix 100 ml of the solvent and 10 g of alkaline byproducts of appropriate size for 20 min. Five time-steps have been used to monitor the pH and temperature values: $t = 0, 1, 5, 10$, and 20 min. After extraction, the sludge is filtered using a vacuum pump through a $2\text{--}3\ \mu\text{m}$ filter. Although 100 ml of 2M HCl is used for Ca extraction from LD-slag, only 60 ml could be obtained after filtration due to filter clogging leading to wastage of some filtrate. Since the cause of the clogging was unknown, the experiment was repeated. No clogging or filtrate wastage was observed the second time and similar ion compositions and efficiencies were obtained as the first, hence the data from the earlier experiment was deemed suitable for further use.

50 ml of the collected filtrate is sent to stage 2 while the remaining filtrate is sent for ICP-OES analysis to determine the ion composition. The filter cake residue is dried at 120°C for 6 h. The dried residue is then subjected to an XRD analysis as well as an elemental analysis using ICP-SFMS. An overview of the three steps, including the Ca extraction, impurity precipitation, and precipitation of the final product $\text{Ca}(\text{OH})_2$, is shown in Fig. 2.

2.2.2. Stage 2: Precipitation of impurities

The filtrate from stage 1 is mixed with a weak base (2M NH_4OH) in stage 2 with the goal of raising the solution's pH to 9.5, which has been found to be ideal for precipitating less soluble species such as $\text{Fe}(\text{OH})_2$, $\text{Fe}(\text{OH})_3$, $\text{Al}(\text{OH})_3$ and $\text{Mg}(\text{OH})_2$ [23]. A $2\text{--}3\ \mu\text{m}$ vacuum filter is used again to filter the solution after it reached the required pH level. Any residue found is dried for 6 h at 120°C . 50 ml of the filtrate is sent to stage 3 for processing while the remaining filtrate is saved for ICP-OES analysis.

2.2.3. Stage 3: Selective precipitation of $\text{Ca}(\text{OH})_2$

To precipitate $\text{Ca}(\text{OH})_2$, a strong base (1M NaOH) is applied in the last step to achieve a pH of 13. The process for filtration and drying is the same as in the earlier steps. In addition to the amount of base added to the filtrate, the temperature, pH, and the amount of precipitated $\text{Ca}(\text{OH})_2$ are also measured. The filtrate from stage 3 is again sent for ICP-OES analysis.

2.3. Analysis techniques

2.3.1. XRD analysis

The original samples, residual filter cakes from stage 1 and the final $\text{Ca}(\text{OH})_2$ precipitates are analyzed using Powder-X-Ray Diffraction (XRD). The instrument used in this work is the D8 Discover Bruker. As an X-Ray source it uses a copper source ($\lambda = 1.5406\ \text{nm}$) in the setup of 40 V/40 mA. The detector (Eiger2) is used in 2D mode with an angle coverage of $10\text{--}80^\circ$, screening at an angle step size of 0.02 (two theta degrees, 2θ).

2.3.2. ICP-OES – ion composition

The assessment of the ion composition in the filtrate is done using Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES). The method allows us to analyze aqueous samples by exciting the ions in plasma. The ions of interest in this study are calcium, aluminum, magnesium, silicon, iron, manganese, vanadium, and chromium. As diluents, 0.5M HNO_3 and 0.5M HCl (spiked with Yttrium) are used.

2.3.3. ICP-SFMS

In this study, Inductively Coupled Plasma – Sector Field Mass Spectroscopy (ICP-SFMS) is used to determine the elemental composition of the solid byproducts. Because of its high sensitivity, ICP-SFMS offers the lowest detection limits of any ICP-MS, making it the ideal option for identifying trace and ultra-trace elements. A wet digestion method using $\text{HNO}_3/\text{HCl}/\text{HF}$ or LiBO_2 fusion method is applied prior to the analysis, depending on which material is being digested and what element is being analyzed.

2.4. Data evaluation

The processing of the ICP-OES data to provide comparable findings is described in the following sections.

2.4.1. Extraction efficiency

The amount of Ca dissolved by the acid in stage 1 ($m_{\text{Ca},1}$) divided by the total amount of Ca present in the alkaline byproduct ($m_{\text{Ca},\text{total}}$) gives the Ca extraction efficiency (Equation (1)).

$$\eta_E = m_{\text{Ca},1} / m_{\text{Ca},\text{total}} \quad (1)$$

2.4.2. Dilution factor (D_f)

In stages 2 and 3 of the experimental method, bases (NH_4OH and NaOH) are used to raise the pH of the filtrate to 9.5 and 13, respectively, to segregate the contaminants and precipitate $\text{Ca}(\text{OH})_2$. The two solvents, NH_4Cl and HCl , which are mild and strong acidic solutions, respectively have different pH after extraction and require different amounts of bases for stage 2 and stage 3. This causes non-similar dilutions of the solvent that have an impact on the ICP-OES measured ion concentrations. To rectify the results and allow for comparison, a dilution factor must be included.

This factor, given in Equation (2), expresses the ratio between the final volume after adding the base (V_{final}) and the volume of filtrate sent to the individual stage from the previous one ($V_{\text{previous stage}}$).

$$D_f = V_{\text{final}} / V_{\text{previous stage}} \quad (2)$$

The relevant D_f is then multiplied by the ion-concentration values determined by ICP-OES for stages 2 or 3 to make them equivalent to the undiluted ion concentration after stage 1. All concentrations reported herein have been corrected using Equation (2).

