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Process Technology

CO₂ capture through mineralization of CaO- containing by-products

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Summary:

The amount of CO₂ in the atmosphere is continuously increasing, resulting in climate change and global warming. Industrial processes contribute a substantial share in the amount of CO₂ released to the atmosphere. On the other hand, different types of wastes and by-products are being produced by different industries which are deemed pollutants and require energy and capital to be safely managed through circular economy perspective. In many cases, the amount of waste is so high that it cannot be handled and is freely piled in nature. Hence, a solution to simultaneously tackle both the CO₂ emission and waste pollution problems would be of high value.

CO₂ sequestration by mineralization of CaO-rich industrial wastes is one potential solution. In such a process, CO₂ reacts with the CaO in the waste and CaCO₃ is produced. This product is thermodynamically stable and has multiple uses. Many studies in the literature have reported use of various CaO-rich wastes to capture CO₂, but they are mostly based on lab-scale experiments, and mostly the focus is on the chemistry of the suggested processes. Hence, there is a need to study the technical and economic feasibility of up-scaled industrial versions of such processes.

In this study and after a comprehensive systematic literature review, four different mineralization processes applying different chemicals and using different CaO-rich wastes, all with a relatively high performance documented from laboratory experiments, were chosen. These processes were scaled up to the industrial size of a pilot plant with capacity of 400 tons of CO₂ to be captured in a year and outlined with the required process equipment. Based on published lab results, mass and energy balances of the up-scaled processes were then performed, and key performance indicators of the processes in three different countries were calculated using an in-house-made process simulation tool. Furthermore, a comparative technical, economic, and environmental analysis was conducted for all processes.

The results indicate that process 2 (using recycled concrete fines as waste and NH₄Cl as reagent) has the highest amount of CO₂ captured per mass of industrial waste (0.33 kg/kg) and consumes the least amount of reagent per mass of captured CO₂ with 0.5 kg/kg. Meanwhile, process 1 (using converter slag as waste and NH₄Cl as reagent) has the least energy consumption per mass of captured CO₂ with 1.48 kWh/kg. Process 3 is characterized by blast furnace slag as waste and HCl as reagent. All processes are economically feasible in each scenario except process 4 (using converter slag as waste and CH₃COOH as reagent). This means that CO₂ sequestration via mineralization using CaO-rich industrial wastes is a promising solution not only in terms of circularity and emitted CO₂ reduction, but also as an attractive business case.

Preface

The urgency of addressing climate change has placed a significant emphasis on finding viable solutions to mitigate greenhouse gas emissions, particularly carbon dioxide. Among the various strategies proposed, carbon capture and storage (CCS) has emerged as a promising avenue to reduce CO₂ emissions from industrial sources. Within the realm of CCS, indirect mineralization processes have drawn attention for their potential to not only capture CO₂ but to also permanently sequester it using from different industries.

This Master's Thesis delves into the realm of CO₂ capture through indirect mineralization processes, focusing on the mechanisms involved, technological advancements, and environmental implications. Through an interdisciplinary approach, this study aims to provide a comprehensive understanding of the technical, economical, and environmental aspects associated with indirect mineralization, shedding light on its feasibility as a sustainable CO₂ mitigation strategy.

The journey embarked upon in this thesis begins with a thorough review of the literature in indirect mineralization processes. By exploring the principles underlying mineral carbonation and its application in CO₂ capture, this thesis lays the groundwork for a deeper exploration into the potential and limitations of indirect mineralization.

By combining theoretical analysis with practical experimentation, this study aims to elucidate key factors influencing the efficiency and scalability of indirect mineralization processes.

Ultimately, this thesis aspires to serve as a catalyst for continued exploration and innovation in the realm of CO₂ capture and utilization. By fostering a deeper understanding of indirect mineralization processes, it is hoped that this research will inspire further advancements towards a sustainable future, wherein CO₂ emissions are not only mitigated but also transformed into valuable resources for benefit.

It is imperative to acknowledge and appreciate the collaborative efforts and support extended by Prof. Lars-Andre Tokheim as my supervisor and professor from University of South-Eastern Norway (USN) whose guidance and feedback played a main role in shaping this thesis.

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Finally, I want to thank my family whose kindness and support, despite the geographical distance between us, were heartwarming while working on the thesis.

May this thesis serve as a testament to the collective commitment towards addressing the global challenge of climate change and ushering in a more sustainable and resilient world.

Porsgrunn, 15 May 2024

Amirhossein Ghazi

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Nomenclature

Symbol or Abbreviation	Meaning	Unit
Al ₂ O ₃	Aluminum oxide	-
BFS	Blast furnace slag	-
C	Specific heat capacity	kJ/kg°C
Ca	Calcium	-
CaCO ₃	Calcium carbonate	-
CaO	Calcium oxide	-
CAPEX	Capital expenditure	USD
CCS	Carbon capture and storage	-
CH ₃ COOH	Acetic acid	-
CO ₂	Carbon dioxide	-
COP	Coefficient of performance	-
CS	Converter slag	-
DOI	Digital object identifier	-
EOR	Enhanced oil recovery	-
F ₈₀	Primary particle size	μm
Fe ₂ O ₃	Ferric oxide	-
g	Acceleration of gravity	m/s ²
h	Head loss of the system	m
HCl	Hydrochloric acid	-
HE	Heat exchanger	-
KPI	Key performance indicator	-
KPP	Key performance parameter	-

Symbol or Abbreviation	Meaning	Unit
LP	Leaching pressure	bar
LT	Leaching temperature	°C
L_v	Latent heat of evaporation	kJ/kg
m	Mass flow rate	kg/s, kg/min
M	Molar mass	kg/mol
MgO	Magnesium oxide	-
Mn	Manganese	-
MnO	Manganese(II) oxide	-
MOF	Metal-Organic Framework	-
MP	Mineralization pressure	MPa, bar, atm
MT	Mineralization temperature	°C
NH ₄ Cl	Ammonium chloride	-
NOAA	National Oceanic and Atmospheric Administration	-
NPSH	Net positive suction head	m
OPEX	Operational expenditure	USD
P	Power	kW
P&ID	Process and instrumentation diagram	-
P ₂ O ₅	Phosphorus pentoxide	-
P ₈₀	Final particle size	μm
PCC	Precipitated calcium carbonate	-
PFD	Process flow diagram	-
PFR	Plug flow reactor	-
P _h	Hydraulic power of pump	kW
ppm	Part per million	-

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Symbol or Abbreviation	Meaning	Unit
q	Flow rate	m ³ /h
Q	Heat	kW
RCF	Recycled concrete fines	-
S	Sulfur	-
S/L	Solid to liquid ratio	g/L
SiO ₂	Silicon dioxide	-
T	Temperature	°C
Ti	Titanium	-
W	Energy consumption	kWh/ton
W _i	Bond's crushing work index	kWh/ton
ZIF	Zeolitic Imidazolate Framework	-
ρ	Density	kg/m ³

1 Introduction

The amount of heavy pollutants generated by industrial facilities has increased dramatically since the start of the industrial revolution, primarily due to the combustion of fossil fuels [1]. The amount of CO₂ in the atmosphere has surpassed 400 ppm and is projected to surpass 500 ppm by 2050, leading to an elevated average world temperature in the event that appropriate measures are not implemented to mitigate the issue [2]. Figure 1.1 shows the increasing trend of CO₂ in the atmosphere up to 2021. The combined land and ocean temperatures have risen since 1850 at an average pace of 0.06 degrees Celsius every decade, according to National Oceanic and Atmospheric Administration's (NOAA) 2023 Annual Climate Report [3].

Considering the projected population growth, by 2050, there will be around 10 billion people on the planet, resulting in a rise in the demand for energy and water. Nevertheless, fossil fuels are still providing more than 84% of the world's energy needs, placing significant pressure on the growing trajectory of greenhouse gas emissions [4]. Taking into account all points mentioned above, utilizing alternative energies such as renewables and CO₂ capture seem to be the only solutions to tackle these challenges.

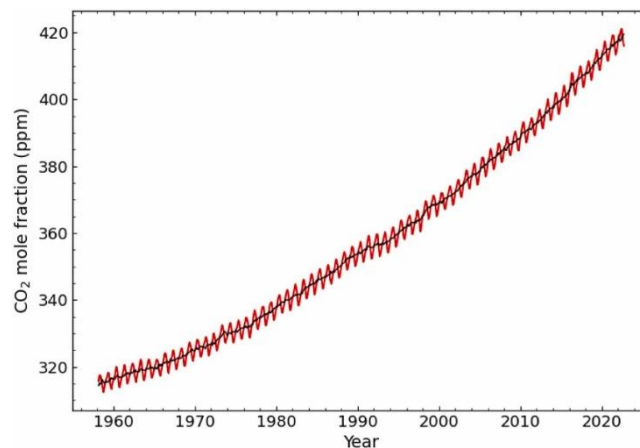


Figure 1.1: Increase in CO₂ mole fraction in the atmosphere in recent decades [4]

1.1 CO₂ Capture methods

Carbon capture and storage (CCS) is the process of capturing CO₂ generated during the production of electricity and other industrial activities and storing it to make sure it is not discharged into the environment. It is possible to significantly reduce CO₂ emissions from energy systems by utilizing CCS methods [5]. Many methods can be used to capture CO₂ at the source [6]. Then, instead of being kept underground, captured CO₂ might occasionally be used to produce goods, in industrial processes, and in other activities. The two primary categories of CO₂ capturing methods are: indirect carbon capture, which includes enhanced weathering-natural absorption, afforestation, reforestation, and bio-energy options with carbon capture, and direct carbon capture, which includes pre-, post-, and oxy-combustion, and air

capture systems [5]. Figure 1.2 depicts these methods. Pre-, post-, and oxy-combustion methods will be briefly discussed in the next section.

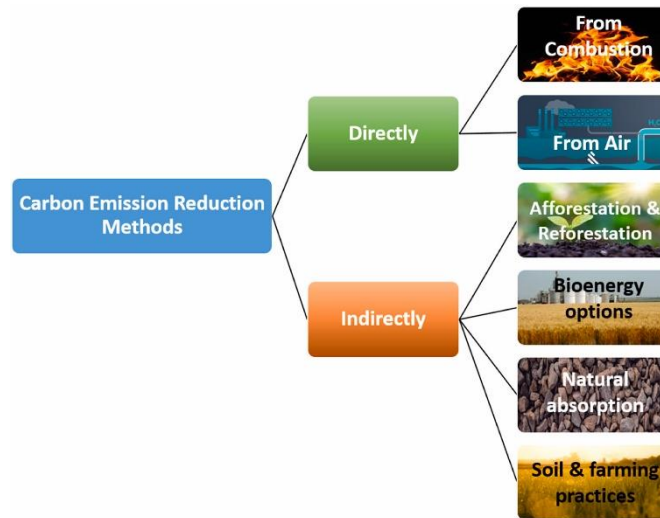


Figure 1.2: CO₂ capture methods [5]

Pre-combustion CO₂ capture methods

Fuel is oxidized through a gasification process in pre-combustion carbon capture, resulting in syngas that is mostly composed of carbon monoxide (CO) and hydrogen (H₂). The generated CO is turned into CO₂, which is then captured before combustion. The studies indicate that this process can reduce the volume of combustible fuel due to separation of CO₂ which may have its drawbacks [7].

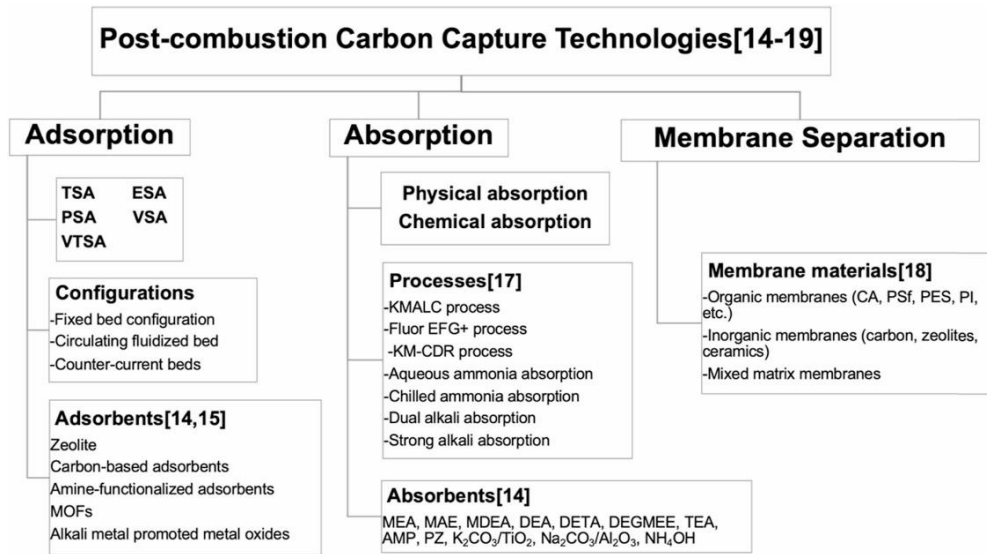
Oxy-combustion CO₂ capture methods

Substitution of oxygen with air for combustion is known as oxy-combustion. With this method, it is ensured that the whole exhaust gas is CO₂ which does not need any further separation. This method has some advantages such as higher flame temperatures since there is no nitrogen gas playing the role of dilutant, and has some disadvantages such as high price of pure oxygen or difficulty in separating the oxygen from air to be used for combustion [8].

Post-combustion CO₂ capture methods

By using physical or chemical adsorption/absorption mechanisms, the post-combustion CO₂ capture method can extract CO₂ and other gases from industrial activities' flue gases. It can be categorized as chemical reactions, adsorption, absorption, and membrane separation capture technologies. Figure 1.3 shows a summary of these technologies [9].

The following section will discuss the post-combustion CO₂ capture.

Figure 1.3: Post-combustion CO₂ capture technologies [9]

1.2 Post-combustion CO₂ capture technologies

There are three main post-combustion CO₂ capture technologies: absorption, membrane separation, and adsorption.

1.2.1 Absorption

Absorption technology is divided into two main categories: physical and chemical absorption.

Physical absorption

Henry's law serves as the foundation for the physical absorption process. High pressure and low temperature are used to absorb CO₂, while low pressure and high temperature are used to desorb it. This technology has been widely used in a variety of industrial operations, including the production of hydrogen with high CO₂ concentrations, synthesis gas, and natural gas. Numerous commercial processes are currently in use, including the Fluor, Purisol, Rectisol, Selexol, and Morphysorb. Dimethylether or propylene glycol is used as an absorbent in the Selexol process; methanol is used in the Rectisol process; N-methylpyrrolidone is used in the Purisol process; morpholine is used in the Morphysorb process; and propylene carbonate is used in the Fluor process [10].

Chemical absorption

The absorber and the stripper, which thermally regenerates the absorbent, are the two main components of a normal chemical absorption process. The flue gas carrying CO₂ enters a packed bed absorber from the bottom and makes counter-current contacts with a CO₂-lean absorbent during the chemical absorption process. Once absorbed, the CO₂-rich absorbent flows into a stripper for thermal regeneration. The CO₂-lean absorbent is fed back to the

absorber for cyclic use following regeneration. Compressed CO₂ is produced exclusively by the stripper and is then stored and transported. The absorber and stripper typically have temperatures between 40 and 60 °C and 120 and 140 °C, respectively, and the operation pressure is around 1.0 bar [10].

1.2.2 Membrane separation

Membranes are used in gas separation to selectively allow certain gaseous components to pass through from one side of a barrier to the other. A concentration gradient causes the components to dissolve in the polymer at one surface and transfer across the membrane. A high partial pressure of the essential gas components on one side of the membrane barrier and a low partial pressure on the other preserves the concentration gradient. A suitable membrane with high CO₂ permeability and selectivity should be used in the CO₂ capture procedure. One of the hardest things in the realm of membrane research is to break through the permeability-selectivity trade-off and attain such a performance [11].

1.2.3 Adsorption

Adsorption is a CO₂ capture method whereby CO₂ is either physically or chemically captured by a solid material.

Physical adsorption

Physical adsorption happens due to weak Van der Waals forces. Less energy is used in the CO₂ capture process when physical sorbents and inorganic porous materials (such as zeolites and carbonaceous materials) are used. This is because significantly less energy is needed for CO₂ regeneration because no new bond is created between the sorbent and sorbate. Some samples of physical adsorbents could be carbon-based materials, mesoporous Silica, Zeolites, Zeolitic Imidazolate Frameworks (ZIFs), Metal-Organic Frameworks (MOFs), and blended adsorbents [12].

Chemical adsorption

Chemical adsorption occurs due to covalent bonding or electrostatic attraction. The chemical sorbents could be amine-based adsorbents, amine grafted, amine-impregnated, metal salts, double salts, and metal oxides such as calcium oxide (CaO) [12]. Figure 1.4 shows the most common physical and chemical adsorbents.

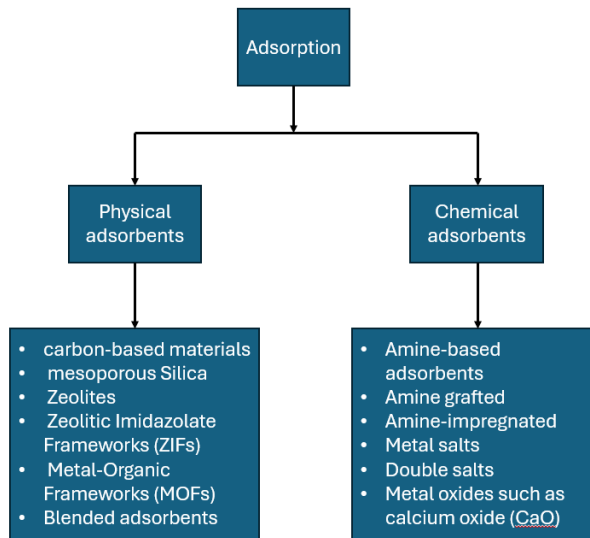


Figure 1.4: Physical and chemical sorbents

1.3 Adsorption with metal oxides

The ability of minerals containing alkaline-earth metals (such as Ca and Mg) to absorb more than 36,000 Gt of CO₂ (or 10,000 Gt of carbon) makes them the world's greatest carbon sink. The current estimate of 780–1,000 Gt of carbon that conventional fossil fuels hold can be adsorbed by this amount. There is no need for continued monitoring because the produced mineral carbonates are highly stable, inert, and unlikely to release the bound CO₂ again into the atmosphere. Thus, carbon dioxide reaction with alkaline-earth metals, known as *mineralization*, is the process of sequestering excess anthropogenic CO₂ into minerals, and it has the potential to be a practical and long-term way to reduce global warming [13].

Mineralization of silicates was introduced by W.Seifritz [14] and is a direct process for carbonation and mineralization of alkaline minerals such as olivine, serpentine and basalt which are natural materials, or industrial alkaline wastes namely ashes and slags. This, due to the slow kinetics of mineral carbonation, needs a large energy demand to accelerate, or a long reaction time under ambient conditions. In addition, directly carbonating alkaline minerals usually produces low-quality products [15]. Figure 1.5 shows different types of direct carbonation.

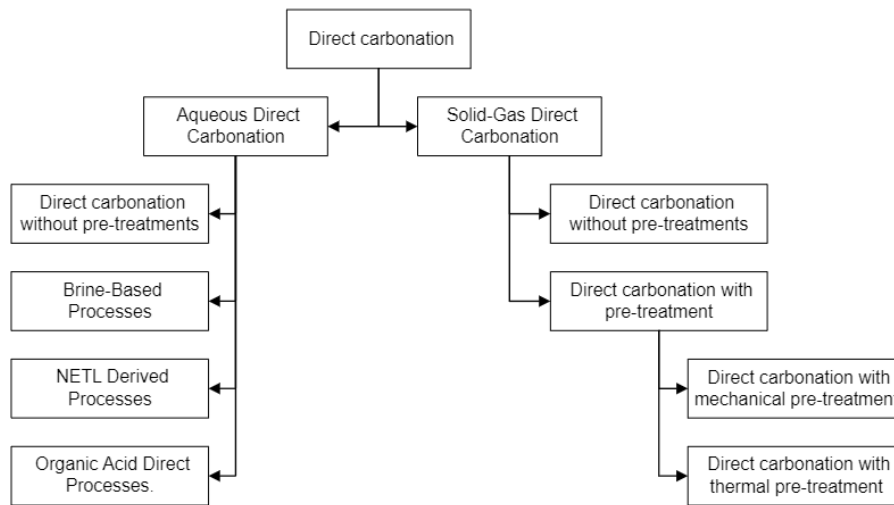


Figure 1.5: Types of direct carbonation [6]

On the other hand, there is the indirect carbonation process, which makes it possible to produce higher-value goods such as pure precipitated calcium carbonate (PCC). Prior to starting the mineral carbonation reaction, an indirect reaction must be used to extract alkaline earth metal ions from silicates using the appropriate organic or inorganic acids or salts. Because the reactions are usually occurring at ambient conditions, this method can, in the best of situations, save energy in the mineral carbonation step. However, the extraction step (also known as the enrichment or separation step) may result in an uneconomical net outcome due to the need of expensive reagents, reagent recovery, and energy consuming processes and equipment [15]. Figure 1.6 shows different types of indirect carbonation and Figure 1.7 shows a schematic of this process; the extraction step is shown within the red dashed line and the mineralization is shown within the green dashed line.

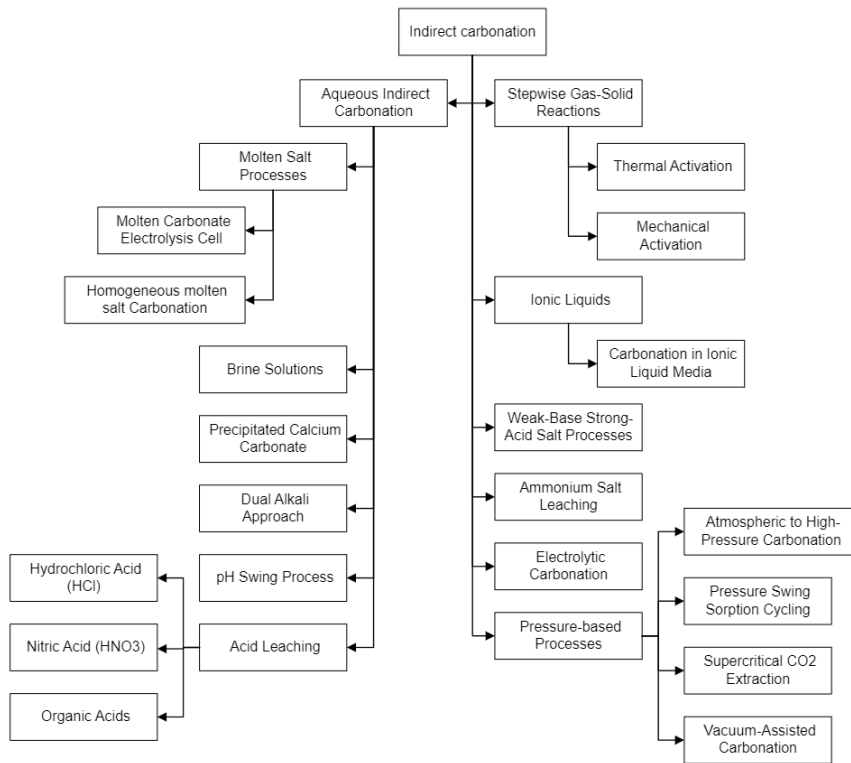


Figure 1.6: Types of indirect carbonation [6]

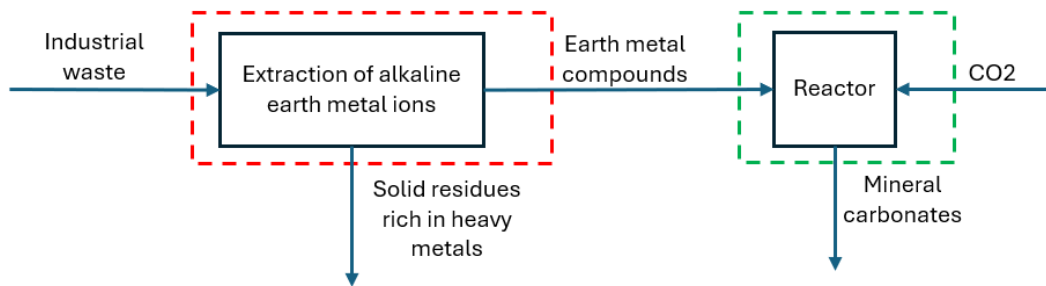


Figure 1.7: A schematic overview of indirect mineralization process

This is an obstacle on the way to implement the mineralization CO₂ capture at an industrial scale. In order to determine which processes have the lowest energy and cost intensity and the greatest amount of CO₂ captured, it is important to conduct process simulation and reaction modelling, environmental impact assessment, energy analysis, and economic evaluation in conjunction with process development. This will help determine the most promising options for scale-up [16].

1.4 Objectives and scope

As shown in Figure 1.5 and Figure 1.6, both direct and indirect processes fall into aqueous and gas-solid reactions. When compared to solid phase mineralization methods, aqueous phase mineralization has demonstrated advantages in terms of process parameters, allowing for a lower utilization of harsh conditions. Promising benefits of aqueous phase mineralization include the potential to speed up the process and the viability of large-scale implementation [16]. To study the feasibility of this technology to meet the objective of CO₂ mitigation, this thesis addresses the most recent advancements in aqueous indirect mineralization from a technical, economical, and environmental standpoint, and provides an update on the state of process development to help closing the gap between bench-scale research and industrial-scale application through the steps below:

- A literature study about utilization of CaO-rich industrial wastes and indirect CO₂ capture through mineralization.
- A subsequent detailed identification of Ca extraction methods from industrial wastes.
- Obtain the composition of the pertinent products from different extraction methods.
- Choosing four published processes and conducting mass and energy balance calculation of a pilot scale plant with capacity of capturing 400 ton of CO₂ per year for them.
- Quantifying the required mass of industrial waste, leaching reagent, and energy consumption to capture one kilogram of CO₂ using the four chosen processes.
- Make a process flow diagram for the processes.
- Simulate the pilot scale plant for the processes using an in-house-made simulation tool.
- Conduct an economic assessment of the processes in different scenarios.
- Evaluate the environmental impact of the processes by calculation of their CO₂ footprint in different scenarios.

1.5 Methodology

The methodology used in this thesis consists of eight steps. First, select four processes from the literature with laboratory study. Second, study the materials, methods, and the results in the four processes. Third, make a preliminary PFD to identify main streams of each process. Fourth, perform scale-up mass balance calculations to simulate the process in large scale. Fifth, make an improved PFD which includes the equipment with energy consumption too. Sixth, conduct detailed mass and energy balance calculations and size the equipment. Seventh, perform calculations on financial viability. Lastly, define some key performance indicators (KPIs) to compare the processes. This workflow is summarized in Figure 1.8.

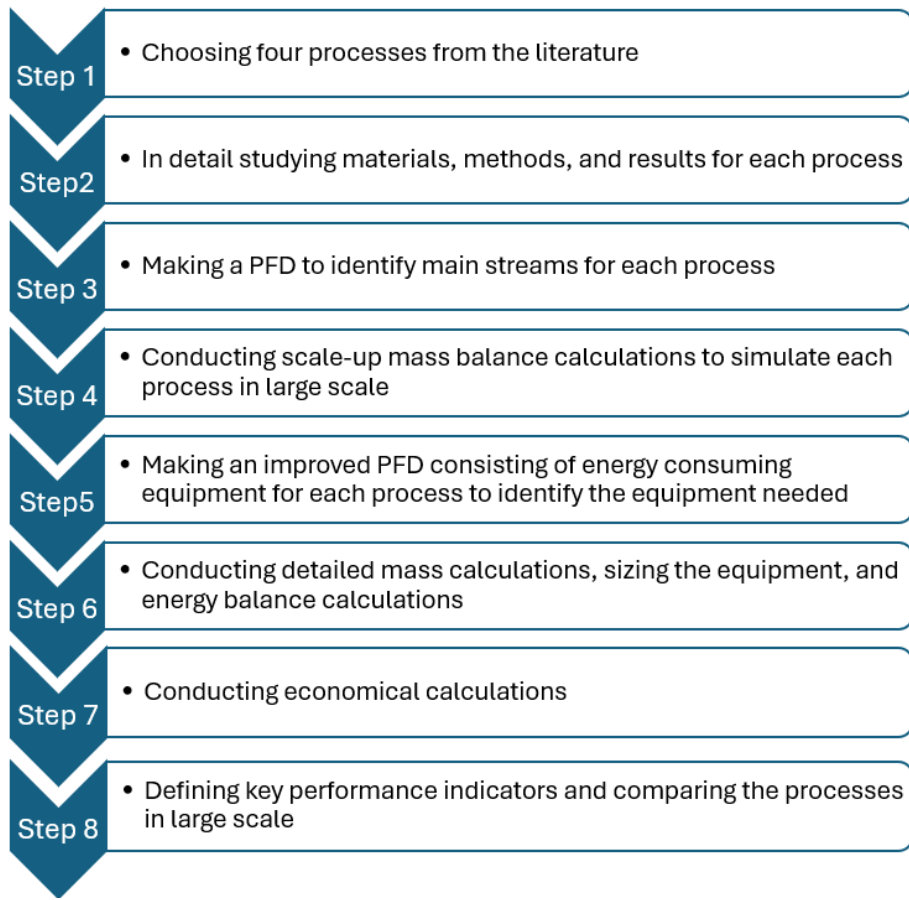


Figure 1.8: Workflow of the thesis and derivation of appropriate KPIs for process comparisons

2 Literature review

Reviews of the existing literature serve as a crucial basis for all kinds of study. They can give evidence of an effect, establish rules for policy and practice, form the basis for knowledge development, and, with careful execution, have the power to inspire new ideas and directions for a certain field. As such, they provide the foundation for upcoming studies and theories. However, carrying out a literature review and assessing its calibre might be demanding tasks. It will be much simpler to identify real research gaps rather than just carrying out the same research over and over again, to create better and more focused hypotheses and research questions, and ultimately to raise the calibre of research if it is known that the research is based on extremely accurate data [17].

2.1 Review procedure

There are a number of actions and choices that need to be made in order to produce a literature review that satisfies publication standards, regardless of the methodology chosen. The following sections outline and analyse the fundamental procedures and crucial decisions that went into performing the literature review shared here, in four stages: (1) planning the review, (2) conducting, (3) analysing, and (4) reporting [17].

2.1.1 Planning the review

This literature review is required in order to have a better overview and understanding of the CO₂ sequestration using the indirect mineralization process of CO₂ using different industrial wastes. In this regard, Zhang et al. [16] have performed a holistic literature review until 2019 which can be improved by reviewing more recent studies. Hence, in this thesis, the literature from 2020 to 2024 will be reviewed. The focus will be on materials and methods, results, and efficiencies.

This review will be beneficial for all the researchers, engineers, and decision makers who want to have an overview of different parameters and criteria playing a role in the indirect mineralization of CO₂ using industrial waste.

In order to choose the review method, a search on the web using Google was done to find out about different literature review methods. First, overall, 19 websites and articles were reviewed, each one introducing different methods. Totally, 21 literature review methods were identified. Then, the methods were reported in a table with the websites or articles in front of them as a reference. Finally, the first four methods with the most references were chosen to study more about them. Table 2.1 shows the results.

Table 2.1: Methods of literature review

No.	Method	Reference
1	Narrative or traditional reviews	[18] [19] [20] [21] [22] [23] [24] [25] [26] [27] [28] [29] [30]
2	Critically Appraised Topic (CAT)	[18]
3	Scoping reviews	[18] [19] [20] [31] [21] [22] [23] [26] [32] [28] [29] [33] [30]
4	Systematic literature reviews	[18] [20] [31] [21] [22] [34] [23] [35] [24] [25] [26] [27] [32] [28] [29] [33] [30]
5	Annotated bibliographies	[18]
6	Descriptive or Mapping Reviews	[19]
7	Aggregative Reviews	[19]
8	Realist Reviews	[19]
9	Critical Reviews	[19] [31] [21] [23]
10	Argumentative literature review	[20]
11	Integrative literature review	[20] [21]
12	Theoretical literature review	[20] [21] [36]
13	Meta analysis	[31] [21] [22] [25] [26] [28] [30]
14	Rapid review	[31] [21] [22] [24] [29] [33]
15	Umbrella review	[31] [32] [33]
16	Historical review	[21] [35]
17	Methodological review	[21] [35] [36] [26]
18	Cross-Disciplinary Review	[21]
19	Descriptive Review	[21]
20	Conceptual Review	[21]
21	Chronological	[36]

The results show that the *systematic, scoping, narrative, and meta-analysis* have been referred the most among these 19 references.

After identifying the most referred methods, the procedure and purpose of each one was studied. The results (in brief) are as follows.

Systematic literature review: Systematic literature reviews have a straightforward goal. Its main purpose is to present a high-level overview of a specific research subject. This question is specifically tailored to align with the literature review on the subject matter. For instance, a targeted inquiry about clinical or medical results. A systematic literature review will adhere to well-defined protocols that are chosen before the review even begins. This calls for careful planning and a search technique that is tailored to the particular research subject. To ensure that the review is focused and that other researchers can trust it, every detail of a systematic literature review—such as the study methodologies, databases used, and search dates—must be made public [37].

Scoping literature review: Comprehensive literature studies that address broad research concerns are called scoping reviews. Their primary goal is to examine the body of literature and determine its extent within a certain field. Rather than providing a specific response, they present the big picture. They also adhere to a strict, methodical process that needs to be open

2 Literature review

and repeatable. While the goals of scoping reviews can vary, they all aim to identify and map the body of accessible data in a particular field [38].

Narrative literature review: First-year college students typically learn how to conduct a narrative review as a general technique. Finding a few studies that discuss an interesting issue is its goal. There is just a topic of interest in narrative reviews; there is no predefined research question or search method. They lack structure and adhere to no set procedure. There are no guidelines or norms for the review. The reviewers will gain knowledge about the issue, but they won't have a thorough grasp of the current state of the science around it [39].

Meta-analysis literature review: A meta-analysis quantitatively synthesizes studies within a study domain in an impartial manner. To give academics and practitioners more clarity on a subject, meta-analysis statistically evaluates the robustness of findings in a field and finds and resolves conflicting findings in earlier studies. Meta-analysis collects, combines, and analyses empirical evidence using highly technical and complex processes. The method's validity and reliability are ensured by this rigorous approach, which also obscures the process for academics and practitioners who would find meta-analyses useful [40].

With attention to the subject of this study, which has clear questions to be answered and specific variables to consider, the systematic review suits the best.

2.1.2 Conducting the review

The present review follows PRISMA guidelines described in [41] [42] [43].

Table 2.2 shows the criteria and limits of this search.

Table 2.2: Search criteria and limits

Search parameter	Criteria and limit
Database	Scencedirect and Scopus
Year	2020, 2021, 2022, 2023, 2024
Search string	indirect AND (sequestration OR carbonation OR mineralization) AND (CO ₂ OR (carbon AND dioxide))
Search area	Title, abstract, and keywords
Type of literature	Articles (Scopus) and Research Articles (Scencedirect)

The reason for choosing the above-mentioned period of time was explained in the previous sections and the logic behind making the shown keyword and search in titles, abstracts and keywords of papers was to make sure that the most relevant papers using the indirect process to mineralize CO₂ using carbonation could be found. Finally, the search was done excluding review articles to ensure that the practical works in the laboratory or in the industry could be observed.

2 Literature review

The search results from both databases were gathered and processed based on below steps:

1. The title, year, DOI, link, and abstract of each paper was exported to a table.
2. The titles were read and assessed to primarily make sure that the paper is relevant.
3. The papers with obvious irrelevance in their titles were deleted.
4. The relevant results with title, year, DOI, link, and abstract were gathered in a second table.
5. The results (in step 4) from Scopus and Sciencedirect were compared, and the duplicated papers were deleted.
6. The unique papers from step 5 were gathered in a third table and the content of the papers was reviewed.
7. The relevant papers (based on the content) were extracted to a fourth table to be analysed.

Table 2.3 shows the number of papers in each step mentioned above.

Table 2.3: Number of papers in each step

Step	No. of papers
1- primary papers	Scopus: 196- Sciencedirect: 82
4- after title assessment	Scopus: 35- Sciencedirect: 23
6- after deleting duplications	41
7- after review of contents	28

As depicted in Table 2.3, 28 relevant papers were found which were sequestering CO₂ by mineralizing it using carbonation of different industrial wastes. Since two papers were using the same process to describe different outcomes, the total number of papers was considered to be 27.

2.1.3 Analyzing

In this stage, some repeated and common key performance parameters (KPPs) and process settings were identified by reviewing the papers, and the values were extracted. These KPPs or settings and their definitions are shown in Table 2.4.

Table 2.4: Parameters and settings with their definition and units

Parameters & settings	Definition & unit(s)
Extracted metal	The metal extracted from the industrial waste to be carbonated
Source	The industrial waste from which the metal is extracted
Composition	The percentage or concentration of different metals (oxides) in the industrial waste in [%] or [wt%]
Grinding / milling	Availability of grinding or milling for the industrial waste before leaching
Particle size	The particle size of the industrial waste to be used for leaching in [μm] or [mm]
Special process	Any special process before or after leaching to be considered
Leaching reagent and concentration	The used reagent for leaching and its concentration in [mol/L] or [g/L] or [wt%] or [mol/kg]
Solid / liquid	The ratio of the waste solved in the reagent solution in [g/g] or [g/L] or [g/mL] or [mol/mol]
Leaching temperature	The temperature for leaching process in [$^{\circ}\text{C}$]
Leaching pressure	The pressure for leaching process in [bar]
Leaching agitation	The agitation of the leaching solution in [rpm]
Leaching time	The time for leaching process in [min]
Leaching efficiency / capacity	The percentage of extracted metal in leaching process compared to the initial amount in the industrial waste in [%] or the concentration of the extracted metal in the leachate solution in [mol/L] or [mg/L]
Mineralization reactor type	The type of reactor used for the mineralization process
Mineralization agitation	The agitation of the mineralization process in [rpm] or [W]
Mineralization time	The time for mineralization process in [min]
Mineralization gas type, concentration, and flow rate	The type of gas or flue gas used for the mineralization and its concentration in [%] or [mol/L] and its flow rate in [L/min] or [mL/min] or [cm^3/min]
Mineralization temperature	The temperature for mineralization process in [$^{\circ}\text{C}$]
Mineralization pressure	The pressure for mineralization process in [bar] or [MPa]
Mineralization product and its purity	The type of mineralization product and its purity in [%]
Mineralization efficiency/ capacity	The conversion of Ca in the leachate solution in [%] or carbonation efficiency (actual sequestered CO_2 compared to the theoretical possible sequestration amount) in [%] or the amount of products and CO_2 per ton of industrial waste in [kg/ton] or [mol/kg]
Reagent recovery/ recycling	The possibility of recovery or recycling of reagents to repeat the cycle

It is noteworthy to indicate is that studies in literature usually assess and analyse the effect of different parameters and settings on the outcomes to find out the best or optimum results. Therefore, the extracted values in this study for parameters or settings are either the optimum or the best results, not the only results. After extraction, different KPPs and settings were compared to have a better overview and understanding of the processes.

2.1.4 Reporting the review

The extracted values were gathered on a table to have access to all of them in one place. This table is available in appendices 1 to 3. Some of the KPPs and settings in the table show N/A (not available) for some papers. This means that the paper is not providing any information or data for that KPP or setting. In addition, if no temperature or pressure for leaching or mineralization process was provided by the paper, the value was assumed to be ambient temperature or pressure. Finally, the charts were used for a visual understanding of some of the values which are shown in the following pages.

Before describing the charts, it is important to mention that all 27 papers are using either industrial waste or ash from different origins as the source to extract a metal (or metals) for CO₂ mineralization.

Figure 2.1 shows the number of papers per extracted metals for mineralization.

Figure 2.2 depicts the need for grinding of milling of the industrial wastes in the papers.

Figure 2.3 refers to the particle sizes of the industrial waste for leaching.

Figure 2.4 talks about the types of reagents used for leaching. NH₄Cl and HCl were chosen as most representative.

Figure 2.5 reveals the distribution of reagent concentrations among papers.

Figure 2.6 takes T = 45 C as a midpoint and shows the leaching temperatures.

Figure 2.7 depicts different agitation speeds for leaching process.