2.4.3. $\text{Ca}(\text{OH})_2$ precipitate

The amount of $\text{Ca}(\text{OH})_2$ precipitate can also be theoretically calculated using the stage 2 and stage 3 ion concentrations from the ICP-OES (mg/L) and the volume of the first filtrate (Equation (3)), which was collected following the 20-min batch reaction in step 1. Here, the dilution factor (D_f) has been used to adjust the concentration and is thus not explicitly shown in the equation. The mass of calcium ions is converted to the mass of calcium-hydroxide ions using a factor of 1.85. It represents the ratio of two substances' respective molar weights (calcium: 40.08 g/mol; calcium-hydroxide: 74.08 g/mol).

$$m_{\text{Ca}(\text{OH})_2} = (C_{\text{Ca, stage 2}} - C_{\text{Ca, stage 3}}) \times V_{\text{filtrate, stage 1}} \times 1.85 \quad (3)$$

Here it is assumed that there is no precipitation of $\text{Ca}(\text{OH})_2$ during stage 2.

2.4.4. Precipitation efficiency

The ratio of Ca in the precipitated portlandite at the end of stage 3 to the total dissolved calcium after stage 1 can be used to measure the Ca precipitation performance (Equation (4)).

$$\eta_p = (C_{\text{Ca, stage 2}} - C_{\text{Ca, stage 3}}) / C_{\text{Ca, stage 1}} \quad (4)$$

The amount of Ca in portlandite that precipitated upon adding 1M NaOH is represented by the difference in normalized concentrations of Ca between stage 2 and stage 3 according to ICP-OES measurements. The number of dissolved calcium ions at the beginning of stage 1 is compared to this difference to determine the precipitation efficiency of Ca.

2.4.5. Total conversion efficiency

The ratio of Ca in the final precipitated slaked lime to the total amount of calcium present in the industrial byproduct used in stage 1 gives the total Ca conversion efficiency. This can be calculated as a product of extraction and precipitation efficiencies as given in Equation (5).

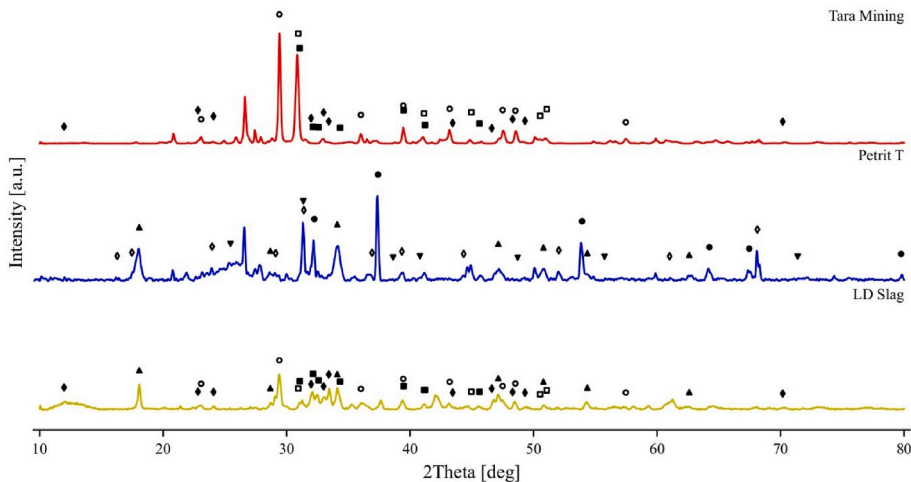


Fig. 3. Main Ca bearing mineral phases identified by PXRD in the raw materials: ● CaO (Lime, PDF#01-082-1690), ▲ $\text{Ca}(\text{OH})_2$ (Portlandite, PDF#01-076-0571), ▼ CaSO_4 (Anhydrite, PDF#01-086-2270), ○ CaCO_3 (Calcite, PDF#01-080-9776), ■ Ca_2SiO_4 (Larnite, PDF#00-033-0302), □ $\text{CaMg}(\text{CO}_3)_2$ (Dolomite), ◆ $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Srebrodolskite, PDF#01-084-9445), ◇ $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (Gehlenite, PDF#01-074-1607), a.u. = arbitrary unit.

$$\eta_T = \eta_E \times \eta_P \quad (5)$$

3. Results and discussion

This section begins by comparing the Ca leaching behavior and $\text{Ca}(\text{OH})_2$ precipitation of three different alkaline by-products, each with distinct origins and compositions, using two different solvents. After this comparison, the analysis focuses on the fate of the most problematic species in waste.

3.1. Sample composition

The mineral and elemental compositions of the three alkaline by-products were characterized using powder X-ray diffraction (PXRD) and inductively coupled plasma-sector field mass spectrometry (ICP-SFMS), with the results presented in Fig. 3 and Table 1, respectively. The PXRD patterns indicate a complex mixture of crystalline calcium-bearing phases, reflecting the varied origins and histories of the materials. Major identified phases include Lime (CaO), Portlandite ($\text{Ca}(\text{OH})_2$), Calcite (CaCO_3), Anhydrite (CaSO_4), Larnite (Ca_2SiO_4), Dolomite ($\text{CaMg}(\text{CO}_3)_2$), Srebrodolskite ($\text{Ca}_2\text{Fe}_2\text{O}_5$), and Gehlenite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$). The identification was supported by matching diffraction peaks with standard reference patterns from the ICDD PDF-4+ database, as detailed in the figure caption. Calcite (CaCO_3) was identified in the LD-slag and Tara mining samples but was absent in the Petrit-T material. In Tara mining waste, CaCO_3 likely originates from co-extracted limestone. In LD-slag, it forms post-production through atmospheric carbonation, as steelmaking temperatures prevent CaCO_3 formation during processing. The absence of calcite in Petrit-T suggests minimal weathering or recent processing.

It can be seen in Table 1 that Ca is the most dominant element in all the industrial byproducts. The rather high fraction of V and Cr in LD-slag and S in Petrit T and Tara mine tailings is noteworthy. Such high concentrations likely make these materials unapplicable for landfilling or other uses in open systems when present in soluble solid minerals.