Figure 2.8 considers 60 minutes as a midpoint and covers the leaching time.

Figure 2.9 shows the mineralization temperatures.

Figure 2.10 talks about the availability of reagent recovery or recycling in the reviewed literature.

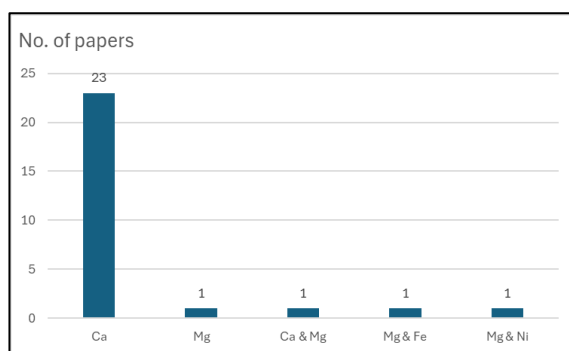


Figure 2.1: Number of papers per each extracted metal

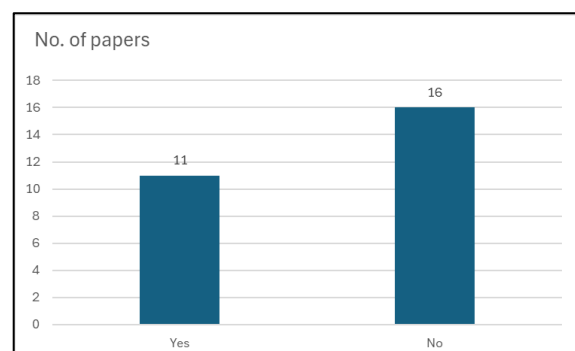


Figure 2.2: No of papers with and without grinding/milling

2 Literature review

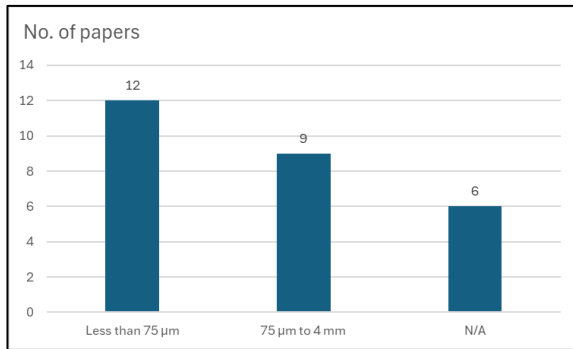


Figure 2.3: Number of papers per particle size of industrial waste

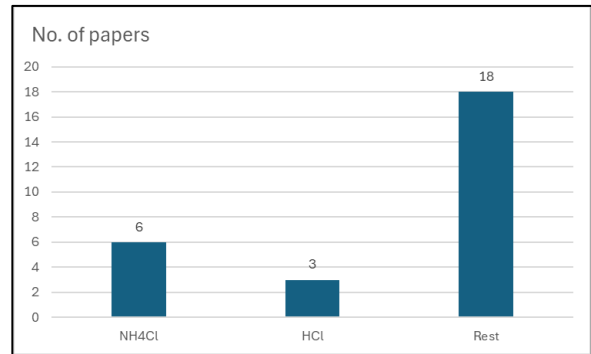


Figure 2.4: Number of papers per type of reagent

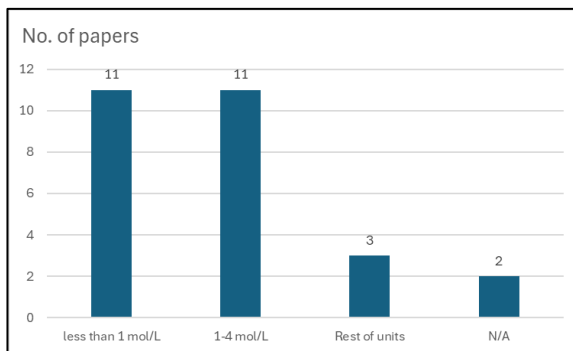


Figure 2.5: Number of papers per concentration of the reagent

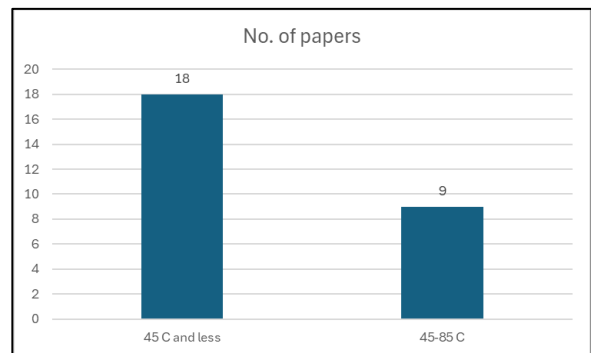


Figure 2.6: Number of papers per leaching temperature

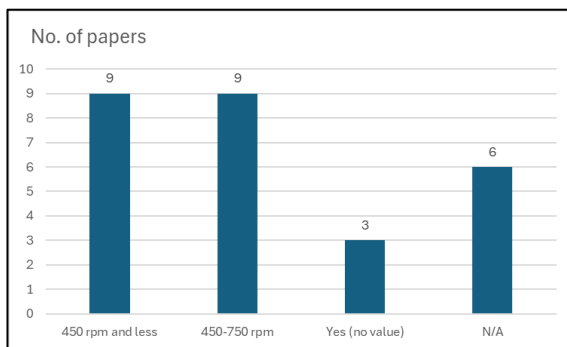


Figure 2.7: Number of papers per different agitation speeds for leaching process

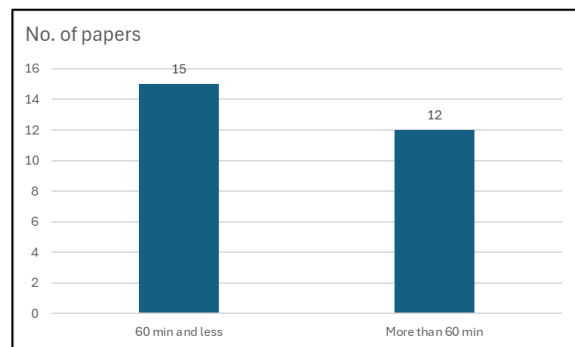


Figure 2.8: Number of papers per leaching time

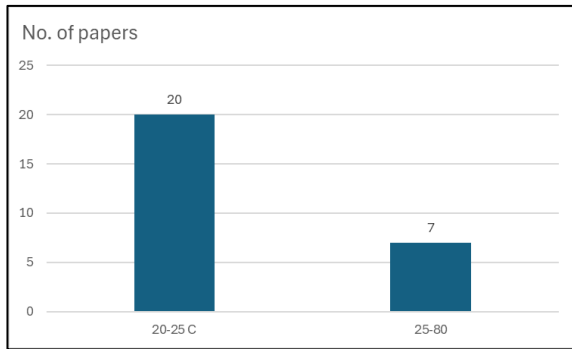


Figure 2.9: Number of papers per mineralization temperature

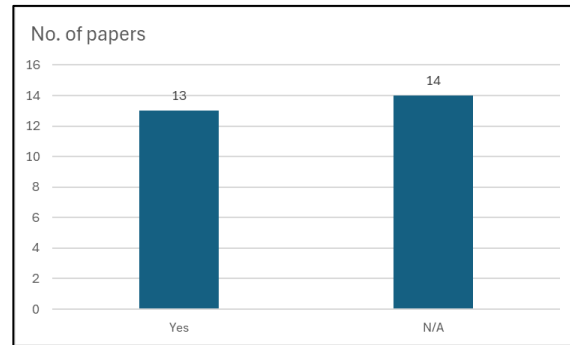


Figure 2.10: Number of papers per availability of reagent recovery or recycling

2.2 Step1: Choosing four processes from literature

With regards to the methodology and Figure 1.8 and having a good overview of the literature, four processes are chosen in the first step to be studied in more detail. The overview of each process is shown in Table 2.5. The reasons to choose these four processes were using different reagents with different concentrations for leaching (extraction) process, using different industrial wastes, and working under different conditions such as temperatures of leaching and mineralization.

Table 2.5: Overview of four chosen processes

Process	Extracted ion	Source	Reagent for leaching (Ca extraction)	Reference
1	Ca	Converter slag (CS)	NH ₄ Cl	[44]
2	Ca	Recycled concrete fines (RCF)	NH ₄ Cl	[45]
3	Ca	Blast furnace slag (BFS)	HCl	[46]
4	Ca	Blast furnace slag (BFS)	CH ₃ COOH	[47]

2.3 Step 2: In detail study of the materials, methods, and results for each process

In the second step, each process was studied deeply to know the materials used, identify the methods implemented, and understand the results from the process. These details are provided in three parts in this report: 1) Waste composition and grinding details, 2) Leaching process details, and 3) Mineralization process details.

2.3.1 Waste composition and pre-treatment details

The details of the percentage of most of the compositions for the industrial wastes and if grinding is included or not, are shown in Table 2.6.

Table 2.6: Composition and pre-treatment of four chosen processes

Process	Source	Composition [%]	Grinding	Reference
1	Converter slag (CS)	CaO: 44.5, Fe ₂ O ₃ : 19.1, MnO: 2.1, Al ₂ O ₃ : 2.3, MgO: 7.6, SiO ₂ : 9.28, P ₂ O ₅ : 1.3	Yes	[44]
2	Recycled concrete fines (RCF)	CaO: 71.11, SiO ₂ : 16.51, Al ₂ O ₃ : 3.28, Fe ₂ O ₃ : 4.19	Yes	[45]
3	Blast furnace slag (BFS)	CaO: 40, SiO ₂ : 29.76, Al ₂ O ₃ : 15.76, Fe ₂ O ₃ : 0.59- MgO: 6.06	No	[46]
4	Blast furnace slag (BFS)	CaO: 40.6, SiO ₂ : 34.1, Fe ₂ O ₃ : 0.901, MgO: 10.7, Al ₂ O ₃ : 9.4, F: 0.07, Cr: 0.003, Ti: 1.03, Mn: 0.376, S: 1.73	No	[47]

2.3.2 Leaching process details

The details of the leaching process such as the reagent type, concentration, agitation, leaching temperature (LT) and pressure (LP), solid to liquid (S/L) ratio, leaching time, and leaching efficiency are shown in Table 2.7. Note that in cases of no data available for the leaching pressure, the pressure has been considered as ambient pressure. The definition of each title has been provided in Table 2.4.

Table 2.7: Leaching process details

Process	Leaching reagent	Reagent concentration [mol/L]	S/L [g/L]	Agitation [rpm]	LT [°C]	LP [bar]	Time [min]	Efficiency [%]
1	NH ₄ Cl	1	63	300	80	1	60	48.1
2	NH ₄ Cl	2	100	500	85	1	60	65.7
3	HCl	4	80	600	80	1	120	91
4	CH ₃ COOH	1.7	16.8	600	25	1	120	100

2.3.3 Mineralization process details

Table 2.8 summarizes the required details of the mineralization process for further calculation for each process. The details are mineralization temperature (MT), pressure (MP), time, and purity of CaCO₃. Noteworthy to indicate is that the process number 4 does not cover the mineralization part and only talks about the leaching process. This process was chosen

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deliberately to calculate the mineralization outcome based on the settings of leaching process and assumptions similar to other three processes and compare the key performance indicators (KPI). Moreover, like leaching process details, if no data for the mineralization pressure is provided, the pressure has been considered as ambient pressure. The definition of each title has been provided in Table 2.4.

Table 2.8: Mineralization process details

Process	MT [°C]	MP [bar]	Time [min]	Purity of CaCO₃ [%]
1	80	1	120	98
2	25	0.25	30	97.8
3	20	1	15	99.5
4	No value (assumed as 25)	No value (assumed as 1)	No value (assumed as 120)	No value

3 Process calculations

In this chapter and following the work procedure in Figure 1.8, the process calculations will be conducted from step 3 to step 8.

3.1 Step 3: Making a PFD to identify main streams for each process

For calculating the mass balances, it is necessary to identify the main streams of a process. To do so, a visual aid like a PFD showing the main units could be helpful. Figure 3.1 shows a PFD of a typical indirect aqueous mineralization process with the main streams.

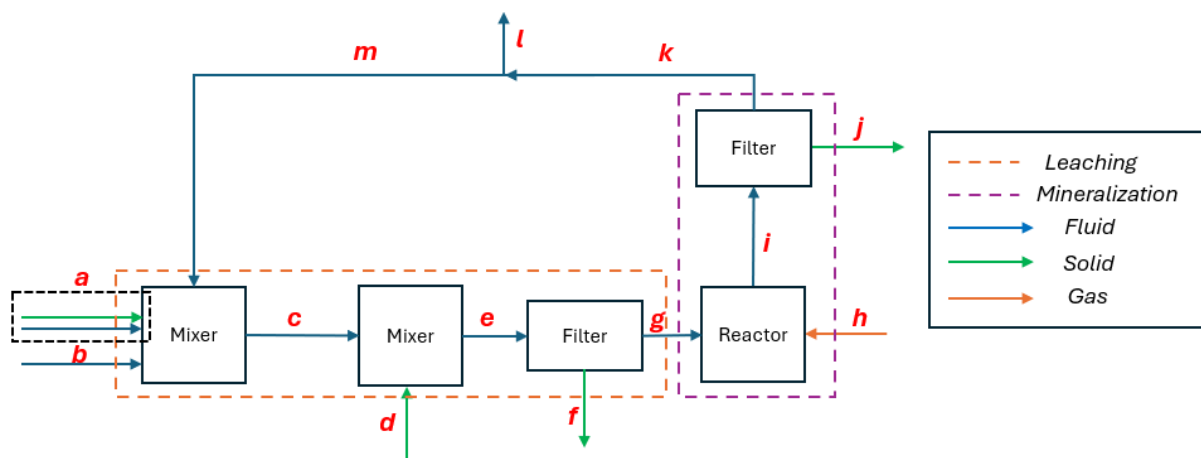


Figure 3.1: PFD of indirect aqueous mineralization process

Definition of the streams in Figure 3.1 are as follows:

- a: Leaching reagent (fluid or solid) make-up to system
- b: Water make-up to system
- c: Leaching solution
- d: Industrial waste
- e: Leachate solution
- f: Solid residues from leachate solution
- g: Filtrate solution rich in Ca
- h: CO₂ introduced to the reaction
- i: Mineralization solution (leaching solution + CaCO₃ + unreacted Ca)
- j: Precipitated CaCO₃
- k: Leaching solution + unreacted Ca
- l: Purge from system
- m: Recovered leaching solution

3.2 Step 4: Conducting scale-up mass balance calculations to simulate each process at large scale

To be able to scale up a process and perform mass balance calculations, it is necessary to conduct the calculations based on the chemical reactions ruling the process. The most important and role-playing reaction in the mineralization process is the sequestration of CO₂ with CaO, which is depicted in R3.1.



Considering reaction R3.1, the procedure for scale-up calculations for a pilot plant with yearly capacity of capturing 400-ton CO₂ (passing through stream h in Figure 3.1) will be explained in the next sections and one set of hand calculations will be conducted for process number 2 as an example. The reason to choose process number 2 is its higher complexity due to different operational temperatures in the leaching and mineralization processes. It should be noted that all of the results are considered to be per annum.

3.2.1 Reagents and product mass calculation

As can be seen in reaction R3.1, 1 mol of CO₂ reacts with 1 mol of CaO which is 1 mol of Ca, and 1 mol of CaCO₃ is produced consequently. In this study, the mass and number of moles of Ca is considered for the calculations since the CaO will turn into Ca and O ions in the leaching process.

To calculate the required amount of Ca, the mass of CO₂ should be turned into the number of moles:

$$\frac{400000 \text{ kg } CO_2}{0.044 \frac{\text{kg}}{\text{mol}}} = 9100000 \text{ mol } CO_2 \quad (3.1)$$

Where 0.044 is showing the molar mass of CO₂.

Hence, the same number of moles as CO₂ is required for Ca which will result in:

$$(9100000 \text{ mol } Ca) \cdot \left(0.04 \frac{\text{kg}}{\text{mol}}\right) = 364000 \text{ kg } Ca \quad (3.2)$$

Where 0.04 is the molar mass of Ca.

Following the same logic, the produced amount of CaCO₃ (passing through stream j in Figure 3.1) will be:

$$(9100000 \text{ mol } Ca) \cdot \left(0.1 \frac{\text{kg}}{\text{mol}}\right) = 910000 \text{ kg } CaCO_3 \quad (3.3)$$

Where 0.1 represents the molar mass of CaCO₃.

3 Process calculations

Considering the non-ideal reactions in a real case field scenario which are unlikely to achieve a conversion of Ca of 100%, and with attention to the need to capture the whole amount of 400-ton CO₂ despite of this fact, an excess amount of Ca should be introduced to CO₂.

Assuming Ca conversion to be 90%, the required mass of Ca will be:

$$(910000 \text{ kg CaCO}_3) \cdot \left(\frac{1}{0.9}\right) \approx 404444 \text{ kg Ca} \quad (3.4)$$

It is important to remember the produced amount of CaCO₃ will not be changed and the non-ideal conversion of Ca (90%) will result in excess Ca ions after the mineralization reaction.

3.2.2 Industrial waste mass calculation

To be able to provide the Ca amount calculated in (3.4), the industrial waste must be leached to extract Ca. The industrial waste used in this process is recycled concrete fines (RCF) which has 71.11% CaO based on Table 2.6 meaning that 1kg of RCF contains 0.7111 kg of CaO. Accordingly, the amount of Ca will be as follows:

$$(0.7111 \text{ kg CaO per kg RCF}) \cdot \left(\frac{0.04 \text{ kg/mol}}{0.056 \text{ kg/mol}}\right) \approx 0.508 \text{ kg Ca per kg RCF} \quad (3.5)$$

Where 0.04 and 0.056 represent Ca and CaO molar masses, respectively.

Considering the leaching (extraction) efficiency (65.7%) shown in Table 2.7, the required amount of RCF (passing through stream d in Figure 3.1) will be:

$$\frac{(404444 \text{ kg Ca})}{0.508 \text{ kg Ca per kg RCF} \times 0.657} \approx 1211967 \text{ kg RCF} \quad (3.6)$$

3.2.3 Leaching solution and leaching reagent mass calculation

Provided by Table 2.6, 100 g of RCF is to be solved in one liter of leaching solution which an aqueous solution with NH₄Cl concentration of 2 mol/L. Hence, the required volume of the leaching solution is going to be:

$$\frac{1211967 \text{ kg RCF}}{0.1} \approx 12119670 \text{ liter} \quad (3.7)$$

Due to comparably lower amount of NH₄Cl solved in the water and unclear effect of it on the volume change, the volume in (3.7) is considered to be the same as the mass of leaching solution (passing through stream c in Figure 3.1) for the upcoming calculations in the next sections.

Consequently, the required mass of NH₄Cl will be calculated as below:

$$12119670 \text{ liter solution} \times 0.054 \frac{\text{kg}}{\text{mol}} \times 2 \text{ mol/L} \approx 1308925 \text{ kg NH}_4\text{Cl} \quad (3.8)$$

Where 0.054 is the molar mass of NH_4Cl .