3.2. Effect of solvent

This section explains how the use of various solvents affects the results of the experiments. For the three by-products, the pH of the filtrate from stage 1 varies between 0.08 and 1.31 (HCl) and 7.3–9.5 (NH_4Cl) depending on the type of extraction agent utilized and the byproduct used. As a result, the amount of base needed for different samples to obtain a pH value of 9.5 during stage 2 varies significantly.

3.2.1. Role of calcination in the extraction of Ca

Depending on whether the material is pre-calcined or not, the procedure using NH_4Cl as a solvent showed highly variable results with respect to Ca-extraction performance. It was found that the extraction efficiency for LD-slag and Tara mining with NH_4Cl was very low for untreated samples as given in Fig. 4 in supplementary document. This is likely due to the lower solubility of calcite in NH_4Cl compared to free CaO or $\text{CaO} \cdot \text{SiO}_2$ [39]. Limestone (CaCO_3), which accounts for a significant weight proportion of the Tara Mining material, is most likely derived from the rock material that is extracted and processed together with the ore. The limestone content, however, can be correlated with the material's freshness in the case of LD-Slag. Due to the high temperatures (1600–1650°C) in the basic oxygen furnace (BOF) and Linz-Donawitz (LD) converter, the by-product of the steelmaking process usually comprises free lime, slaked lime, or lime silicates but not calcium carbonate [40]. The material is typically exposed to atmospheric CO_2 , resulting in the formation of calcium carbonate in the pile's outer layer.

The samples need to be calcined to bring them to the state in which free lime or slaked lime are present. It is important to point out that the current batch of LD-slag had a long history of storage outside, but in a continuous process, no or limited carbonation is expected in produced LD-slag as we do not expect that it will be stored outside for such long times when used in the current process. Tara mine tailings, on the other hand, inherently have large fractions of calcite which need to be considered in any extraction process. The Petrit T sample had solely free lime (CaO) from the start, which made it possible to extract significant amounts of Ca with NH_4Cl even without calcination. Since CaCO_3 can be easily dissolved by HCl, calcining the minerals is expected to have no impact on how well the extraction works for this solvent. It is also important to note that many negative CO_2 emissions pathways are likely to involve CO_2 storage, which would necessitate a calcination step to regenerate $\text{Ca}(\text{OH})_2$ from the carbonated material. This regenerated $\text{Ca}(\text{OH})_2$ could then be reused in subsequent carbonation cycles. As a result, any initial carbon present in the material could potentially be addressed as part of this integrated process and is likely to be relatively minor in any case.

To improve calcium extraction using NH_4Cl , the LD slag and Tara mining samples were calcined in air at 950°C for 5 h. This

Table 1
Elemental composition (wt.%) of LD slag, Petrit T and Tara mine tailings as determined by ICP-SFMS.

Element	Ca	Fe	Mg	Si	Al	Zn	V	Mn	S	Cr	Ti	Na	P
LD slag	33.80	15.50	4.78	5.60	0.67	- ^a	1.81	2.31	0.09	0.35	0.63	0.04	0.24
Petrit T	22.40	8.66	0.42	9.17	3.10	0.001	0.06	0.08	0.71	0.01	0.24	0.17	0.05
Tara Mine Tailings	28.50	3.42	3.85	13.00	2.25	0.29	0.01	0.29	2.38	0.10	0.10	0.32	0.05

^a Below limit of detection.

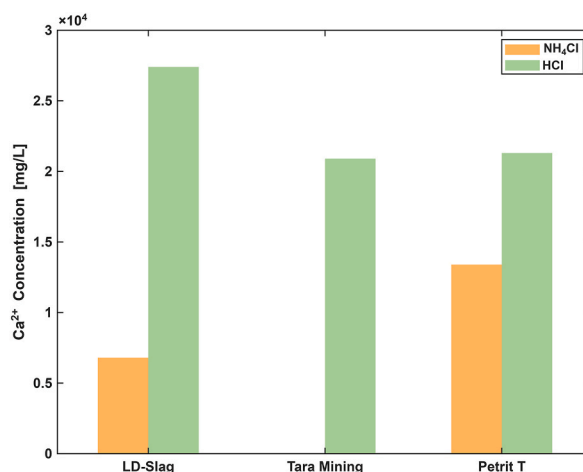


Fig. 4. Dissolved Ca-ion concentrations for both solvents determined by ICP-OES for non-calcined materials.

calcination step primarily aimed to decompose calcium carbonate (CaCO_3) into calcium oxide (CaO) and carbon dioxide (CO_2), thereby increasing the amount of extractable calcium from CaO using the mild NH_4Cl extractant. The PXRD patterns of the calcined materials, shown in Fig. 5, confirm the formation of CaO (lime) and the absence of CaCO_3 . Other phases such as larnite (Ca_2SiO_4), srebrodolskite ($\text{Ca}_2\text{Fe}_2\text{O}_5$), and gehlenite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) are also present, reflecting the complex mineralogy of these by-products. This transformation enhances calcium availability for subsequent extraction.

The main phases identified in the calcined LD slag and Tara mining samples as well as the non-calcined Petrit T sample are given in Table 2. The XRD patterns, along with the identified peaks, are included in the supplementary material.

As seen in Table 2 and Fig. 5, there is no CaCO_3 detected by XRD after the calcination of LD slag and Tara mining samples. The XRD plot of the Petrit T sample indicates the presence of significant amounts of lime and portlandite. Calcium is also found in calcium sulphate (anhydrite) and Ca-Al silicate (gehlenite) in Petrit T. The dominant phases in the diffractogram of calcined LD slag sample are lime, larnite and srebrodolskite whereas the diffractogram of Tara mining shows a mixture of lime, portlandite, larnite, srebrodolskite and gehlenite. In addition, several new minor phases appeared, indicating that other reactions and transformations also happened during the high-temperature process.