3.2.4 Rest of mass calculations

The mass of mixture of leaching solution and the industrial waste (leachate solution) after the leaching mixer (passing through stream e in Figure 3.1) will be:

$$12119670 \text{ kg leaching solution} + 1211967 \text{ kg RCF} \approx 13331637 \text{ kg} \quad (3.9)$$

It is assumed by the authors of the paper [45] that the leaching process is highly selective and almost all of the heavy metals except Ca will precipitate in the leaching process in form of solid residues. Therefore, they can be separated from the leaching solution. The mass of solid residues filtered out of the leaching solution (passing through stream f in Figure 3.1) will be:

$$1211967 \text{ kg RCF} - 404444 \text{ kg Ca} \approx 807523 \text{ kg} \quad (3.10)$$

The filtrate solution after filtering which is rich in Ca (passing through stream g in Figure 3.1) will weigh as below:

$$13331637 \text{ kg leachate solution} - 807523 \text{ kg solid residues} \approx 12524114 \text{ kg} \quad (3.11)$$

After introducing CO_2 to the filtrate solution in the reactor, the mineralization solution will consist of recovered leaching solution (3.7), unreacted Ca, and produced CaCO_3 (3.3). The amount of unreacted Ca will be the real extracted amount (3.4) deducted by the amount reacted based on the stoichiometric relation (3.2), which will be:

$$404444 \text{ kg Ca extracted} - 364000 \text{ kg Ca reacted} \approx 40444 \text{ kg} \quad (3.12)$$

So, the mineralization solution (passing through stream i in Figure 3.1) will weigh:

$$12119670 \text{ kg leaching solution} + 40444 \text{ kg Ca unreacted} \\ + 910000 \text{ kg CaCO}_3 \approx 13070114 \text{ kg} \quad (3.13)$$

After filtering the CaCO_3 out of the mineralization solution, the rest (passing through stream k in Figure 3.1) will have below mass:

$$13070114 \text{ kg mineralization solution} - 910000 \text{ kg CaCO}_3 \approx 12160114 \text{ kg} \quad (3.14)$$

Since the filtering units are never 100% ideal and since there will be some heavy metals solved in the solution despite the assumption by the authors, there will be accumulation of heavy metals and CaCO_3 in the process. To tackle this, a purge stream is considered. The accurate amount of this purge to prevent high accumulation can be calculated by measuring the amount of heavy metals and CaCO_3 in the process after filtering, which is not possible now. Hence, an assumed percentage of 10% was considered for this (passing through stream l in Figure 3.1) which will be:

$$12160114 \text{ kg} \times 0.1 \approx 1216011 \text{ kg} \quad (3.15)$$

3 Process calculations

Finally, the recovered leaching solution will turn back to the first stage of the process completing a whole cycle. This recovered leaching solution (passing through stream m in Figure 3.1) will weigh as follows:

$$12160114 \text{ kg} - 1216011 \text{ kg purge} \approx 10944103 \text{ kg} \quad (3.16)$$

There are two losses in this process. One is the loss of NH_4Cl in the purge and by evaporation of ammonia in the process, and the other is the loss of leaching solution which is mostly water.

The mass of lost ammonia by evaporation has not been covered by the authors of the paper but can be measured in the laboratory, which is not possible now. So, an approximate percentage of 5% was assumed for it.

Therefore, the lost amount of NH_4Cl is going to be 15% (10% purge and 5% evaporation) in the process, and the loss of water will be 10% (only purge).

Considering above explanation, the make-up mass of NH_4Cl (passing through stream a in Figure 3.1) and water (passing through stream b in Figure 3.1) will be calculated as below:

$$1308925 \text{ kg } \text{NH}_4\text{Cl} \times 0.15 \approx 196338 \text{ kg } \text{NH}_4\text{Cl} \quad (3.17)$$

$$12119670 \text{ kg water} \times 0.1 \approx 1211967 \text{ kg water} \quad (3.18)$$

3.2.5 Flow rate calculations

After calculating the required masses to capture 400 tons of CO_2 in a year, it is possible to calculate the flow rates. For this process the flow rates are calculated based on the rationale outlined below:

1. Flow rate per second, to enable further energy consumption calculations since it is a metric unit.
2. Flow rate per minute, to be shown on the PFD or P&ID.

The results are illustrated in

Table 3.1.

Table 3.1: Details of flow rates for each stream

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	196338	0.0062	0.4
Water make-up [kg]	b	1211967	0.0384	2.3
Leaching solution [kg]	c	12119670	0.3843	23.1
RCS [kg]	d	1211967	0.0384	2.3
Leachate solution [kg]	e	13331637	0.4227	25.4
Solid residues [kg]	f	807523	0.0256	1.5
Filtrate solution [kg]	g	12524114	0.3971	23.8
CO ₂ [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i	13070114	0.4145	24.9
Precipitated CaCO ₃ [kg]	j	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	12160114	0.3856	23.1
Purge [kg]	l	1216011	0.0386	2.3
Recovered leaching solution [kg]	m	10944103	0.3470	20.8

3.3 Step 5: Making an improved PFD consisting of energy-consuming equipment

With the data calculated in section 3.4 for the main streams and with attention to the preliminary PFD in Figure 3.1, it is possible to draw a more detailed PFD with different equipment and more detailed streams. Figure 3.2 is a suggested improved PFD for process number 2.

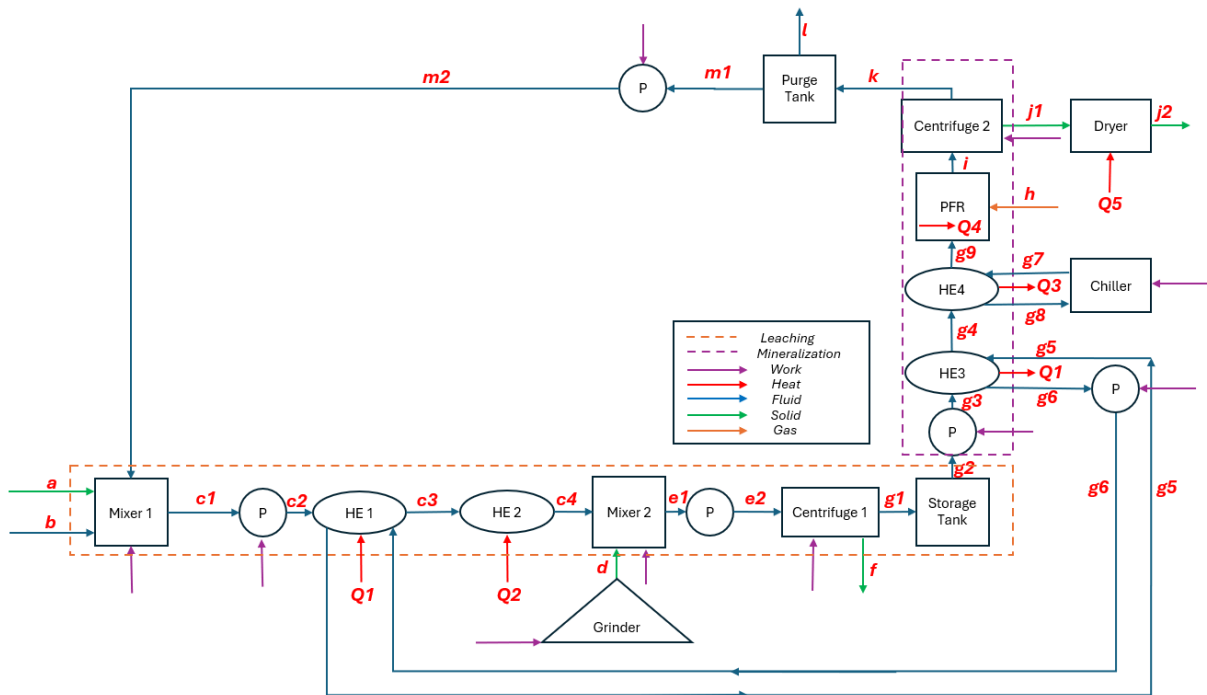


Figure 3.2: Improved PFD for process number 2

3 Process calculations

Figure 3.2 shows a detailed process with more streams and some critical equipment compared to the PFD in Figure 3.1. In this section, the equipment will be introduced, and the detailed calculations of flow rates, equipment sizing, and energy balance calculations will be provided in the next section.

Mixer 1 is the mixing tank where the leaching solution is prepared. This tank is the connecting point of the recovered leaching solution and make up water and NH_4Cl and supplies the net positive suction head (NPSH) for the pump.

Heat Exchanger 1 (HE1) is foreseen to recover the heat in the process. Since the leaching and mineralization processes operate at different temperatures, there is the need to heat and cool the streams. In order to save energy, part of the energy in the cooling can be reused to heat the stream before leaching.

Heat Exchanger 2 (HE2) is to provide the rest of the heat required to increase the stream temperature up to the leaching temperature.

Grinder crushes and grinds the RCF to the required size before being added to the leaching process.

Mixer 2 is the mixing tank where the leaching solution and RCF mix for 60 minutes, and Ca is extracted. This tank supplies the NPSH for the pump too.

Centrifuges in this process are used to separate the solids (residues from leaching process and CaCO_3 from mineralization process) using high centrifugal force.

The **storage tank**'s duty is to accumulate the filtrate solution before the mineralization process to ensure the continuity of the process and reaction and to supply the NPSH for the pump too.

Heat Exchanger 3 (HE3) is coupled with HE1 to transfer the heat extracted from the stream before mineralization to be applied to the stream before leaching.

Heat Exchanger 4 (HE4) is used to extract the rest of the heat that cannot be recovered.

Chiller supplies the cooling water for HE4.

PFR, which stands for Plug Flow Reactor is the reactor where CO_2 is introduced to the reaction with the filtrate solution. Although the papers chosen in this study use batchwise mineralization process, a continuous mineralization process was considered here due to the more efficient nature of PFR in terms of conversion.

The **dryer** evaporates the moisture content of the produced CaCO_3 to increase the quality of the product.

Purge tank is where the recovered leaching solution is accumulated, and the purge happens through this tank. The other duty of the purge tank is to control the continuity of the process and to supply the NPSH for the pump.

Pumps in this process are to ensure and control the calculated flow rates and overcome the head loss in the system.

3.4 Step 6: Conducting detailed mass calculations, sizing the equipment, and energy balance calculations

Using the now improved PFD shown in Figure 3.2, it is possible to calculate the energy consumption and capacity of different equipment in the process and the details of flow rate and temperature of each stream. In section 3.6, these calculations will be covered. The whole energy consumption of the process divides into energy consumption of pumps, agitators, centrifuges, grinder, drying of CaCO_3 , heating, and cooling.

3.4.1 Detailed flow rate calculations

By comparing Figure 3.2 and Figure 3.1, it is visible that the main streams in Figure 3.1 have been broken into more streams in Figure 3.2. This will not affect the mass calculations and flow rates which has already been done in section 3.4. In addition, streams g5, g6, g7, g8 have been added (the heat recovery line and cooling water line) whose flow rates will be calculated in section 3.4.8. Considering above,

Table 3.1 can be updated as Table 3.2.

Table 3.2: Details of flow rates for each stream based on notations in Figure 3.2

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	196338	0.0062	0.4
Water make-up [kg]	b	1211967	0.0384	2.3
Leaching solution [kg]	c1, c2, c3, c4	12119670	0.3843	23.1
RCS [kg]	d	1211967	0.0384	2.3
Leachate solution [kg]	e1, e2	13331637	0.4227	25.4
Solid residues [kg]	f	807523	0.0256	1.5
Filtrate solution [kg]	g1, g2, g3, g4, g9	12524114	0.3971	23.8
CO_2 [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i	13070114	0.4145	24.9
Precipitated CaCO_3 [kg]	j1, j2	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	12160114	0.3856	23.1
Purge [kg]	l	1216011	0.0386	2.3
Recovered leaching solution [kg]	m1, m2	10944103	0.3470	20.8

The details of the streams shown in Table 3.2 such as composition and the temperature will be provided in section 3.4.9.

3.4.2 Sizing of equipment

By knowing the flow rates in the process and time of leaching and mineralization process, the capacity of the equipment such as mixers, purge tank, and PFR can be calculated. As explained in section 3.2.5, the flow rate per 30 minutes is calculated to find the PFR capacity since the

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mineralization needs 30 minutes (considered as reactor residence time) and the flow rate per 60 minutes is calculated to find the capacity of the mixers, storage tank, and purge tank since leaching process requires 60 minutes.

To consider the worst case, the maximum flow rates were chosen from Table 3.2 to size the capacities:

- For flow rate per 30 minutes: 761 kg equal to 761 L equal to 0.761 m³
- For flow rate per 60 minutes: 1522 kg equal to 1522 L equal to 1.522 m³

Hence, the capacity of the PFR will be 761 L and the capacity of the mixers, storage tank, and purge tank will be 1522 L.

Since the leaching process needs more time than mineralization process and is operating batchwise while mineralization is operating continuously, it is not possible to use one set of equipment as shown in Figure 3.2. The actual required number of the mixers, storage tank, and purge tank in the process will be explained in section 4.1.4.

3.4.3 Energy consumption of pumps

The duty of the pumps in the process is to ensure and control the flow rate and continuity of the process and to overcome the head loss in the system. The hydraulic power of a pump can be calculated using equation (3.19).

$$P_h(kW) = \frac{\rho g h q}{3.6 \times 10^6} \quad (3.19)$$

Where:

ρ = flow density [kg/m³]

g = acceleration of gravity [m/s²]

h = head loss of the system [m]

q = flow rate [m³/h]

Considering the small size of the system and since the real-life scale is not clear yet, the head loss of the system is assumed to be 5 m.

The flow density cannot be specified accurately and needs to be measured in the laboratory. But due to the small fraction of solved NH₄Cl and since the solid residues and products are separated shortly after emerging in the solution, the density of the flow is assumed to be that of water at 1000 kg/m³.

The flow rate was calculated to be 1522 kg/h in the stream e1 which equals to 1.522 m³/h considering the density. To calculate the hydraulic power of pump, the maximum flowrate in the system (e1) was used to forecast the worst case.

With above calculation, the power of pump and the yearly energy consumption will be as follows in (3.20) and (3.21):

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$$P_h(kW) = \frac{1000 \frac{kg}{m^3} \times 9.81 \frac{m}{s^2} \times 5 m \times 1.522 m^3/h}{3.6 \times 10^6} \approx 0.0207 kW \quad (3.20)$$

$$P_h(kW) \times 365 \text{ days} \times 24 \text{ hours} = 0.0207 \times 365 \times 24 \approx 182 kWh \quad (3.21)$$

It is important to remember that the energy calculated in (3.21) is the sum of all pumps in the system and will be shared among the real number of pumps when the detailed engineering is being done and the real head loss of each part of the system is calculated.

3.4.4 Energy consumption of agitators in mixers

To calculate the energy consumption of agitators in the process, a table with the agitator power per volume of a reactor and rotational speed of approximately 60-80 rpm was found in the website of a reactor manufacturer [48]. The summary of this table is shown in Table 3.3.

Table 3.3: Agitator motor power per reactor volume

Reactor volume [L]	Motor power [kW]
500	4
1000	5.5
2000	7.5
3000	15
5000	15
10000	18

This table was used to calculate the agitators' energy consumption in the four processes which were studied. It is worth noting that for process number 4 where the required size of reactor is approximately 20000 L, it was considered that two agitators with 18 kW power have been used in a single tank.

With respect to the explanations provided, the agitators' power for process number 2 will be calculated based on the capacity of mixer 1 and mixer 2 which is 1522 L. The corresponding motor power will be 7.5 kW in Table 3.3 (the reactor with 2000 L was chosen in Table 3.3). The yearly energy consumption will be:

$$P(kW) \times 365 \text{ days} \times 24 \text{ hours} = 7.5 \times 365 \times 24 \approx 65700 kWh \quad (3.22)$$

Since there are two agitators (mixer 1 and 2), the total yearly energy consumption will be:

$$65700 \times 2 \approx 131400 kWh \quad (3.23)$$

Although the effect of agitation's rotational speed on leaching and mineralization was one of the variables to study in some of the papers, it was not considered in this study. The reason is that most of the papers used magnetic stirrers in high rotational speeds and they have not

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provided information about the length of the stirrer compared to the diameter of the container of the solution which could give a good idea of the size of the industrial impellers compared to the diameter of the reactor or mixer. Therefore, it was decided to not consider this effect on scale-up calculations of energy consumption.

3.4.5 Energy consumption of centrifuges

A centrifuge is an equipment which can separate different phases using strong centrifugal force. This force is generated by a cylinder with high rotational speed. Two centrifuges have been foreseen in the processes in this study to separate filtrate solution and solid residues originating from the industrial wastes in leaching process and to separate the recovered reagent and CaCO_3 after mineralization process. The average energy consumption of a centrifuge could be considered as 1.5 kWh/m^3 of liquid which is filtered this way [49].

To calculate the total yearly energy consumption, the yearly flow rate must be multiplied by the number provided. Similar to previous energy calculations, the biggest flow rate in the system has been chosen which belongs to stream e1:

$$\text{Decanters energy (kWh)} = \frac{13331637 \text{ L}}{1000 \text{ L/m}^3} \times 2 \times 1.5 \text{ kWh/m}^3 \approx 39995 \text{ kWh} \quad (3.24)$$

3.4.6 Energy consumption of grinding

The wastes as the source of Ca do not have a homogenous size. They can consist of particles as small to be considered as powder and particles as big as gravel. To increase the extraction efficiency of Ca, it is important to leach small particles since this will increase their solution by the reagent.

To achieve this, it is important to grind the waste before the leaching process. To calculate the energy consumption of grinding Bond's equation [44] shown in (3.25) was used:

$$W \text{ (kWh/ton)} = W_i \left(\frac{10}{P_{80}^{0.5}} - \frac{10}{F_{80}^{0.5}} \right) \quad (3.25)$$

Where:

W is energy consumption in [kWh/ton],

W_i is Bond's crushing work index in [kWh/ton],

P_{80} is the final particle size in [μm]. The index 80 means that 80% of the particles have considered to have the final particle size or less.

F_{80} is the primary particle size in [μm]. The index 80 means that 80% of the particles have considered to have the primary particle size or less.

By reviewing the literature, it can be found out that the smaller the particles, the better the extraction efficiency. But due to relatively high energy consumption of grinding equipment, many previous studies have used $150 \mu\text{m}$ for P_{80} .

And $20000 \mu\text{m}$ can be a good average representative primary size [44] of the wastes (F_{80}).

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Moreover, the W_i found to be approximately 18 kWh/ton for the wastes used in this study [50].

So, the energy consumption of grinding will be:

$$\text{Grinding energy (kWh)} = W \text{ (kWh/ton)} \times \text{Ton of ground waste} \quad (3.26)$$

$$\begin{aligned} \text{Grinding energy (kWh)} &= 18 \frac{\text{kWh}}{\text{ton} \left(\frac{10}{150 \mu\text{m}^{0.5}} - \frac{10}{20000 \mu\text{m}^{0.5}} \right)} \times 1211967 \text{ ton} \quad (3.27) \\ &\approx 16270 \text{ kWh} \end{aligned}$$

3.4.7 Energy consumption of drying CaCO₃

In order to have a higher quality of CaCO₃, different parameters play a role. One of these parameters is the moisture content. The less the moisture, the higher the quality. Therefore, a dryer has been foreseen for the process to dry the produced CaCO₃.

The required energy for this process divides into two stages:

1. Increase the temperature of CaCO₃ and its moisture content to 100 °C
2. Add more heat to evaporate the moisture content at the constant temperature (100 °C)

To achieve this, equations (3.28) and (3.29) will be used, the first one to calculate the heat to increase temperatures, and the latter to calculate the heat to evaporate moisture.

$$Q \text{ (kW)} = m \text{ (kg/s)} \times C \left(\frac{\text{kJ}}{\text{kg}^\circ\text{C}} \right) \times (T_2(\text{C}) - T_1(\text{C})) \quad (3.28)$$

$$Q \text{ (kW)} = m \text{ (kg/s)} \times L_v \left(\frac{\text{kJ}}{\text{kg}} \right) \quad (3.29)$$

Where:

m in (3.28) is the mass flow rate of CaCO₃ and the moisture content in [kg/s].

C is the specific heat capacity of CaCO₃ and water in [kJ/kg°C].

T_2 is the evaporation temperature of water in atmospheric conditions in [°C] (considered to be 100 °C).

T_1 is the primary temperature of CaCO₃ and the moisture content before drying, in [°C]. T_1 is 25 C following the assumption that the product is first cooled down to ambient temperature (due to logistic and storage) and then heated to be dried.

m in (3.29) is the flow rate of the moisture content in [kg/s].

L_v is the latent heat of evaporation for water at atmospheric conditions in [kJ/kg].

With the above explanations, there will be two heat calculations with (3.28) for CaCO₃ and moisture (Q_a and Q_b) and one heat calculation with (3.29) for evaporation (Q_c).

The mass flow rate of CaCO₃ is 0.0289 kg/s.

Assuming 10% of moisture content, the moisture content's flow rate is 0.00289 kg/s.

C of water is 4.2 kJ/kg°C.

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C of CaCO_3 is $0.8343 \text{ kJ/kg}^\circ\text{C}$.

L_v is 2260 kJ/kg .