3.2.2. Calcium dissolution and precipitation of impurities

The Ca concentrations in the filtrates after each stage are shown in Fig. 6. In the case of extraction with HCl, changes in the Ca concentration in filtrates from stages 1 and 2 are observed as seen in Fig. 6 (a). The drop in Ca concentration in stage 2 can be due to co-precipitation of Ca along with other impurities.

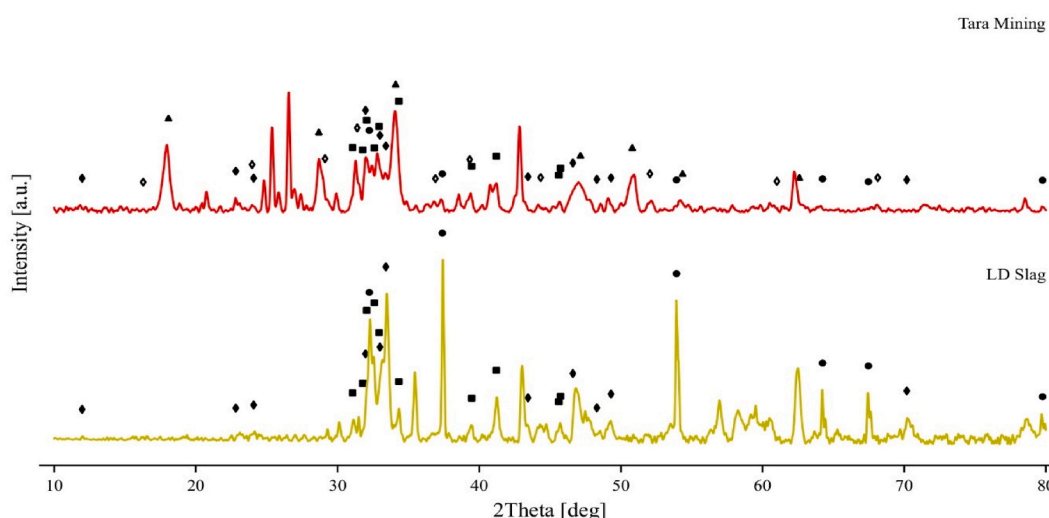
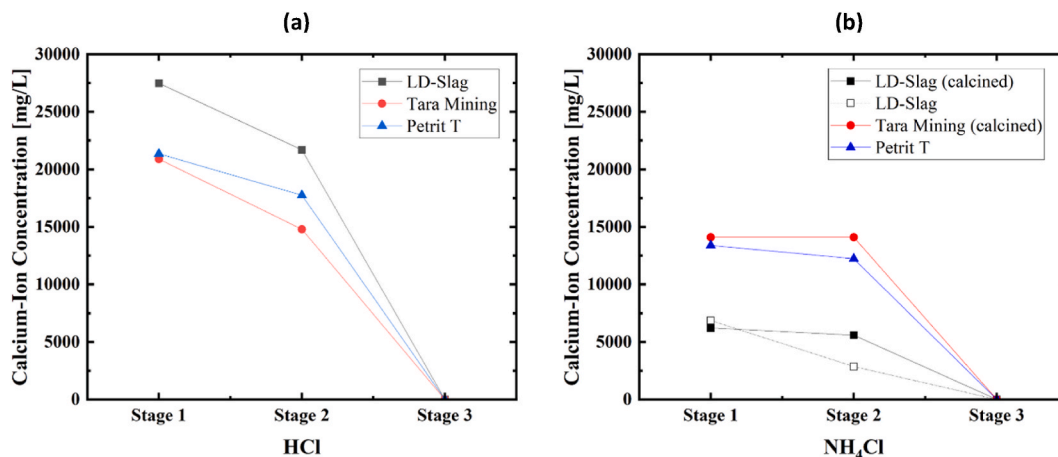


Fig. 5. Main Ca bearing mineral phases identified by PXRD in the calcined materials: ● CaO (Lime, PDF#01-082-1690), ▲ $\text{Ca}(\text{OH})_2$ (Portlandite, PDF#01-076-0571), ■ Ca_2SiO_4 (Larnite, PDF#00-033-0302), ◆ $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Srebrodolskite, PDF#01-084-9445), ◇ $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (Gehlenite, PDF#01-074-1607).

Table 2

Main phases of the raw materials identified by PXRD.

LD slag (<i>calcined</i>)	Tara Mining (<i>calcined</i>)	Petrit-T
MgO (<i>Periclase</i>)	MgO (<i>Periclase</i>)	SiO ₂ (<i>Quartz</i>)
CaO (<i>Lime</i>)	SiO ₂ (<i>Quartz</i>)	CaO (<i>Lime</i>)
Ca ₂ Fe ₂ O ₅ (<i>Srebrodolskite</i>)	CaO (<i>Lime</i>)	Ca(OH) ₂ (<i>Portlandite</i>)
Ca ₂ SiO ₄ (<i>Larnite</i>)	Ca ₂ Fe ₂ O ₅ (<i>Srebrodolskite</i>)	CaSO ₄ (<i>Anhydrite</i>)
Fe ₃ O ₄ (<i>Magnetite</i>)	Ca(OH) ₂ (<i>Portlandite</i>)	Fe ₃ O ₄ (<i>Magnetite</i>)
MgFe ₂ O ₄ (<i>Magnesioferrite</i>)	CaSO ₄ (<i>Anhydrite</i>)	Ca ₂ Al ₂ SiO ₇ (<i>Gehlenite</i>)
Fe ₂ VO ₄ (<i>Vanadium ferrite</i>)	Ca ₂ SiO ₄ (<i>Larnite</i>)	CaAl ₂ Si ₂ O ₈ (<i>Anorthite</i>)
	Ca ₂ MgSi ₂ O ₇ (<i>Åkermanite</i>)	
	Ca ₂ Al ₂ SiO ₇ (<i>Gehlenite</i>)	
	FeS ₂ (<i>Marcasite</i>)	

**Fig. 6.** Calcium concentrations evolving over the three stages for solvent (a) HCl and (b) NH₄Cl for calcined and non-calcined samples.

As seen in Fig. 6 (b), after the calcination, the Ca extraction with NH₄Cl enhanced significantly for the calcined Tara mining sample. However, the extracted Ca from the calcined LD-Slag sample was surprisingly only slightly lower than that from the un-calcined sample. Since there are no unexpected chemical phase changes during calcination as seen in XRD analyses, the observed reduction in calcium dissolution from LD slag after calcination may be attributed to a physical phase transformation occurring during the high-

Table 3

Extraction efficiencies (based on elemental analysis of the alkaline byproducts, residues from stage 1 using ICP-SFMS and the ICP-OES measurements of the solutions).

		LD slag	Tara mining	Petrit T
Industrial byproduct	g	10.04	10.01	10.01
Ca content	%	33.8	28.5	22.4
Total Ca in byproduct	g	3.39	2.85	2.24
HCl				
Stage 1 residue	g	2.87	3.93	5.07
Residue Ca composition	%	13.10	6.80	3.92
Ca in stage 1 residue	g	0.37	0.26	0.20
Ca extracted	g	3.02	2.59	2.04
Extraction efficiency (η_E)	%	88.9	90.6	91.1
Precipitation efficiency (η_P)	%	78.9	70.8	83.2
Total Ca conversion efficiency (η_T)	%	70.1	64.1	75.8
NH ₄ Cl ²				
Ca concentration in stage 1 filtrate with HCl	mg/L	27480.71	20887.42	21343.51
Ca concentration in stage 1 filtrate with NH ₄ Cl	mg/L	6212.31	14097.32	13394.90
Ca extracted	g	0.68	1.75	1.28
Extraction efficiency (η_E)	%	20.0	61.1	57.2
Precipitation efficiency (η_P)	%	90.1	100.0	91.3
Total Ca conversion efficiency (η_T)	%	18.0	61.1	52.2

²LD slag and Tara mining samples have been pre-calcined for use with NH₄Cl.

temperature calcination process. This transformation likely leads to the formation of less soluble phases that accumulate on the surface of the LD slag particles, thereby hindering the solvent's ability to access the more soluble phases within the particles. The Ca concentrations in the filtrate after stages 1 and 2 using NH_4Cl are quite similar because with NH_4Cl , there is no precipitation of any impurities in stage 2 due to the partial selectivity of NH_4Cl for Ca. Only the non-calcined LD-slag sample shows a difference in Ca concentration between stages 1 and 2.

The PXRD scans of the precipitated material from stage 2 were very noisy, making it difficult to determine the mineral phase of calcium. This issue requires a detailed investigation.

The Ca extraction efficiencies for both solvents are shown in Table 3. The total calcium extracted in stage 1 using HCl as a solvent has been calculated based on the elemental compositions and mass balances of the industrial byproduct samples and the residues from stage 1. The amount of calcium extracted using NH_4Cl is estimated by comparing its calcium extraction to that of HCl. This is done by first determining the ratio of calcium concentrations in NH_4Cl and HCl for a given byproduct using ICP-OES. Since both extractions used the same amount of raw material and solvents, the calcium extracted with NH_4Cl is calculated by multiplying this ratio by the amount of calcium extracted with HCl. The Ca extraction efficiencies are then calculated using Equation (1).

Since the Ca extraction efficiency depends on the type of solvent, the concentration of solvent, particle size, solid to liquid ratio (SLR) and the temperature, it is not easy to have a direct comparison with other studies, however, the Ca extraction efficiency values reported in Table 3 for HCl match well with the maximum efficiencies reported in the literature ($\geq 90\%$) since the parameters in this study were already optimized [23,41]. The Ca extraction efficiency for LD slag using NH_4Cl is also close to what has been reported by Lee ($\sim 28\%$) [23]. However, better efficiencies have been obtained using Tara mine tailings and Petrit T.

3.2.3. $\text{Ca}(\text{OH})_2$ precipitation

After increasing the pH of the filtrate from stage 2 to 3 by adding 1M NaOH, the extremely alkaline conditions cause all the calcium from stage 2 to precipitate for all the cases. The precipitate is examined using PXRD. Since the diffraction patterns of the final precipitate for all six cases, independent of the extraction method, match well with the pure $\text{Ca}(\text{OH})_2$ (portlandite) [42], it is safe to assume that the purity is sufficient.

The precipitated $\text{Ca}(\text{OH})_2$ obtained in stage 3 was dried at 120°C for 6 h prior to weighing. The amount of precipitated $\text{Ca}(\text{OH})_2$ was also calculated using Equation (3), which considers the Ca concentrations in the filtrate after stage 2 and stage 3 (ICP-OES) and the original volume of filtrate after extraction (stage 1). These calculations are denoted as theoretical values. Variations between the gravimetrically determined amount and the theoretical values are given in Fig. 7.

The theoretically calculated amounts of $\text{Ca}(\text{OH})_2$ using ICP-OES results are consistently higher than the gravimetrically determined amounts of $\text{Ca}(\text{OH})_2$ except for the case where Ca from LD slag is extracted with NH_4Cl . The Petrit T sample using HCl as solvent was found to have the highest deviation with a variance of factor 2 for $\text{Ca}(\text{OH})_2$ precipitated in the lab. Compared to the lower values, the other variances ranged from 3.4% to 17%. For LD slag, the amount of $\text{Ca}(\text{OH})_2$ precipitated varies the most between the two solvents. The discrepancies observed may result from product loss occurring during filtration, washing, or drying steps, leading to a lower measured mass experimentally.