The energy consumption of drying will be as follows:

$$Q_a \text{ (kW)} = 0.0289 \frac{\text{kg}}{\text{s}} \times 0.8343 \text{ kJ/kg}^\circ\text{C} \times (100 \text{ C} - 25 \text{ C}) \approx 1.808 \text{ kW} \quad (3.30)$$

$$Q_b \text{ (kW)} = 0.00289 \frac{\text{kg}}{\text{s}} \times 4.2 \text{ kJ/kg}^\circ\text{C} \times (100 \text{ C} - 25 \text{ C}) \approx 0.91 \text{ kW} \quad (3.31)$$

$$Q_c \text{ (kW)} = 0.00289 \frac{\text{kg}}{\text{s}} \times 2260 \text{ kJ/kg} \approx 6.53 \text{ kW} \quad (3.32)$$

$$\text{Drying energy} = (Q_a + Q_b + Q_c) \times 365 \text{ days} \times 24 \text{ hours} \approx 80907 \text{ kWh} \quad (3.33)$$

3.4.8 Energy consumption of heating and cooling

The leaching process operates at $85 \text{ }^\circ\text{C}$ and the mineralization process operates at $25 \text{ }^\circ\text{C}$ in process number 2.

So, the stream should be cooled down before entering the reactor in g9 and should be heated up again before leaching in c4. Heating up and then cooling down, which means removing the same heat added to the system is costly.

Meanwhile, the mineralization reaction (R3.1) is an exothermic reaction with enthalpy change of -178 kJ/mol meaning that heat is being generated in the reactor. This will help to reduce the heating demand before leaching, but it is still expensive to add and remove heat to and from the system.

As it was explained in section 3.5, a heat recovery loop consisting of two heat exchangers has been used for the system to recover the possible amount of heat. This will reduce the costs.

To calculate the energy consumption of heating and cooling in the system, the temperatures of the lines must be specified. To start calculating the temperatures, it is a good idea to start with calculating the temperature increase in the reactor due to exothermic nature of the reaction.

Using the mass flow rate of produced CaCO_3 and its molar mass, the molar flow rate of CaCO_3 will be calculated as below:

$$\frac{m \left(\frac{\text{kg}}{\text{s}} \right)}{M \left(\frac{\text{kg}}{\text{mol}} \right)} = \frac{0.0289 \text{ kg/s}}{0.1 \text{ kg/mol}} = 0.289 \text{ mol/s} \quad (3.34)$$

Using the molar flow rate and the enthalpy of reaction, the heat produced in the reactor (Q_4) can be calculated:

$$Q_4 \text{ (kW)} = 0.289 \text{ mol/s} \times 178 \text{ kJ/mol} = 51.442 \text{ kW} \quad (3.35)$$

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By knowing the mass flow rate after the reactor (stream i), the heat in (3.35), assuming the specific heat capacity of stream i to be the same as water (since the amount of solids and dissolved reagent is relatively small), and using equation (3.28), the temperature at the outlet of the reactor can be calculated:

$$T_i(C) = T_{g9}(C) + Q(kW)/(m\left(\frac{kg}{s}\right) \times c\left(\frac{kJ}{kgC}\right)) \quad (3.36)$$

$$T_i(C) = 25 C + \frac{51.442 kW}{0.4145 \frac{kg}{s} \times 4.2 kJ/kgC} \approx 55C \quad (3.37)$$

It is recommended to use insulation for the piping, purge tank, mixers 1 and 2, and the storage tank to prevent the heat loss in the system since the generated heat in the reactor can help to reduce the requirement to add external heat to the system and helps for heat recovery.

With attention to (3.37), the temperature of streams i, k, l, m1, m2, c1, and c2 is 55 °C.

The temperatures before HE 1 in c2 and before HE 3 in g3 are known to be 55 °C and 85 °C, respectively. Considering temperature difference of 10 °C for each one to ensure a good heat transfer, the temperature in g5 and g6 (heat recovery line) will be 65 °C and 75 °C, respectively.

By knowing the temperatures in g5 and g6, assuming a flow rate equal to stream i, and using water as the fluid in g5 and g6, the heat recovered (Q_1) in the system can be calculated using (3.28):

$$Q_1 (kW) = 0.4145 kg/s \times 4.2 kJ/kgC \times (75 C - 65 C) \approx 18 kW \quad (3.38)$$

By using the same logic as (3.36), the temperature in c3 (after HE 1) can be calculated:

$$T_{c3}(C) = T_{c2}(C) + Q_1(kW)/(m\left(\frac{kg}{s}\right) \times c\left(\frac{kJ}{kgC}\right)) \quad (3.39)$$

$$T_{c3}(C) = 55 C + \frac{18 kW}{0.4145 \frac{kg}{s} \times 4.2 kJ/kgC} \approx 65C \quad (3.40)$$

In case of lack of heat recovery loop, the temperature in c2 (55 °C) had to be increased to the leaching temperature (85 °C) which is a 30 °C increase. Now, by having the heat recovery loop, the increase will be 20 °C meaning that the requirement of external heating has been approximately reduced by 33.3%.

Now, and by knowing the temperature in c3 and operating temperature of leaching process, the required external heat to increase the temperature can be calculated using equation (3.28). This is shown with Q_2 supplied via HE 2:

$$Q_2 (kW) = 0.4145 kg/s \times 4.2 kJ/kgC \times (85 C - 65 C) \approx 35 kW \quad (3.41)$$

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Using the energy balance relation, the heat that must be removed (Q_3) from the process (by HE 4) to reach the operational temperature of mineralization process can be calculated:

$$Q_{in} - Q_{out} + Q_{generation} = dQ/dt \quad (3.42)$$

$$Q_2 - Q_3 + Q_4 = dQ/dt \quad (3.43)$$

Assuming the process to be in steady state, the term dQ/dt will be zero. Hence:

$$35 \text{ kW} - Q_3 + 51 \text{ kW} = 0 \quad (3.44)$$

$$Q_3 = 86 \text{ kW} \quad (3.45)$$

To remove Q_3 via HE 4, a chiller supplying cooling water (through streams g7 and g8) was chosen. The temperature in g7 was assumed to be 10 °C and the one for g8 to be 20 °C. Knowing these and using equation (3.28), the mass flow rate of cooling water will be:

$$m \text{ (kg/s)} = Q_3 \text{ (kW)} / (C \left(\frac{\text{kJ}}{\text{kgC}} \right) \times (T_{g8}(C) - T_{g7}(C))) \quad (3.46)$$

$$m \text{ (kg/s)} = \frac{86 \text{ kW}}{4.2 \frac{\text{kJ}}{\text{kgC}} \times (20 \text{ C} - 10 \text{ C})} \simeq 2 \text{ kg/s} \quad (3.47)$$

To remove the Q_3 , the chiller does work. This work can be calculated using the definition of Coefficient of Performance (COP) for refrigeration systems:

$$COP = Q_c/W \quad (3.48)$$

Where:

Q_c is the heat removed in [kW] which is Q_3 , and

W is the work in [kW]

Assuming a regular compression chiller with average COP of 5 [51], the work will be:

$$W \text{ (kW)} = 86 \text{ kW} / 5 \simeq 17 \text{ kW} \quad (3.49)$$

Finally, considering (3.41) and (3.49), the yearly energy consumption to heat and cool the process will be:

$$Q_2 \text{ (kW)} \times 365 \text{ days} \times 24 \text{ hours} = 35 \times 365 \times 24 \simeq 304969 \text{ kWh} \quad (3.50)$$

$$W \text{ (kW)} \times 365 \text{ days} \times 24 \text{ hours} = 17 \times 365 \times 24 \simeq 150983 \text{ kWh} \quad (3.51)$$

The methodology which was used in section 3.6.8 is applicable for processes number 2 and 3 where the operating temperature of the leaching and mineralization processes are different. For processes number 1 and 4 where the operating temperature of the leaching and mineralization processes is the same, the heat recovery system does not exist and only the generated heat in

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the reactor and the cooling demand are calculated using the same equations as in 3.6.8. It is important to indicate that to calculate the cooling demand, the lost heat from purge tank and mixer 1 due to heat transfer (which is beneficial for process 1 and 4) was calculated and the procedure is explained in section 4.2.4.

3.4.9 Summary of the mass flow rates, temperatures, and energy consumption calculations

In this section, an overview of the mass and energy consumption calculations is provided. The details of each stream are as follows and Table 3.4 shows the details of energy consumption and the total yearly amount of it.

a: NH_4Cl make up at 0.6 kg/min

b: Water make up at 2.3 kg/min

c1: aqueous solution of NH_4Cl (2mol/L) at 23.1 kg/min at $T = 55\text{ }^\circ\text{C}$

c2: aqueous solution of NH_4Cl (2mol/L) at 23.1 kg/min at $T = 55\text{ }^\circ\text{C}$

c3: aqueous solution of NH_4Cl (2mol/L) at 23.1 kg/min at $T = 65\text{ }^\circ\text{C}$

c4: aqueous solution of NH_4Cl (2 mol/L) at 23.1 kg/min at $T = 85\text{ }^\circ\text{C}$

d: RCF at 2.3 kg/min

e1: Leachate solution at 25.4 kg/min at $T = 85\text{ }^\circ\text{C}$

e2: Leachate solution at 25.4 kg/min at $T = 85\text{ }^\circ\text{C}$

f: Solid residues at 1.5 kg/min

g1: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85\text{ }^\circ\text{C}$

g2: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85\text{ }^\circ\text{C}$

g3: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85\text{ }^\circ\text{C}$

g4: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 75\text{ }^\circ\text{C}$

g5: Heat recovery line (water) at 24.9 kg/min at $T = 65\text{ }^\circ\text{C}$

g6: Heat recovery line (water) at 24.9 kg/min at $T = 75\text{ }^\circ\text{C}$

g7: Inlet cooling water at 123 kg/min at $T = 10\text{ }^\circ\text{C}$

g8: Outlet cooling water at 123 kg/min at $T = 20\text{ }^\circ\text{C}$

g9: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 25\text{ }^\circ\text{C}$

h: CO_2 at 0.8 kg/min

i: Solution after mineralization (NH_4Cl solution + CaCO_3 + unreacted Ca) at 24.9 kg/min at $T = 55\text{ }^\circ\text{C}$

j1: Solid CaCO_3 at 1.7 kg/min

k: Recovered reagent (NH_4Cl solution + unreacted Ca) at 23.1 kg/min at $T = 55\text{ }^\circ\text{C}$

l: Purge at 2.3 kg/min (10%) at $T = 55\text{ }^\circ\text{C}$

m1: Return flow (NH_4Cl solution) at 20.8 kg/min at $T = 55\text{ }^\circ\text{C}$

m2: Return flow (NH_4Cl solution) at 20.8 kg/min at $T = 55\text{ }^\circ\text{C}$

j2: Dried CaCO_3 at 1.7 kg/min

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Table 3.4: Details of yearly energy consumption and the total amount

Pumps [kWh]	Agitators [kWh]	Centrifuges [kWh]	Grinding [kWh]	Drying CaCO ₃ [kWh]	Heating [kWh]	Cooling [kWh]	Total [kWh]
182	131400	39995	16270	80907	304969	150983	724706

3.5 Step 7: Conducting economic calculations

The energy consumption, NH₄Cl, and the process water have costs and the produced CaCO₃ can be sold for revenue. In step seven, different scenarios are to be assumed and the corresponding yearly costs and revenues will be calculated.

It is of high importance to indicate that the costs calculated in this study are a part of the operational expenditures (OPEX) and the capital expenditure (CAPEX) has not been calculated. The reason is the assumption that the CAPEX is much higher than the OPEX. This is because there will be multiple number of tanks and mixers to ensure the continues nature of mineralization process which is in contrast with the batchwise nature of leaching process. Also, there will be auxiliary pumps in the process to be substituted in case of any failure in one of them. And finally, there will be several controlling instruments. All of this will result in much more CAPEX than OPEX. Meanwhile, the lack of time to be spent on detailed and extensive economic calculations is also another reason to neglect the CAPEX.

3.5.1 Energy cost calculation

The energy cost was decided to be calculated in Middle East with progressive countries where CO₂ production is comparably high in electricity generation, Norway as the representative for a progressed European country where CO₂ production is comparably low in electricity generation, and the United States (US) where CO₂ production is comparably moderate in electricity generation. The reason was to consider different countries to install the pilot plant and assess the impact of energy price on the economy of the processes.

The energy carrier in this study was assumed to be electricity and the average industrial price of electricity in the above-mentioned countries was found by searching online databases.

The representative electricity price for Middle East was found to be 47.96 USD/MWh [52]. Norway has a rate of 42.63 USD/MWh [53], and this price is 79 USD/MWh for the US [54].

Using the total energy consumption in Table 3.4, the energy costs will be calculated as below:

$$\begin{aligned}
 & \text{Total energy consumption}(kWh) \times \text{Electricity price} \left(\frac{USD}{kWh} \right) \\
 & = \text{Energy cost (USD)}
 \end{aligned} \tag{3.52}$$

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$$724706 \text{ kWh} \times 47.96 \frac{\text{USD}}{\text{kWh}} \text{ in Saudi Arabia} \approx 34757 \text{ USD in Middle East} \quad (3.53)$$

$$724706 \text{ kWh} \times 42.63 \frac{\text{USD}}{\text{kWh}} \text{ in Norway} \approx 30894 \text{ USD in Norway} \quad (3.54)$$

$$724706 \text{ kWh} \times 79 \frac{\text{USD}}{\text{kWh}} \text{ in the US} \approx 57252 \text{ USD in the US} \quad (3.55)$$

3.5.2 Material cost calculation

Due to the purge considered in the system and because of loss of NH_4Cl in the process due to evaporation, the system is continuously losing NH_4Cl and water. Hence, there must be a continuous make-up of both added into the system. The streams a and b show this make-up.

Similar to what was done in 3.7.1, the supplying origin of NH_4Cl was to be different areas. In this case, East Asia, US, and European Union (EU) were chosen to supply NH_4Cl from them.

It is important to say that the costs of shipping and transportation from these origins to the plant have not been calculated due to lack of knowledge about the number of shipments, weight of each shipment, and their time.

The NH_4Cl price is 0.08 USD/kg, 0.27 USD/kg, and 0.56 USD/kg in East Asia, US, and EU respectively [55].

Using the required mass of NH_4Cl per year (stream a) in Table 3.2, the costs considering supplying from the above-mentioned origins will be calculated as follows:

$$\text{Yearly mass of } \text{NH}_4\text{Cl} \text{ (kg)} \times \text{NH}_4\text{Cl price} \left(\frac{\text{USD}}{\text{kg}} \right) = \text{NH}_4\text{Cl cost (USD)} \quad (3.56)$$

$$196338 \text{ kg} \times 0.08 \frac{\text{USD}}{\text{kg}} \text{ in East Asia} \approx 15707 \text{ USD} \quad (3.57)$$

$$196338 \text{ kg} \times 0.27 \frac{\text{USD}}{\text{kg}} \text{ in the US} \approx 53011 \text{ USD} \quad (3.58)$$

$$196338 \text{ kg} \times 0.56 \frac{\text{USD}}{\text{kg}} \text{ in the US} \approx 109950 \text{ USD} \quad (3.59)$$

On the other hand, the make-up water known as the process water also has its own price. This price found to be approximately 0.00755 USD/kg [56]. By knowing the required yearly mass of the process water in Table 3.2 (stream b), the cost of process water can be calculated as below:

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$$\text{Yearly mass of process water (kg)} \times \text{Process water price} \left(\frac{\text{USD}}{\text{kg}} \right) = \quad (3.60)$$

Process water cost (USD)

$$1211967 \text{ kg} \times 0.00755 \frac{\text{USD}}{\text{kg}} \approx 9150 \text{ USD} \quad (3.61)$$

3.5.3 Calculation of revenue from sales of CaCO₃

The CaCO₃ produced in the process has numerous uses in different industries. Therefore, it could be sold as a product which can either at least moderate the operational expenses or even turn the process into a beneficial one. The average price of CaCO₃ in the literature [56] was found to be approximately 0.31 USD/kg. Therefore, the yearly revenue from sales of this product will be:

$$\text{Yearly mass of CaCO}_3 \text{ (kg)} \times \text{CaCO}_3 \text{ price} \left(\frac{\text{USD}}{\text{kg}} \right) \quad (3.62)$$

= CaCO₃ sales revenue(USD)

$$910000 \text{ kg} \times 0.31 \frac{\text{USD}}{\text{kg}} \approx 282100 \text{ USD} \quad (3.63)$$

3.5.4 Calculation of revenue from sales of captured CO₂

Assuming the plant to be based in the US and considering the Section 45Q of the U.S. Internal Revenue Code, a tax credit is provided for each ton of the captured CO₂. If the CO₂ is captured in a geologic formation, the benefit from each ton will be 85 USD/ton and if it is used for Enhanced Oil Recovery (EOR), this amount will be 60 USD/ton [57].

Although the captured CO₂ in this study turns into a thermodynamically stable mineral which can be found in geologic formations, the CO₂ has not been captured exactly in the formation. And it cannot be used for EOR as well. Hence, to assume the worst scenario, it was considered that the captured CO₂ will bring 60 USD/ton of benefit.

The whole yearly revenue from this will be calculated as follows:

$$\text{Yearly mass of CO}_2 \text{ (ton)} \times \text{CO}_2 \text{ price} \left(\frac{\text{USD}}{\text{ton}} \right) = \text{CO}_2 \text{ sales revenue(USD)} \quad (3.64)$$

$$400 \text{ ton} \times 60 \frac{\text{USD}}{\text{ton}} \approx 24000 \text{ USD} \quad (3.65)$$

3.5.5 Economic assessment of the process

Based on the calculations in sections 3.5.1, 3.5.2, 3.5.3, and 3.5.4, the potential revenue from the process and a part of OPEX have been calculated. Therefore, it is possible to estimate the profitability of the process in the mentioned three countries considering the supply of the materials from different origins. The general method to estimate the profit is shown below in (3.66). If the result is positive, the process is profitable, if negative, it will be non-profitable.

$$\text{Process profit} = \text{Revenue of CaCO}_3 + \text{Revenue of CO}_2 - \text{Energy cost} - \text{Materials' cost} \quad (3.66)$$

Table 3.5, Table 3.6, and Table 3.7 show the results for Middle East, Norway, and US, respectively.

Table 3.5: Economic assessment of the process in Middle East

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-34757	-34757	-34757
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-59614	-96919	-153857
Yearly profit (sum of total revenue and total cost) [USD]	222486	185181	128243

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Table 3.6: Economic assessment of the process in Norway

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-30894	-30894	-30894
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-55752	-93056	-149994
Yearly profit (sum of total revenue and total cost) [USD]	226348	189044	132106

Table 3.7: Economic assessment of the process in the US

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	24000	24000	24000
Total revenue [USD]	306100	306100	306100
Energy cost [USD]	-57252	-57252	-57252
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-82109	-119414	-176352
Yearly profit (sum of total revenue and total cost) [USD]	223991	186686	129748

3.6 Step 8: Defining and calculating Key Performance Indicators (KPIs)

KPIs can have numerous benefits and help with continuous improvement, progress measurement, prioritizing, decision making, benchmarking and comparison. In order to evaluate the performance of four processes and compare them, some KPIs were defined and calculated in this study. These KPIs and their definitions are as follows:

- **KPI1:** Mass of captured CO₂ per mass of industrial waste in [kg/kg]
- **KPI2:** Mass of produced CaCO₃ per mass of industrial waste in [kg/kg]
- **KPI3:** Mass of make-up reagent (NH₄Cl in process 2) per mass of captured CO₂ in [kg/kg]
- **KPI4:** Mass of make-up reagent (NH₄Cl in process 2) per mass of produced CaCO₃ in [kg/kg]
- **KPI5:** Energy consumption per mass of captured CO₂ in [kWh/kg]
- **KPI6:** Energy consumption per mass of produced CaCO₃ in [kWh/kg]
- **KPI7:** Mass of unutilized industrial waste (solid residues filtered out of the process) per mass of primary added industrial waste (the industrial waste added to the process to supply Ca) in [kg/kg]
- **KPI8:** Mass of produced CO₂ by the process (due to energy consumption) in Middle East (KPI8-1), in Norway (KPI8-2), and in the US (KPI8-3) per mass of captured CO₂ by the process in [kg/kg]
- **KPI9:** Cost of reagent (NH₄Cl in process 2) from East Asia per mass of captured CO₂ in [USD/kg]
- **KPI10:** Cost of reagent (NH₄Cl in process 2) from the US per mass of captured CO₂ in [USD/kg]
- **KPI11:** Cost of reagent (NH₄Cl in process 2) from the EU per mass of captured CO₂ in [USD/kg]
- **KPI12:** Total costs in Middle East considering the reagent (NH₄Cl in process 2) from East Asia per mass of captured CO₂ in [USD/kg]
- **KPI13:** Total costs in Middle East considering the reagent (NH₄Cl in process 2) from the US per mass of captured CO₂ in [USD/kg]
- **KPI14:** Total costs in Middle East considering the reagent (NH₄Cl in process 2) from the EU per mass of captured CO₂ in [USD/kg]
- **KPI15:** Total costs in Norway considering the reagent (NH₄Cl in process 2) from East Asia per mass of captured CO₂ in [USD/kg]
- **KPI16:** Total costs in Norway considering the reagent (NH₄Cl in process 2) from the US per mass of captured CO₂ in [USD/kg]
- **KPI17:** Total costs in Norway considering the reagent (NH₄Cl in process 2) from the EU per mass of captured CO₂ in [USD/kg]

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- **KPI18:** Total costs in the US considering the reagent (NH₄Cl in process 2) from East Asia per mass of captured CO₂ in [USD/kg]
- **KPI19:** Total costs in the US considering the reagent (NH₄Cl in process 2) from the US per mass of captured CO₂ in [USD/kg]
- **KPI20:** Total costs in the US considering the reagent (NH₄Cl in process 2) from the EU per mass of captured CO₂ in [USD/kg]
- **KPI21:** CaCO₃ revenue per mass of captured CO₂ in [USD/kg]
- **KPI22:** Yearly profit in Middle East with regards to the origin of supplying the reagent per mass of captured CO₂ in [USD/kg]
- **KPI23:** Yearly profit in Norway with regards to the origin of supplying the reagent per mass of captured CO₂ in [USD/kg]
- **KPI24:** Yearly profit in the US with regards to the origin of supplying the reagent per mass of captured CO₂ in [USD/kg]

3.6.1 KPIs calculations

In this section, the defined KPIs are calculated as follows:

$$KPI1: \frac{400000 \text{ kg } CO_2}{1211967 \text{ kg Waste}} = 0.33 \text{ kg/kg} \quad (3.67)$$

$$KPI2: \frac{910000 \text{ kg } CO_2}{1211967 \text{ kg Waste}} = 0.751 \text{ kg/kg} \quad (3.68)$$

$$KPI3: \frac{196338 \text{ kg } NH_4Cl}{400000 \text{ kg } CO_2} = 0.491 \text{ kg/kg} \quad (3.69)$$

$$KPI4: \frac{196338 \text{ kg } NH_4Cl}{910000 \text{ kg } CaCO_3} = 0.216 \text{ kg/kg} \quad (3.70)$$

$$KPI5: \frac{724706 \text{ kWh}}{400000 \text{ kg } CO_2} = 1.812 \text{ kWh/kg} \quad (3.71)$$

$$KPI6: \frac{724706 \text{ kWh}}{910000 \text{ kg } CaCO_3} = 0.796 \text{ kWh/kg} \quad (3.72)$$

$$KPI7: \frac{807523 \text{ kg Unutilized waste}}{1211967 \text{ kg Utilized Waste}} = 0.666 \text{ kg/kg} \quad (3.73)$$

For KPI8 to be calculated, the energy, which is assumed to be carried by electricity, must be multiplied by the amount of produced CO₂ while generating electricity. This amount was found

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to be 0.558 kg/kWh in Middle East, 0.029 kg/kWh in Norway, and 0.368 kg/kWh in the US [58].

$$KPI8 - 1: \frac{\left(\text{Energy consumption (kWh)} \times \text{CO}_2 \text{ production} \left(\frac{\text{kg}}{\text{kWh}} \right) \right)}{\text{CO}_2 \text{ captured (kg)}} \quad (3.74)$$

$$= \frac{724706 \text{ kWh} \times 0.558 \frac{\text{kg}}{\text{kWh}}}{400000 \text{ kg CO}_2 \text{ captured}} = \frac{404386 \text{ kg CO}_2 \text{ produced}}{400000 \text{ kg CO}_2 \text{ captured}} \\ = 1.011 \text{ kg/kg}$$

$$KPI8 - 2: \frac{724706 \text{ kWh} \times 0.029 \frac{\text{kg}}{\text{kWh}}}{400000 \text{ kg CO}_2 \text{ captured}} = \frac{21016 \text{ kg CO}_2 \text{ produced}}{400000 \text{ kg CO}_2 \text{ captured}} \quad (3.75) \\ = 0.053 \text{ kg/kg}$$

$$KPI8 - 3: \frac{724706 \text{ kWh} \times 0.368 \frac{\text{kg}}{\text{kWh}}}{400000 \text{ kg CO}_2 \text{ captured}} = \frac{266692 \text{ kg CO}_2 \text{ produced}}{400000 \text{ kg CO}_2 \text{ captured}} \quad (3.76) \\ = 0.667 \text{ kg/kg}$$

$$KPI9: \frac{15707 \text{ USD}}{400000 \text{ kg CO}_2} = 0.039 \text{ USD/kg} \quad (3.77)$$

$$KPI10: \frac{53011 \text{ USD}}{400000 \text{ kg CO}_2} = 0.133 \text{ USD/kg} \quad (3.78)$$

$$KPI11: \frac{109950 \text{ USD}}{400000 \text{ kg CO}_2} = 0.275 \text{ USD/kg} \quad (3.79)$$

$$KPI12: \frac{59614 \text{ USD}}{400000 \text{ kg CO}_2} = 0.149 \text{ USD/kg} \quad (3.80)$$

$$KPI13: \frac{96919 \text{ USD}}{400000 \text{ kg CO}_2} = 0.242 \text{ USD/kg} \quad (3.81)$$

$$KPI14: \frac{153857 \text{ USD}}{400000 \text{ kg CO}_2} = 0.385 \text{ USD/kg} \quad (3.82)$$

$$KPI15: \frac{55752 \text{ USD}}{400000 \text{ kg CO}_2} = 0.139 \text{ USD/kg} \quad (3.83)$$

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$$KPI16: \frac{93056 \text{ USD}}{400000 \text{ kg CO}_2} = 0.233 \text{ USD/kg} \quad (3.84)$$

$$KPI17: \frac{149994 \text{ USD}}{400000 \text{ kg CO}_2} = 0.375 \text{ USD/kg} \quad (3.85)$$

$$KPI18: \frac{82109 \text{ USD}}{400000 \text{ kg CO}_2} = 0.205 \text{ USD/kg} \quad (3.86)$$

$$KPI19: \frac{119414 \text{ USD}}{400000 \text{ kg CO}_2} = 0.299 \text{ USD/kg} \quad (3.87)$$

$$KPI20: \frac{176352 \text{ USD}}{400000 \text{ kg CO}_2} = 0.441 \text{ USD/kg} \quad (3.88)$$

$$KPI21: \frac{282100 \text{ USD}}{400000 \text{ kg CO}_2} = 0.705 \text{ USD/kg} \quad (3.89)$$

The KPIs 22, 23, and 24 are shown in Table 3.8 below.

Table 3.8: KPI22, KPI23, and KPI24

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
KPI22: Yearly profit in Middle East per mass of captured CO ₂ in [USD/kg]	0.56	0.46	0.11
KPI23: Yearly profit in Norway per mass of captured CO ₂ in [USD/kg]	0.57	0.47	0.11
KPI24: Yearly profit in the US per mass of captured CO ₂ in [USD/kg]	0.56	0.46	0.11

4 Results and discussion

In this chapter, the improved PFDs and the results of the calculations based on the discussed method in chapter 3 for all 4 processes in Table 2.5 will be presented, discussed, and compared with each other. The purpose of this work is to find the advantages and disadvantages of each process in technical, economical, and environmental terms.

4.1 Improved PFD for process number 1, 2, 3, and 4

Figure 4.1, Figure 4.2, Figure 4.3, and Figure 4.4 show the improved PFD for processes number 1, 2, 3, and 4, respectively.

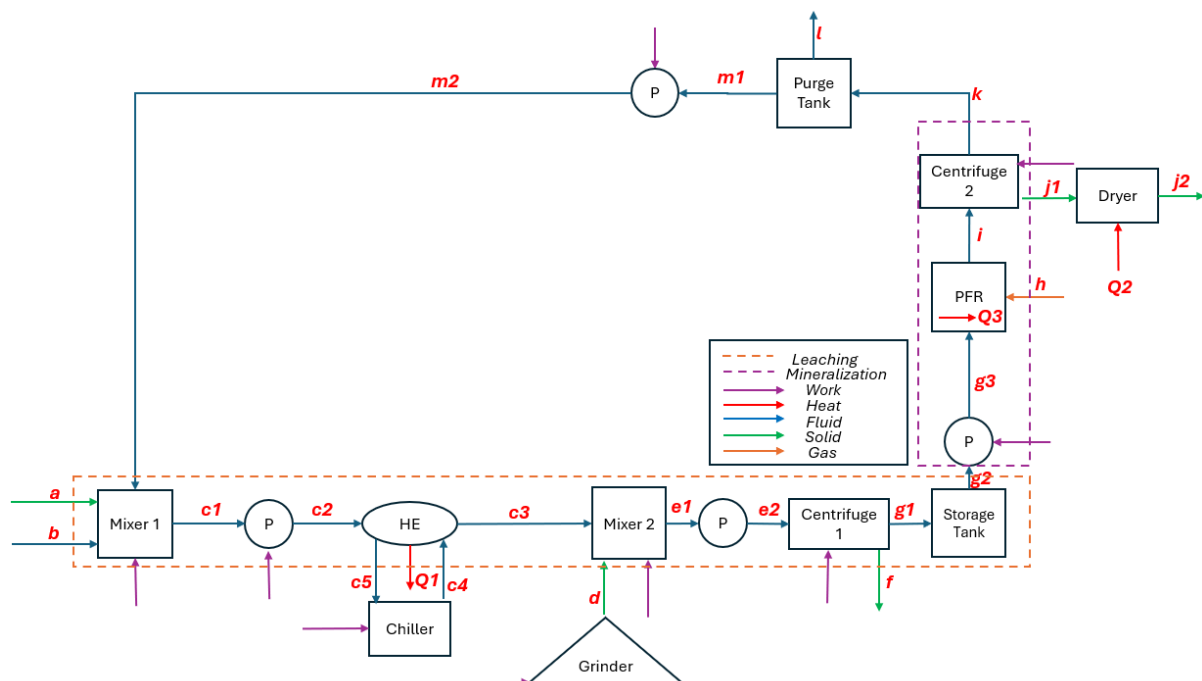


Figure 4.1: The improved PFD for process number 1

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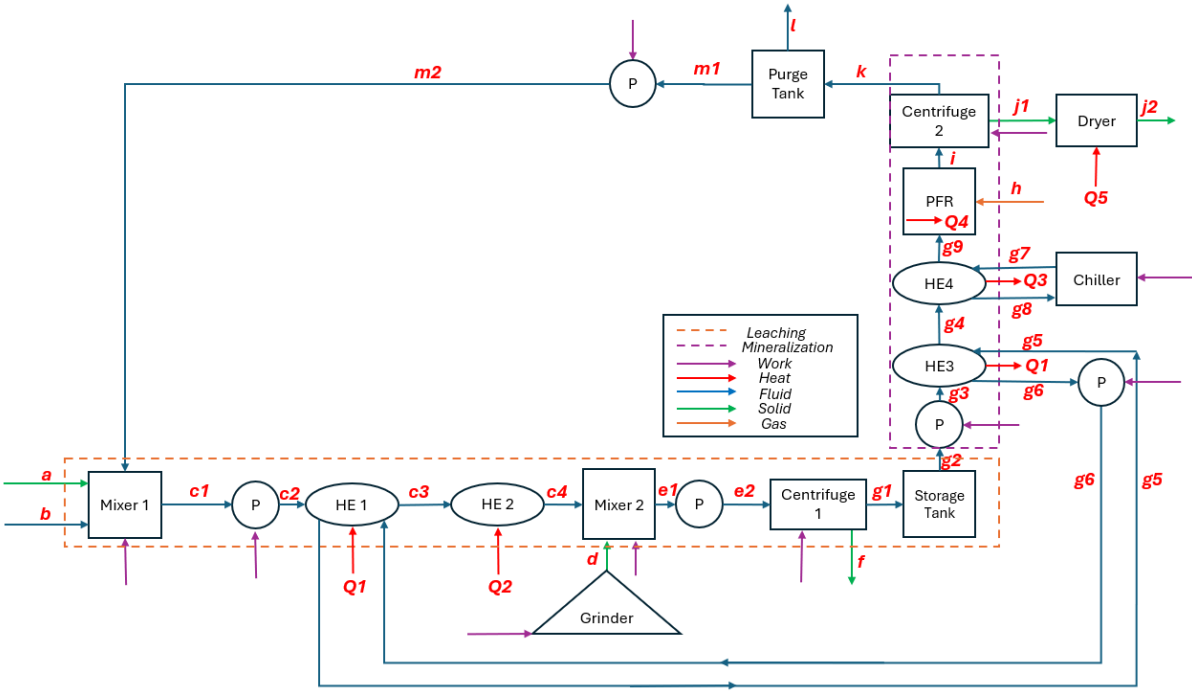


Figure 4.2: The improved PFD for process number 2

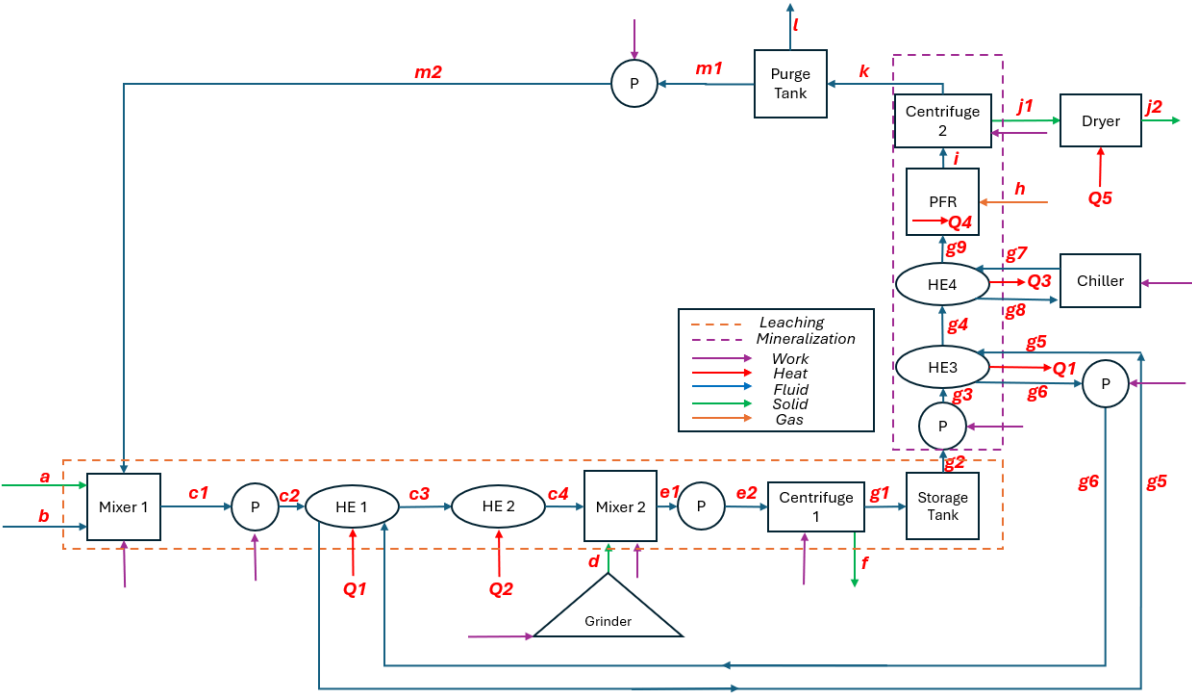


Figure 4.3: The improved PFD for process number 3

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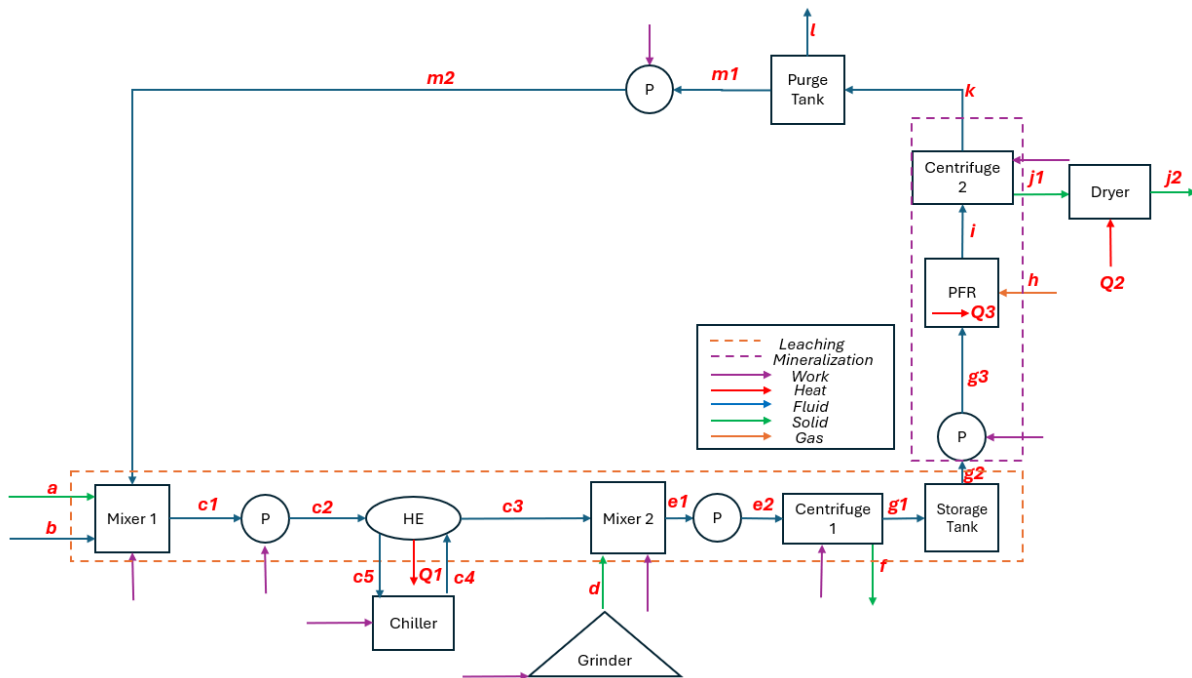


Figure 4.4: The improved PFD for process number 4

As can be seen from Figure 4.1 to Figure 4.4, the four processes are similar to each other with some minor differences. In Figure 4.1 and Figure 4.4, the water and reagent are mixed in mixer 1 making the leaching solution and then pumped through the heat exchanger (HE) where the stream is cooled down to the leaching and mineralization temperature. The leaching and mineralization temperatures are the same in process 1 and 4. After that, the industrial waste is added to the leaching solution in mixer 2 where the leaching process happens in the leaching time. After leaching, the leachate is pumped into the centrifuge 1 where the solid residues are separated, and the filtrate solution (rich in Ca) accumulates in the storage tank. Then the filtrate solution is pumped to the plug flow reactor (PFR) where CO_2 is introduced to be mineralized, generating heat due to exothermic nature of the reaction. After the PFR, the mineralization solution, containing CaCO_3 and unreacted Ca, passes through the centrifuge 2 where the precipitated CaCO_3 is separated from the stream. The recovered reagent, then, accumulates in the purge tank and a part of it is purged out of the process to prevent accumulation of heavy metals and undesired materials. Finally, the recovered reagent is pumped in the mixer 1 to repeat the cycle.

The process is the same in Figure 4.2 and Figure 4.3. The only differences are the number of heat exchangers, where the chiller is located, and the heat recovery line which are due to the different operational temperature of leaching and mineralization in two of these four processes.

The nature of the equipment used in the processes are the same since the main purpose of the processes is the same. There are mixers to mix the reagent with water and leaching solution with the industrial waste. There are centrifuges to separate the solid phase from liquid phase. There are reactors for the reaction, purge tanks and storage tanks to purge and supply the NPSH of the pumps. There are heat exchangers to transfer the heat and chillers to cool down the

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stream. There are grinders to grind the industrial waste to the desired size and dryer to remove the moisture from the produced CaCO_3 . And finally, there are pumps to run the process.