A difference in precipitation performance between the two methods calculated using Equation (4) is given in Table 3. Although the HCl approach dissolves more calcium ions than the NH_4Cl method, the precipitation efficiency is, however, lower. This is also consistent with the values shown in Fig. 6 where the Ca ion concentrations fall for the HCl approach in stage 2. While between 90 and

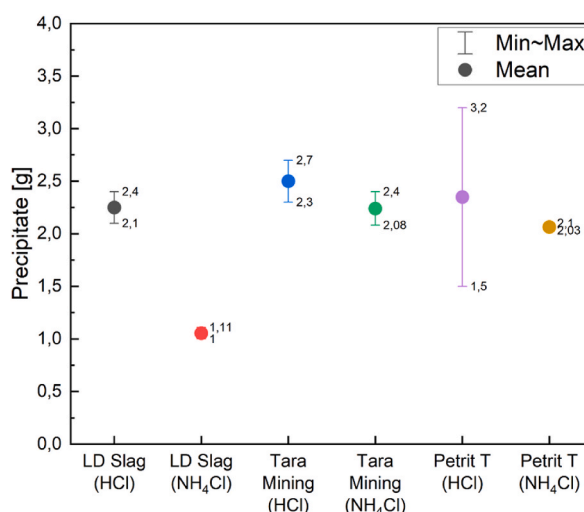


Fig. 7. Difference in gravimetric and calculated values for the final precipitate of Portlandite, $\text{Ca}(\text{OH})_2$ from the three waste products and the two solvents

¹The $\text{Ca}(\text{OH})_2$ precipitate using LD-slag with HCl solvent is calculated and measured for stage 1 filtrate of 60 ml and not 100 ml due to solvent wastage because of filter clogging.

100% of the dissolved calcium in the NH_4Cl precipitates in the form of $\text{Ca}(\text{OH})_2$, the loss of calcium during stage 2 prevents a high precipitation efficiency with HCl. As mentioned above, the most probable reason for this is that some of the calcium may have precipitated with impurities during the first stage of pH-swing (stage 2). Since Fe bearing phases do not dissolve in NH_4Cl , there is no substantial precipitation of impurities during stage 2. This is evident from the mineral compositions of stage 1 residues determined using PXRD given in Table 5.

3.2.4. Calcium conversion

The results regarding how much of the Ca in the initial samples of the industrial byproducts end up in the final $\text{Ca}(\text{OH})_2$ precipitate are presented in this section.

The Ca extraction, $\text{Ca}(\text{OH})_2$ precipitation and total Ca conversion efficiencies for both solvents are shown in Table 3. The total Ca conversion efficiencies have been calculated using Equation (5).

Even though the HCl method's precipitation efficiency, as determined by Equation (4), is lower, the yields of $\text{Ca}(\text{OH})_2$ precipitate for the LD slag and the Tara mining sample with HCl as extraction agent are higher than those of NH_4Cl as shown in Fig. 7 due to a higher total conversion efficiency (Table 3). When extracting with NH_4Cl , the calcined LD slag has a poor Ca extraction performance leading to poor overall Ca conversion efficiency, which is consistent with the final low amount of $\text{Ca}(\text{OH})_2$ precipitate measured in the lab. Although the conversion efficiencies using ICP-OES values indicate the reverse (Table 3), the lab measurements for the Petrit T in Fig. 7 show that less $\text{Ca}(\text{OH})_2$ is precipitated with HCl compared to the NH_4Cl procedure.

3.3. Effect of particle size

Smaller particles have been shown to result in higher extraction performance, due to higher surface to volume ratios. Additional experiments were also performed here on the non-calcined LD slag sample comprising more fines (0-90 μm) using NH_4Cl solvent, with the goal to study the impact of particle size on Ca leaching and $\text{Ca}(\text{OH})_2$ precipitation. For the non-calcined LD slag with particle size distributions of 90-250 μm and 0-90 μm , the $\text{Ca}(\text{OH})_2$ precipitates formed at the end of stage 3 were weighed and reported in Table 4. It is obvious from the results that small particles in the range of 0-90 μm result in better Ca conversion compared to the 90-250 μm range.

3.4. Effect of material composition

Using PXRD, the mineral compositions of the original samples and the phases that disappeared after the first stage extraction using NH_4Cl and HCl are shown in Table 5. The XRD patterns and the corresponding identified peaks are provided in supplementary material.

It is clear from the results shown in Table 5 that extraction in NH_4Cl is very specific in the dissolution of pure calcium minerals whereas this method does not dissolve the calcium present in the aluminum silicate minerals. On the other hand, extraction with HCl dissolves the pure calcium and magnesium minerals as expected but also less soluble minerals such as CaSO_4 , Ca_2SiO_4 . However, HCl doesn't dissolve all CaSO_4 from Tara mining sample and in fact, results in the formation of a new sulfur containing phase, Barite (BaSO_4). HCl is efficient in dissolving sulfide containing minerals such as FeS_2 and K_2S_2 from Tara mining and Petrit T, respectively. Another significant difference is the dissolution of iron bearing minerals in HCl from all three materials. Interestingly, the vanadium containing phase Fe_2VO_4 in LD slag doesn't completely dissolve in either HCl or NH_4Cl and is present in the residue from stage 1, likely making it unsuitable for landfilling due to acid leaching and metal dissolution.

3.5. Fate of non-Ca ions

For all the cases investigated in this study, the filtrates from stage 1 have been examined for seven additional ions: iron, manganese, aluminum, silicon, magnesium, chromium, and vanadium. Fig. 8 presents a plot of the different ion concentrations after stage 1 extraction with HCl and NH_4Cl .