The PFDs of processes 1 and 4 are the same and the PFDs of processes 2 and 3 are the same.

In processes 1 and 4, the leaching and mineralization processes have the same temperature (80 °C and 25 °C, respectively). Hence, the produced heat in the reactor will increase the temperature after mineralization. This extra heat needs to be extracted out of the process before leaching. So, a heat exchanger is foreseen before mixer 2 to cool down the stream's temperature to the operational temperature of both leaching and mineralization. To achieve this, a chiller is designed to supply the cooling water.

In processes 2 and 3, the operational temperatures of leaching and mineralization are different. For process 2, the leaching temperature and mineralization temperature are 85 °C and 25 °C, respectively and for process 3 these are 80 °C and 20 °C respectively. This means that the temperature of the stream should be reduced to the operation temperature of mineralization before the reactor. So, there is a need to have a heat exchanger before the reactor to do this duty. Meanwhile, although the generated heat in the reactor heats up the stream after it, the stream must be heated up much more before leaching to reach the operational temperature of leaching. A part of the heat which must be extracted before mineralization can be recovered to increase the temperature of the stream before leaching. Therefore, a heat recovery line with two heat exchangers (HE 1 and HE 3) are designed for this purpose. The rest of the heat to be added to and to be extracted from the process will be transferred through heat exchangers 2 and 4 (HE 2 and HE 4) and the chiller is there to supply the cooling water. This heat recovery line helps to optimize the energy consumption in processes 2 and 3.

4.2 Mass and flow rates for process number 1, 2, 3, and 4

Table 4.1, Table 4.2, Table 4.3, and Table 4.4 provide the mass flow rates of the streams in processes number 1, 2, 3, and 4, respectively.

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Table 4.1: Details of flow rates for each stream based on notations in Figure 4.1

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	340521	0.0108	0.6
Water make-up [kg]	b	4203960	0.1333	8
Leaching solution [kg]	c1, c2, c3	42039602	1.3331	80
CS [kg]	d	2645342	0.0839	5
Leachate solution [kg]	e1, e2	44684944	1.417	85
Solid residues [kg]	f	2240898	0.0711	4.3
Filtrate solution [kg]	g1, g2, g3	42444047	1.3459	80.8
CO ₂ [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i	42990047	1.3632	81.8
Precipitated CaCO ₃ [kg]	j1,j2	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	42080047	1.3343	80.1
Purge [kg]	l	4208005	0.1334	8
Recovered leaching solution [kg]	m1, m2	37872042	1.2009	72.1
Cooling water from chiller [kg]	c4, c5	11668112	0.37	22

Table 4.2: Details of flow rates for each stream based on notations in Figure 4.2

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	196338	0.0062	0.4
Water make-up [kg]	b	1211967	0.0384	2.3
Leaching solution [kg]	c1, c2, c3, c4	12119670	0.3843	23.1
RCS [kg]	d	1211967	0.0384	2.3
Leachate solution [kg]	e1, e2	13331637	0.4227	25.4
Solid residues [kg]	f	807523	0.0256	1.5
Filtrate solution [kg]	g1, g2, g3, g4, g9	12524114	0.3971	23.8
CO ₂ [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i, g5, g6	13070114	0.4145	24.9
Precipitated CaCO ₃ [kg]	j1,j2	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	12160114	0.3856	23.1
Purge [kg]	l	1216011	0.0386	2.3
Recovered leaching solution [kg]	m1, m2	10944103	0.347	20.8
Cooling water from chiller [kg]	g7, g8	64706899	2.0518	123

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Table 4.3: Details of flow rates for each stream based on notations in Figure 4.3

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	425367	0.0135	0.8
Water make-up [kg]	b	1944444	0.0617	3.7
Leaching solution [kg]	c1, c2, c3, c4	19444444	0.6166	37
BFS [kg]	d	1555556	0.0493	3
Leachate solution [kg]	e1, e2	21000000	0.6659	40
Solid residues [kg]	f	1151111	0.0365	2.2
Filtrate solution [kg]	g1, g2, g3, g4, g9	19848889	0.6294	37.8
CO ₂ [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i, g5, g6	20394889	0.6467	38.8
Precipitated CaCO ₃ [kg]	j1,j2	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	19484889	0.6179	37.1
Purge [kg]	l	1948489	0.0618	3.7
Recovered leaching solution [kg]	m1, m2	17536400	0.5561	33.4
Cooling water from chiller [kg]	g7, g8	79356444	3	151

Table 4.4: Details of flow rates for each stream based on notations in Figure 4.4

Stream definition	Stream	Per year	Per second	Per minute
Leaching reagent make-up [kg]	a	1245211	0.0395	2.4
Water make-up [kg]	b	8301405	0.2632	15.8
Leaching solution [kg]	c1, c2, c3	83014049	2.6324	157.9
CS [kg]	d	1394636	0.0442	2.7
Leachate solution [kg]	e1, e2	84408685	2.6766	160.6
Solid residues [kg]	f	990192	0.0314	1.9
Filtrate solution [kg]	g1, g2, g3	83418493	2.6452	158.7
CO ₂ [kg]	h	400000	0.0127	0.8
Mineralization solution [kg]	i	83964493	2.6625	159.7
Precipitated CaCO ₃ [kg]	j1,j2	910000	0.0289	1.7
Leaching solution + unreacted Ca [kg]	k	83054493	2.6336	158
Purge [kg]	l	8305449	0.2634	15.8
Recovered leaching solution [kg]	m1, m2	74749044	2.3703	142.2
Cooling water from chiller [kg]	c4, c5	35462706	1.1245	67

The masses and flow rates of different streams in the processes are considerably different with each other.

Talking about the stream d where the industrial waste is added, the different mass flow rate originates from the different Ca content in each industrial waste and the different leaching efficiency in each process.

Consequently, the liquid to solid ratio, which is different in each process, is the reason to have different flow rate for leaching solution and the different flow rate of the reagent stems in the different concentrations of each one in the leaching solution.

Considering the above, stream f which is the solid residues exiting the processes and the rest of the streams will have different mass flow rates.

Coming to the temperatures, the temperatures in the streams are different due to different operational temperatures for each process and for leaching and mineralization. The results for each stream are shown in section 4.1.3.

Meanwhile, what is the same for each process, is that the temperature after the reactor is more than that of before the reactor. The reason is the generated heat in the reactor which increases the temperature of the streams differently since the mass flow rates are different.

4.3 Details of the streams for process number 1, 2, 3, and 4

The details of each stream including the temperatures are provided in this section.

Process number 1:

a: NH_4Cl make up at 0.6 kg/min

b: Water make up at 8 kg/min

c1: Aqueous solution of NH_4Cl (1mol/L) at 80 kg/min at $T = 83\text{ }^\circ\text{C}$

c2: Aqueous solution of NH_4Cl (1mol/L) at 80 kg/min at $T = 83\text{ }^\circ\text{C}$

c3: Aqueous solution of NH_4Cl (1mol/L) at 80 kg/min at $T = 80\text{ }^\circ\text{C}$

c4: Inlet cooling water from chiller at 9 kg/min at $T = 10\text{ }^\circ\text{C}$

c5: Outlet cooling water from chiller at 9 kg/min at $T = 20\text{ }^\circ\text{C}$

d: CS at 5 kg/min

e1: Leachate solution at 85 kg/min at $T = 80\text{ }^\circ\text{C}$

e2: Leachate solution at 85 kg/min at $T = 80\text{ }^\circ\text{C}$

f: Solid residues at 4.3 kg/min

g1: Filtrate solution (rich in Ca) at 80.8 kg/min at $T = 80\text{ }^\circ\text{C}$

g2: Filtrate solution (rich in Ca) at 80.8 kg/min at $T = 80\text{ }^\circ\text{C}$

g3: Filtrate solution (rich in Ca) at 80.8 kg/min at $T = 80\text{ }^\circ\text{C}$

h: CO_2 at 0.8 kg/min

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i: Solution after mineralization (NH_4Cl solution + CaCO_3 + unreacted Ca) at 81.8 kg/min at $T = 89^\circ\text{C}$

j1: Solid CaCO_3 at 1.7 kg/min

k: Recovered reagent (NH_4Cl solution + unreacted Ca) at 80.1 kg/min at $T = 89^\circ\text{C}$

l: Purge at 8 kg/min (10%) at $T = 89^\circ\text{C}$

m1: Return flow (NH_4Cl solution) at 72.1 kg/min at $T = 89^\circ\text{C}$

m2: Return flow (NH_4Cl solution) at 72.1 kg/min at $T = 89^\circ\text{C}$

j2: Dried CaCO_3 at 1.7 kg/min

Process number 2:

a: NH_4Cl make up at 0.6 kg/min

b: Water make up at 2.3 kg/min

c1: aqueous solution of NH_4Cl (2 mol/L) at 23.1 kg/min at $T = 55^\circ\text{C}$

c2: aqueous solution of NH_4Cl (2 mol/L) at 23.1 kg/min at $T = 55^\circ\text{C}$

c3: aqueous solution of NH_4Cl (2 mol/L) at 23.1 kg/min at $T = 65^\circ\text{C}$

c4: aqueous solution of NH_4Cl (2 mol/L) at 23.1 kg/min at $T = 85^\circ\text{C}$

d: RCF at 2.3 kg/min

e1: Leachate solution at 25.4 kg/min at $T = 85^\circ\text{C}$

e2: Leachate solution at 25.4 kg/min at $T = 85^\circ\text{C}$

f: Solid residues at 1.5 kg/min

g1: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85^\circ\text{C}$

g2: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85^\circ\text{C}$

g3: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 85^\circ\text{C}$

g4: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 75^\circ\text{C}$

g5: Heat recovery line (water) at 24.9 kg/min at $T = 65^\circ\text{C}$

g6: Heat recovery line (water) at 24.9 kg/min at $T = 75^\circ\text{C}$

g7: Inlet cooling water at 123 kg/min at $T = 10^\circ\text{C}$

g8: Outlet cooling water at 123 kg/min at $T = 20^\circ\text{C}$

g9: Filtrate solution (rich in Ca) at 23.8 kg/min at $T = 25^\circ\text{C}$

h: CO_2 at 0.8 kg/min

i: Solution after mineralization (NH_4Cl solution + CaCO_3 + unreacted Ca) at 24.9 kg/min at $T = 55^\circ\text{C}$

j1: Solid CaCO_3 at 1.7 kg/min

k: Recovered reagent (NH_4Cl solution + unreacted Ca) at 23.1 kg/min at $T = 55^\circ\text{C}$

l: Purge at 2.3 kg/min (10%) at $T = 55^\circ\text{C}$

m1: Return flow (NH_4Cl solution) at 20.8 kg/min at $T = 55^\circ\text{C}$

m2: Return flow (NH_4Cl solution) at 20.8 kg/min at $T = 55^\circ\text{C}$

j2: Dried CaCO₃ at 1.7 kg/min

Process number 3:

a: HCl make up at 0.8 kg/min

b: Water make up at 3.7 kg/min

c1: aqueous solution of HCl (4 mol/L) at 37 kg/min at T = 39 °C

c2: aqueous solution of HCl (4 mol/L) at 37 kg/min at T = 39 °C

c3: aqueous solution of HCl (4 mol/L) at 37 kg/min at T = 60 °C

c4: aqueous solution of HCl (4 mol/L) at 37 kg/min at T = 80 °C

d: BFS at 3 kg/min

e1: Leachate solution at 40 kg/min at T = 80 °C

e2: Leachate solution at 40 kg/min at T = 80 °C

f: Solid residues at 2.2 kg/min

g1: Filtrate solution (rich in Ca) at 37.8 kg/min at T = 80 °C

g2: Filtrate solution (rich in Ca) at 37.8 kg/min at T = 80 °C

g3: Filtrate solution (rich in Ca) at 37.8 kg/min at T = 80 °C

g4: Filtrate solution (rich in Ca) at 37.8 kg/min at T = 59 °C

g5: Heat recovery line (water) at 38.8 kg/min at T = 30 °C

g6: Heat recovery line (water) at 38.8 kg/min at T = 70 °C

g7: Inlet cooling water at 151 kg/min at T = 10 °C

g8: Outlet cooling water at 151 kg/min at T = 20 °C

g9: Filtrate solution (rich in Ca) at 37.8 kg/min at T = 20 °C

h: CO₂ at 0.8 kg/min

i: Solution after mineralization (HCl solution + CaCO₃ + unreacted Ca) at 38.8 kg/min at T = 39 °C

j1: Solid CaCO₃ at 1.7 kg/min

k: Recovered reagent (HCl solution + unreacted Ca) at 37.1 kg/min at T = 39 °C

l: Purge at 3.7 kg/min (10%) at T = 39 °C

m1: Return flow (HCl solution) at 33.4 kg/min at T = 39 °C

m2: Return flow (HCl solution) at 33.4 kg/min at T = 39 °C

j2: Dried CaCO₃ at 1.7 kg/min

Process number 4:

a: Acetic acid make up at 2.4 kg/min

b: Water make up at 15.8 kg/min

c1: aqueous solution of acetic acid (10 wt%) at 157.9 kg/min at T = 29 °C

c2: aqueous solution of acetic acid (10 wt%) at 157.9 kg/min at T = 29 °C

4 Results and discussion

- c3: aqueous solution of acetic acid (10wt%) at 157.9 kg/min at T = 25 °C
 c4: inlet cooling water from chiller at 67 kg/min at T = 10 °C
 c5: outlet cooling water from chiller at 67 kg/min at T = 20 °C
 d: CS at 2.7 kg/min
 e1: Leachate solution at 160.6 kg/min at T = 25 °C
 e2: Leachate solution at 160.6 kg/min at T = 25 °C
 f: Solid residues at 1.9 kg/min
 g1: Filtrate solution (rich in Ca) at 158.7 kg/min at T = 25 °C
 g2: Filtrate solution (rich in Ca) at 158.7 kg/min at T = 25 °C
 g3: Filtrate solution (rich in Ca) at 158.7 kg/min at T = 25 °C
 h: CO₂ at 0.8 kg/min
 i: Solution after mineralization (acetic acid solution + CaCO₃ + unreacted Ca) at 159.7 kg/min at T = 30 °C
 j1: Solid CaCO₃ at 1.7 kg/min
 k: Recovered reagent (acetic acid solution + unreacted Ca) at 158 kg/min at T = 30 °C
 l: Purge at 15.8 kg/min (10%) at T = 30 °C
 m1: Return flow (acetic acid solution) at 142.2 kg/min at T = 30 °C
 m2: Return flow (acetic acid solution) at 142.2 kg/min at T = 30 °C
 j2: Dried CaCO₃ at 1.7 kg/min

4.4 Sizes and required numbers of equipment for process number 1, 2, 3, and 4

Table 4.5 represents the sizes of the equipment for each process.

Table 4.5: Capacity and required number of equipment for each process

Process	Mixer 1 (capacity [L]/ required number)	Mixer 2 (capacity [L]/ required number)	Storage Tank (capacity [L]/ required number)	Purge tank (capacity [L]/ required number)	PFR (capacity [L]/ required number)
1	5101/ 2	5101/ 3	5101/ 2	5101/ 2	10202/ 1
2	1522/ 2	1522/ 3	1522/ 2	1522/ 2	761/ 1
3	4795/ 2	4795/ 3	4795/ 2	4795/ 2	599/ 1
4	19271/ 2	19271/ 3	19271/ 2	19271/ 2	19271/ 1

4 Results and discussion

There will be one PFR required for each process since the nature of the mineralization has been assumed to be continuous, not batchwise.

Since the nature of the leaching is batchwise and time consuming, the process needs to have three mixers for leaching to ensure the continuity of the mineralization process and supply the NPSH of the pump.

At the same time, one mixer is supplying the centrifuge 1, the other is mixing to extract Ca, and the last one is being filled with the leaching solution and the industrial waste. Figure 4.5, Figure 4.6, and Figure 4.7 are simplified PFDs and show the way these three tanks work (assuming leaching time to be 60 minutes).