When using NH_4Cl as a solvent, only Ca was detected in the filtrate by the ICP-OES. However, impurities resulting in HCl as the solvent are detected but remain at a level below 3500 mg/L. Only the HCl method in the case of LD slag extracted vanadium (237 mg/L), which is consistent with the total elemental composition of LD slag in Table 3. Neither silicon nor chromium is found among the other ions. The HCl filtrate also includes iron (1914 mg/L), which is one of the dominant components in LD slag. Moreover, using HCl solvent, other than Calcium, Magnesium (2222 mg/L) is also recovered from the Tara mining sample due to its abundance in Mg-bearing phases. Iron (3452 mg/L) and aluminum (798 mg/L) are also extracted from the Petrit T sample material using HCl.

Considering the final disposal of the rest products after extraction, it might be advantageous to use HCl as extraction medium in case of Petrit T because some of the problems which occur in depositing sulfide containing materials such as formation of sulfuric acid

Table 4
Ca(OH)₂ precipitate yield for non-calcined LD-slag using NH_4Cl solvent.

	Precipitate – lab (g)
LD slag (90-250 μm)	0.74
LD slag (0-90 μm)	1.13

Table 5

Mineral phase composition of the original samples and the changes in phase observed in the residue from stage 1.

	Chemical composition of original samples as determined by XRD	Minerals disappeared in the residue after stage 1 (with NH_4Cl)	Minerals disappeared in the residue after stage 1 (with HCl)
LD slag (calcined)	MgO (Periclase) CaO (Lime) $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Srebrodolskite) Ca_2SiO_4 (Larnite) Fe_3O_4 (Magnetite) MgFe_2O_4 (Magnesioferrite) Fe_2VO_4 (Vanadium ferrite)	CaO (Lime)	CaO (Lime) Ca_2SiO_4 (Larnite) Fe_3O_4 (Magnetite) New Phases FeO (Iron oxide)
Tara Mining (calcined)	MgO (Periclase) SiO_2 (Quartz) CaO (Lime) $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Srebrodolskite) $\text{Ca}(\text{OH})_2$ (Portlandite) CaSO_4 (Anhydrite) Ca_2SiO_4 (Larnite) $\text{Ca}_2\text{MgSi}_2\text{O}_7$ (Åkermanite) $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (Gehlenite) FeS_2 (Marcasite)	CaO (Lime) $\text{Ca}(\text{OH})_2$ (Portlandite)	MgO (Periclase) CaO (Lime) $\text{Ca}(\text{OH})_2$ (Portlandite) FeS_2 (Marcasite) $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Srebrodolskite) Ca_2SiO_4 (Larnite) $\text{Ca}_2\text{MgSi}_2\text{O}_7$ (Åkermanite) New Phases BaSO_4 (Barite) KAlSi_3O_8 (Microcline intermediate) CaSiO_3 (Wollastonite) Fe_2O_3 (Hematite) CaO (Lime) $\text{Ca}(\text{OH})_2$ (Portlandite) CaSO_4 (Anhydrite) K_2S_2 (Potassium sulfide)
Petrit T	SiO_2 (Quartz) CaO (Lime) $\text{Ca}(\text{OH})_2$ (Portlandite) CaSO_4 (Anhydrite) Fe_3O_4 (Magnetite) $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (Gehlenite) $\text{CaAl}_2\text{Si}_2\text{O}_8$ (Anorthite) K_2S_2 (Potassium sulfide)	CaO (Lime) $\text{Ca}(\text{OH})_2$ (Portlandite)	

might be avoided. We would like to stress that these are the first results and sulfide-related chemistry needs to be evaluated in detail. Even though NH_4Cl only dissolves a few calcium-bearing phases and S, V and Cr may not be fully leached for most cases, treating the smaller, easier-to-handle residues from stages 1 and 2 for these problematic species is still more practical.

4. Conclusions

Alkaline slags and wastes offer valuable sources of calcium for producing calcium hydroxide, which not only helps achieve negative emissions but also makes waste disposal more manageable. This study used a pH-swing method for extracting calcium and precipitating $\text{Ca}(\text{OH})_2$ utilizing three common industrial waste products (LD slag, Tara mine tailings, and Petrit-T) using two solvents (HCl and NH_4Cl) and provided a comprehensive overview of the method's applicability. The waste products investigated come from sectors with significant direct or indirect CO_2 emissions, making negative emissions particularly relevant.

The findings indicate that NH_4Cl provides a more selective extraction and higher calcium conversion, reaching up to 100%. This method eliminates the need for intermediate pH adjustments, simplifying the process and reducing reliance on compounds like NH_4OH . Although the total process efficiency of NH_4Cl is between 18 and 61%, compared to 64–75% for HCl, it remains the preferred option for Tara Mining and Petrit T due to its high yield of $\text{Ca}(\text{OH})_2$ and process simplicity.

In terms of final disposal of the residues after extraction, using HCl as the extraction medium for Petrit T could be beneficial. Since HCl dissolves all the sulfur containing phases, the issues associated with sulfide-containing materials, such as the formation of sulfuric acid during deposition, are avoided. Although NH_4Cl dissolves only a limited number of calcium-bearing phases and may not completely leach sulfur, vanadium, and chromium, it is still more practical to manage the smaller residues when dealing with these problematic elements.

In all cases, the process yielded a pure $\text{Ca}(\text{OH})_2$ precipitate suitable for cost-effective negative emission technologies, confirming the effectiveness of both solvents for the three waste products. The study also emphasized the significant influence of particle size on extraction efficiency. Optimizing factors such as the solid-to-liquid ratio, solvent concentration, and particle size could further improve calcium recovery. Future research should focus on analyzing the physical properties of the $\text{Ca}(\text{OH})_2$ precipitate and conducting carbonation tests. Additionally, reducing solvent costs and exploring solvent regeneration will be critical for the wider applicability of these extraction methods. The long-term sustainability and economic viability of the process rely on effective solvent management. A major challenge ahead is assessing whether solvents, particularly NH_4Cl , can be regenerated and reused. Without feasible recycling options, the process may become too costly due to the high price and energy demands of producing fresh chemicals. Future studies should explore solvent recovery techniques and strategies to minimize chemical consumption.

Looking forward, two important questions remain: the long-term stability of the precipitated $\text{Ca}(\text{OH})_2$ under atmospheric carbonation conditions, and the scalability of the pH-swing extraction method. Understanding how $\text{Ca}(\text{OH})_2$ behaves over extended periods and under variable environmental conditions is essential for ensuring its performance in real-world CO_2 sequestration

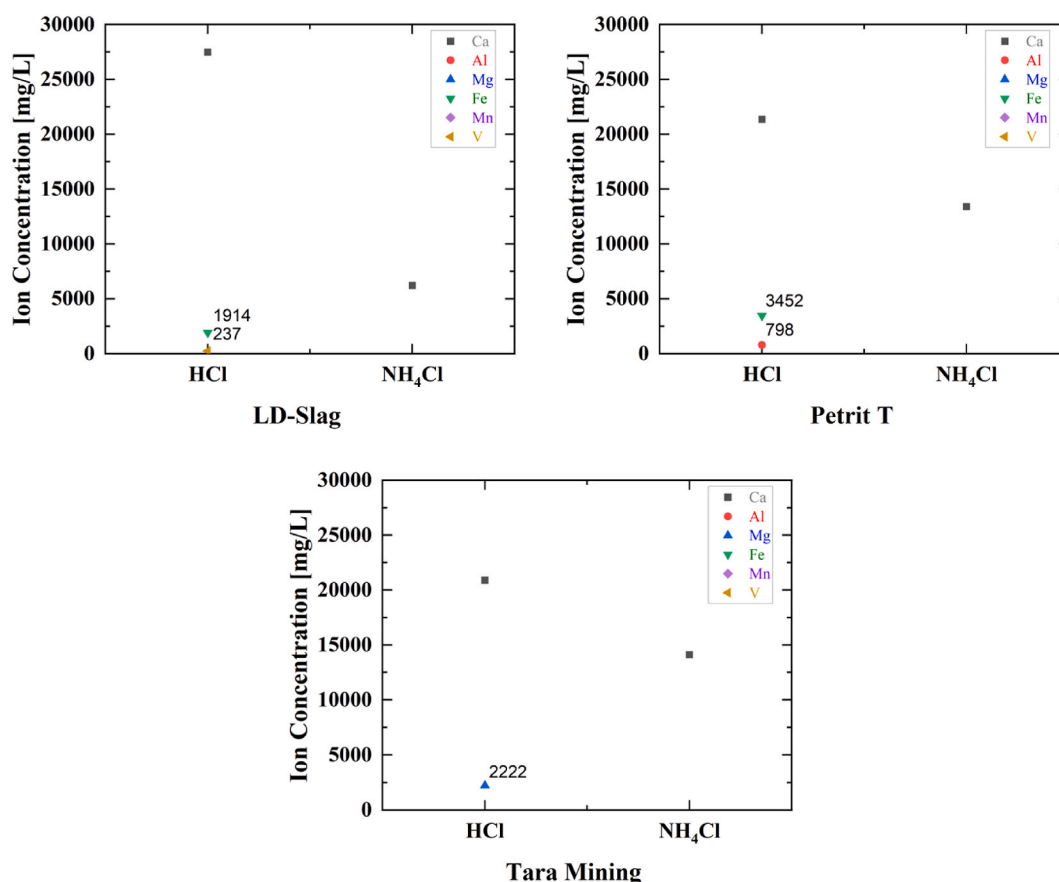


Fig. 8. Non-Ca ion concentration in the filtrate after extraction in stage 1 for the three industrial by-products for both solvents.

applications. Likewise, transitioning the pH-swing process from laboratory to industrial scale will require careful evaluation of its energy requirements, material handling challenges, and integration potential with existing waste management systems. Addressing these open questions will be key to advancing this promising approach toward practical, sustainable, and economically viable carbon capture solutions.

CRediT authorship contribution statement

Muhammad Nauman Saeed: Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation, Data curation. **Anna Stedt:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Julian Scheer:** Visualization, Validation, Software, Methodology, Investigation, Data curation, Conceptualization, Formal analysis, Writing – original draft, Writing – review & editing. **Tobias Mattisson:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Zareen Abbas:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation. **Mika Järvinen:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Investigation, Conceptualization. **Carl Linderholm:** Writing – review & editing, Visualization, Funding acquisition, Conceptualization, Investigation, Methodology, Supervision, Validation.

Declaration of the use of generative AI and AI-assisted technologies

During the preparation of this work, ChatGPT has been used to improve readability and language. After using this tool, the content has been reviewed and edited as needed. Full responsibility for the content of the publication is taken.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Muhammad Nauman Saeed reports financial support was provided by Swedish Energy Agency. Carl Linderholm reports

financial support was provided by Swedish Energy Agency. Tobias Mattisson reports financial support was provided by Swedish Energy Agency. Zareen Abbas reports financial support was provided by Swedish Energy Agency. Anna Stedt reports financial support was provided by Swedish Energy Agency. Mika Jarvinen reports financial support was provided by K H Renlund Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We would like to acknowledge the financial support for this project from the Swedish Energy Agency (Project P2022-00211) and Renlund Foundation (Finland).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.heliyon.2026.e45140>.

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