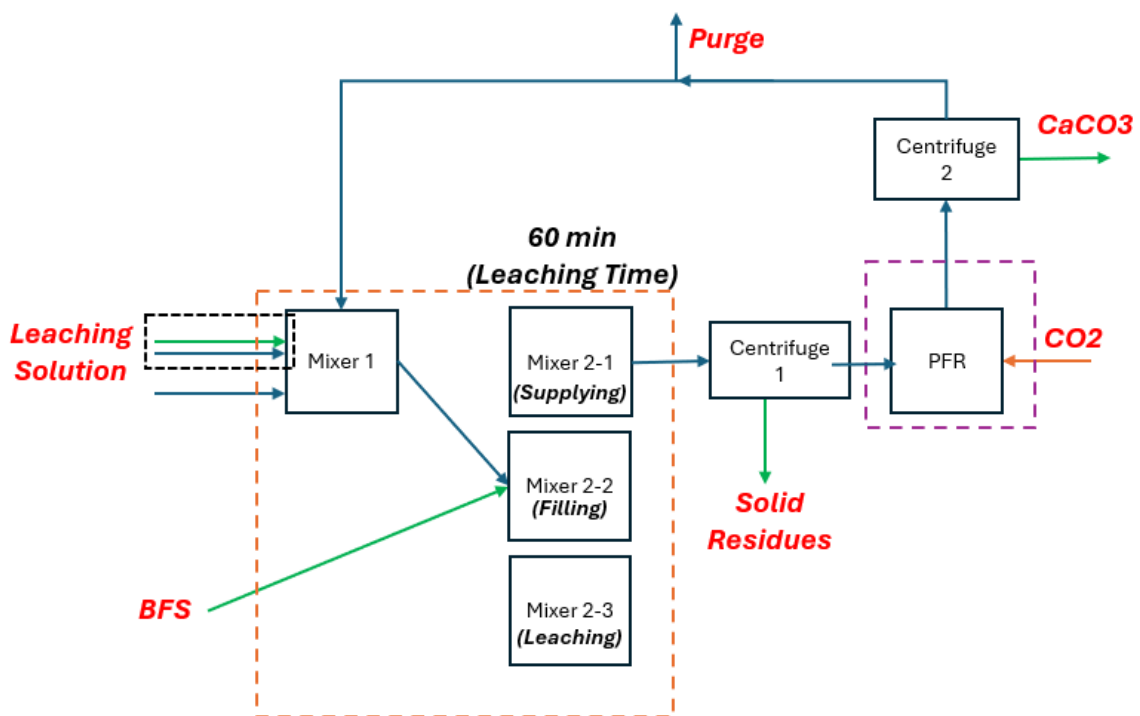


Figure 4.5: Performance of leaching mixers from time 0 to 60 minutes

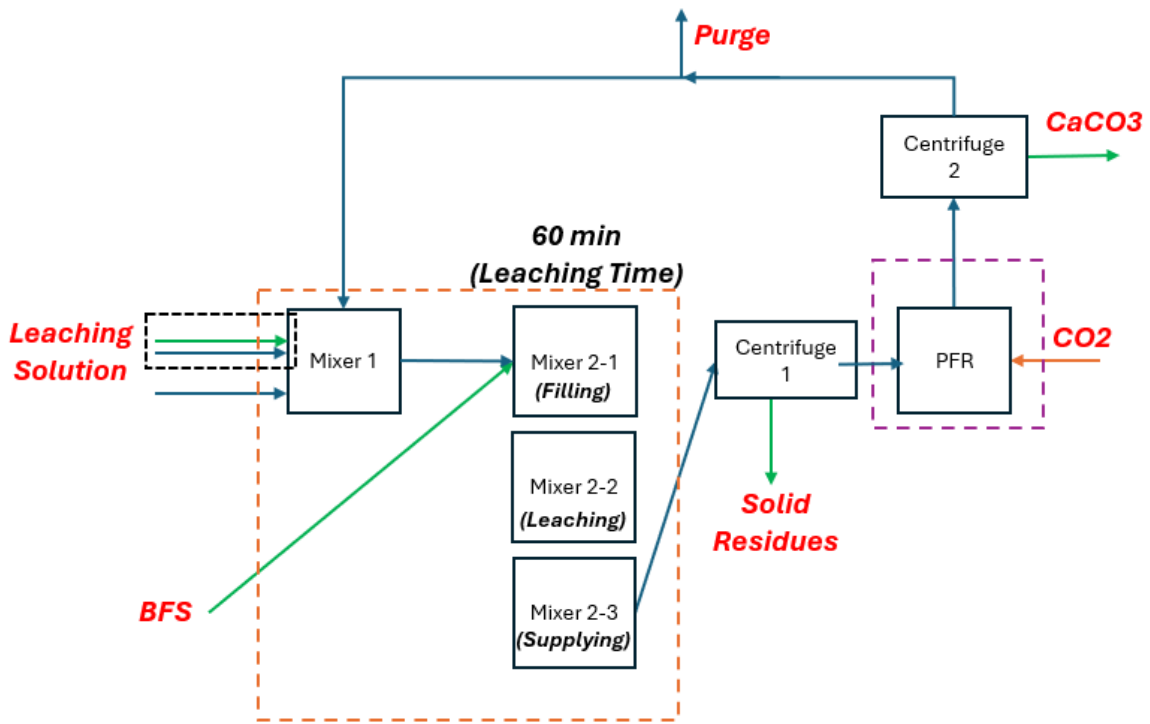


Figure 4.6: Performance of leaching mixers from time 60 to 120 minutes

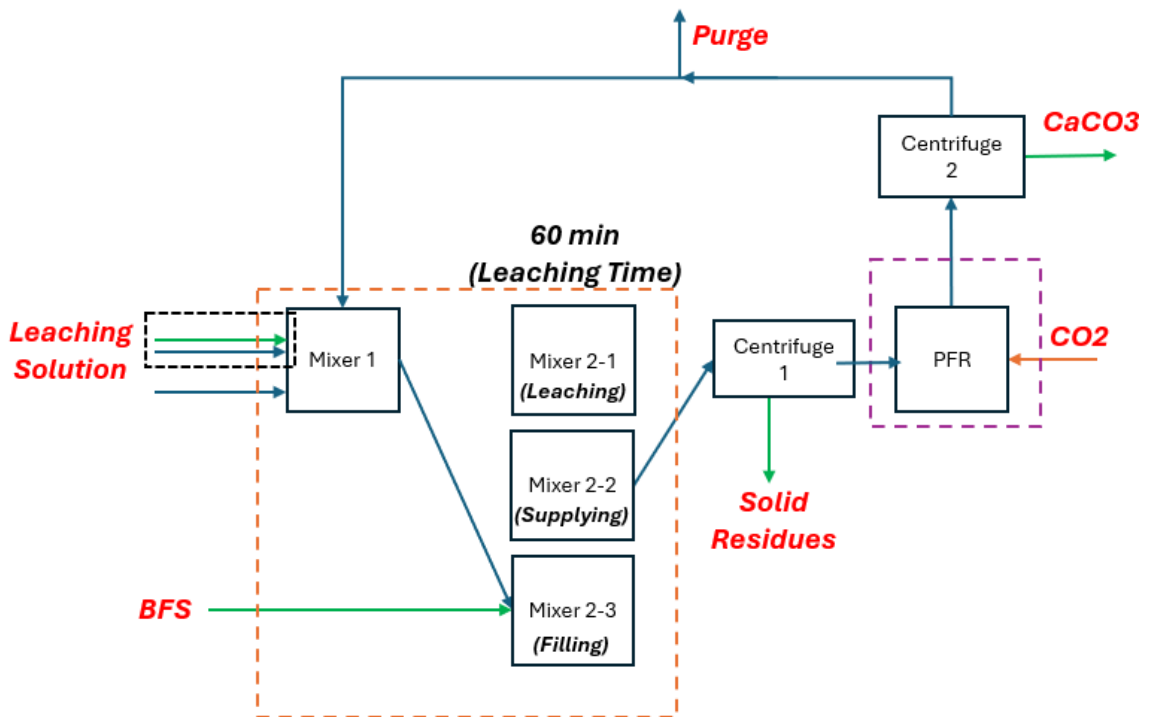


Figure 4.7: Performance of leaching mixers from time 120 to 180 minutes

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The required number of mixers 1, storage tank, and purge tank are calculated based on the above logic but with this consideration that they do not require a time for any process. They get filled in and then get empty to supply the line or the next stage of the process (mixer 1 is mixing the reagent with water and recovered leaching solution, but it does not need any time to be done. It can be done while the mixer is getting filled in).

Finally, it is obvious that due to safety concerns and probable failure of any equipment or line, there will be the need to have more than one for some of the equipment (the auxiliary equipment) such as pumps or even the reactor. But this issue has not been taken into consideration in this study.

The limiting condition for sizing of mixers, storage tank, and purge tank is the leaching time. Leaching is a batchwise process in all four processes with a specific operating time. Therefore, calculating the flow rate per leaching time gives the size of the mentioned equipment. As is obvious from Table 4.5, the sizes of mixers 1 and 2, storage tank, and purge tank are the same for all four processes.

On the other hand, the limiting condition for sizing of the reactors is mineralization time. The mineralization time is the residence time in the reactor. So, calculating the flow rate per mineralization time results in the size of reactor.

For process 1 where the leaching time is half of mineralization time, the size of the rest of the equipment is half of the reactor.

In the case of process 2, where leaching time is double as mineralization time, the size of reactor is half of the rest of the equipment.

Talking about process 3, the mineralization time is one eighth of leaching time which shows itself in the size of the reactor which is one eighth of other equipment.

Finally, for process 4, where the leaching and mineralization time are the same, the sizes of all equipment are the same.

4.5 Energy consumption for process number 1, 2, 3, and 4

This section provides the details of energy consumption of each process for energy consuming equipment and energy consuming processes such as heating or cooling in a year. Table 4.6 provides this data.

Table 4.6: Details of yearly energy consumption and the total amount for each process

Process	Pumps [kWh]	Agitators [kWh]	Centrifuges [kWh]	Grinding [kWh]	Drying CaCO ₃ [kWh]	Heating [kWh]	Cooling [kWh]	Total [kWh]
1	609	315360	134055	35511	80907	0	27226	593668
2	182	131400	39995	16270	80907	304969	150983	724706
3	286	262800	63000	20882	80907	475881	185165	1088921
4	1150	630720	253226	18722	80907	0	82746	1067471

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The total energy consumption of each process consists of the sum of consumed energies by pumps, agitators, centrifuges, grinder, dryer, heating, and cooling (chiller), provided in Table 4.6.

Talking about the pumps, as seen in Table 4.6, process 4 has the highest energy consumption for pumps (1150 kWh) while process 2 has the lowest (182 kWh). Process 3 and 1 are between these amounts. The only reason is the different flow rates of all processes since other role-playing variables such as density and head loss have been assumed to be the same for all 4 processes.

The agitators' energy consumption has been calculated based on the power of each agitator with regards to the size of the tank which is being agitated. The corresponding power for each size has been found online and the numbers are provided in Table 3.3. As expected, the highest energy consumption of agitators belongs to process 4 with 630720 kWh. After that, processes 1, 3, and 2 stand with 315360 kWh, 262800 kWh, and 131400 kWh, respectively.

The energy consumption of the centrifuges was found to be a function of the flow rate, so, not surprisingly, process 4 has the highest and process 2 owns the lowest energy consumption for centrifuges with 262226 and 39995 kWh, respectively.

When it comes to grinding energy consumption, process number 4 has the second lowest energy consumption of 18722 kWh due to its comparably low solid to liquid ratio and high extraction efficiency. The only lower energy consumption belongs to process 2 with 16270 kWh and the highest is for process 1 with 35511 kWh.

The drying energy consumption is the same for all 4 processes (80907 kWh) since the amount of the produced CaCO_3 is the same and the moisture content was assumed to be constant for all processes.

The heating demand for processes number 2 and 3 was calculated to be 304969 kWh and 475881 kWh and the cooling demand was calculated to be 150983 kWh and 185165 kWh, respectively. It is good to remember that these values could have been approximately 30% more if the heat recovery line did not exist in the system.

Processes number 1 and 4 do not have the heating demand since they have the same operational temperature for both leaching and mineralization. There is only the need to remove the generated heat in the reactor to cool down the stream to the operational temperatures. The energy consumption of the chiller to provide the cooling water for this purpose is 27226 and 82746 kWh for processes 1 and 4.

Noteworthy to indicate is that contrary to processes 2 and 3 where insulation is suggested to save the generated heat in the reactor which helps to reduce the heating demand, it is suggested to have no insulation for processes number 1 and 4 to lose some heat in purge tank and mixer 1 to reduce the energy consumption of the chiller.

This lost heat has been calculated and considered in the calculation of chiller's energy consumption for processes 1 and 4 using equation (4.1).

$$Q_{loss} (W) = h (W/m^2C) \times A (m^2) \times (T_s(C) - T_a(C)) \quad (4.1)$$

Where:

Q_{loss} is the lost heat due to heat transfer in [W],

h is the heat transfer coefficient of air with no wind in [$W/m^2\text{C}$],

A is the heat transfer area in [m^2],

T_s is the temperature of the surface of the purge tank and mixer 1 in [$^{\circ}\text{C}$],

T_a is the ambient temperature in [$^{\circ}\text{C}$].

For process 1, the temperature of the stream after the reactor is calculated to be 89°C which is assumed to be the T_s . The T_a is also assumed to be 25°C . The heat transfer coefficient was assumed as $10\text{ W}/m^2\text{C}$ [59]. The area of mixer 1 and the purge tank was assumed based on their volume which is 5101 L equal to 5.101 m^3 . To consider more volume due to real-time conditions, it was assumed that the volume will be 12 m^3 which could be a tank of $2\text{ m} \times 2\text{ m} \times 3\text{ m}$. Therefore, the area of each tank is 28 m^2 and the whole area is 56 m^2 .

So, the heat loss will be:

$$Q_{loss} (W) = 10 \frac{W}{m^2C} \times 56\text{ m}^2 \times (89\text{ C} - 25\text{ C}) \approx 36000\text{ W} = 36\text{ kW} \quad (4.2)$$

Using energy balance equation in (3.42), the heat that should be removed by chiller will be calculated as 16 kW resulting in chiller power to be approximately 3 kW and its yearly energy consumption will be 27226 kWh , calculated using the method in chapter 3.

A similar procedure has been conducted for process 4 and the heat loss from the pipes has been neglected due to small surface of them and for simplification.

4.6 Economics of process number 1, 2, 3, and 4

The economic assessment of each process in Middle East, Norway, and US are provided in this section. The calculations have been conducted based on the procedure in section 3.5. Table 4.7, Table 4.8,

Table 4.9, and Table 4.10 provide this assessment for processes number 1, 2, 3, and 4 in Middle East. Table 4.11, Table 4.12, Table 4.13, and Table 4.14 show the results for the mentioned processes in Norway, and Table 4.15, Table 4.16, Table 4.17, and Table 4.18 represent the results for the processes in the US.

4 Results and discussion

Table 4.7: Economic assessment of the process No. 1 in Middle East

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-28472	-28472	-28472
NH ₄ Cl cost [USD]	-27242	-91941	-190692
Process water cost [USD]	-31740	-31740	-31740
Total cost (part of OPEX) [USD]	-87454	-152153	-250904
Yearly profit (sum of total revenue and total cost) [USD]	194646	129947	31196

Table 4.8: Economic assessment of the process No. 2 in Middle East

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-34757	-34757	-34757
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-59614	-96919	-153857
Yearly profit (sum of total revenue and total cost) [USD]	222486	185181	128243

Table 4.9: Economic assessment of the process No. 3 in Middle East

Parameter	HCl bought in East Asia	HCl bought in the US	HCl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-52225	-52225	-52225
HCl cost [USD]	-17015	-59551	-42537
Process water cost [USD]	-14681	-14681	-14681
Total cost (part of OPEX) [USD]	-83920	-126457	-109442
Yearly profit (sum of total revenue and total cost) [USD]	198180	155643	172658

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Table 4.10: Economic assessment of the process No. 4 in Middle East

Parameter	Acetic acid bought in East Asia	Acetic acid bought in the US	Acetic acid bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-51196	-51196	-51196
Acetic Acid cost [USD]	-585249	-510536	-846743
Process water cost [USD]	-62676	-62676	-62676
Total cost (part of OPEX) [USD]	-699121	-624408	-960615
Yearly profit (sum of total revenue and total cost) [USD]	-417021	-342308	-678515

As seen from the Table 4.7 to Table 4.10, process number 1, 2, and 3 have positive yearly profit in Middle East which means they are profitable. On the other hand, process 4 has negative yearly profit which means it is not profitable in Middle East. The other important point is that for processes 1, 2, and 3, the process will have more profit if the origin of the reagent is East Asia.

Table 4.11: Economic assessment of the process No. 1 in Norway

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-25308	-25308	-25308
NH ₄ Cl cost [USD]	-27242	-91941	-190692
Process water cost [USD]	-31740	-31740	-31740
Total cost (part of OPEX) [USD]	-84290	-148989	-247740
Yearly profit (sum of total revenue and total cost) [USD]	197810	133111	34360

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Table 4.12: Economic assessment of the process No. 2 in Norway

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-30894	-30894	-30894
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-55752	-93056	-149994
Yearly profit (sum of total revenue and total cost) [USD]	226348	189044	132106

Table 4.13: Economic assessment of the process No. 3 in Norway

Parameter	HCl bought in East Asia	HCl bought in the US	HCl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-46421	-46421	-46421
HCl cost [USD]	-17015	-59551	-42537
Process water cost [USD]	-14681	-14681	-14681
Total cost (part of OPEX) [USD]	-78116	-120653	-103638
Yearly profit (sum of total revenue and total cost) [USD]	203984	161447	178462

Table 4.14: Economic assessment of the process No. 4 in Norway

Parameter	Acetic acid bought in East Asia	Acetic acid bought in the US	Acetic acid bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	0	0	0
Total revenue [USD]	282100	282100	282100
Energy cost [USD]	-45506	-45506	-45506
Acetic Acid cost [USD]	-585249	-510536	-846743
Process water cost [USD]	-62676	-62676	-62676
Total cost (part of OPEX) [USD]	-693431	-618718	-954925
Yearly profit (sum of total revenue and total cost) [USD]	-411331	-336618	-672825

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With attention to Table 4.11 to Table 4.14 which show the yearly profit in Norway, it is clear that the trend is similar to Middle East, but the numbers are different. Processes 1, 2, and 3 are slightly more profitable in Norway than in Middle East and process 4 is observing less negative profit.

Table 4.15: Economic assessment of the process No. 1 in the US

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	24000	24000	24000
Total revenue [USD]	306100	306100	306100
Energy cost [USD]	-46900	-46900	-46900
NH ₄ Cl cost [USD]	-27242	-91941	-190692
Process water cost [USD]	-31740	-31740	-31740
Total cost (part of OPEX) [USD]	-105881	-170580	-269331
Yearly profit (sum of total revenue and total cost) [USD]	200219	135520	36769

Table 4.16: Economic assessment of the process No. 2 in the US

Parameter	NH ₄ Cl bought in East Asia	NH ₄ Cl bought in the US	NH ₄ Cl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	24000	24000	24000
Total revenue [USD]	306100	306100	306100
Energy cost [USD]	-57252	-57252	-57252
NH ₄ Cl cost [USD]	-15707	-53011	-109950
Process water cost [USD]	-9150	-9150	-9150
Total cost (part of OPEX) [USD]	-82109	-119414	-176352
Yearly profit (sum of total revenue and total cost) [USD]	223991	186686	129748

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Table 4.17: Economic assessment of the process No. 3 in the US

Parameter	HCl bought in East Asia	HCl bought in the US	HCl bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	24000	24000	24000
Total revenue [USD]	306100	306100	306100
Energy cost [USD]	-86025	-86025	-86025
HCl cost [USD]	-17015	-59551	-42537
Process water cost [USD]	-14681	-14681	-14681
Total cost (part of OPEX) [USD]	-117720	-160257	-143242
Yearly profit (sum of total revenue and total cost) [USD]	188380	145843	162858

Table 4.18: Economic assessment of the process No. 4 in the US

Parameter	Acetic acid bought in East Asia	Acetic acid bought in the US	Acetic acid bought in the EU
Revenue of CaCO ₃ [USD]	282100	282100	282100
Revenue of CO ₂ [USD]	24000	24000	24000
Total revenue [USD]	306100	306100	306100
Energy cost [USD]	-84330	-84330	-84330
Acetic Acid cost [USD]	-585249	-510536	-846743
Process water cost [USD]	-62676	-62676	-62676
Total cost (part of OPEX) [USD]	-732255	-657542	-993749
Yearly profit (sum of total revenue and total cost) [USD]	-426155	-351442	-687649

Table 4.15 to Table 4.18 and comparing the yearly profit of process 1 in Norway, it is clear that process 1 is slightly more profitable in the US than Norway. Processes 2 and 3 are more profitable in Norway than the US and process 4 has a worse condition in the US than Norway.

4.7 KPIs of process number 1, 2, 3, and 4

Following the calculations in section 3.6, the introduced KPIs are calculated for all four processes and the results are shown in diagrams in Figure 4.8 to Figure 4.39. The exact definition of KPIs is provided in section 3.6, in the figures, only a brief definition is provided only for overview.

4 Results and discussion

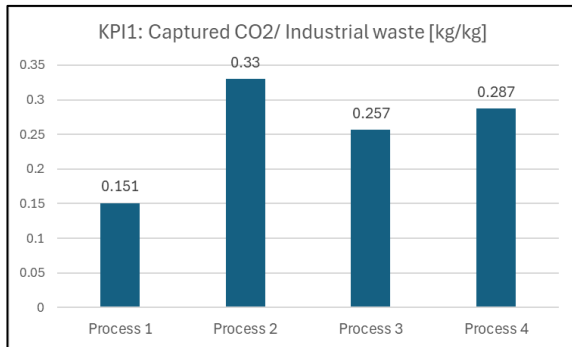


Figure 4.8: KPI1, captured CO₂ per consumed industrial waste

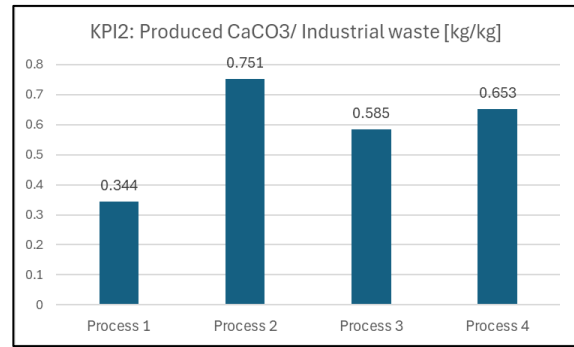


Figure 4.9: KPI2, produced CaCO₃ per consumed industrial waste

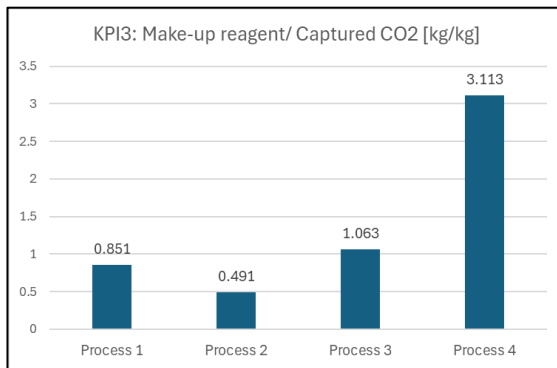


Figure 4.10: KPI3, mass of make-up reagent per mass of captured CO₂

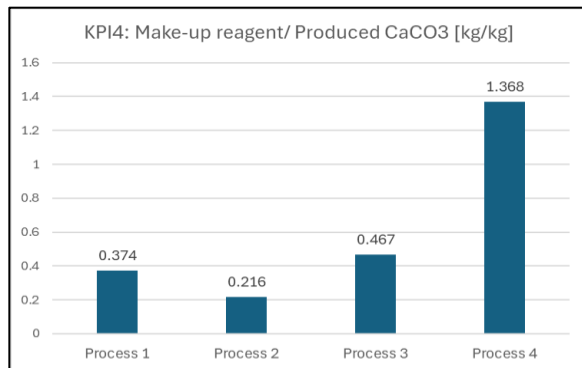


Figure 4.11: KPI4, mass of make-up reagent per mass of produced CaCO₃

4 Results and discussion

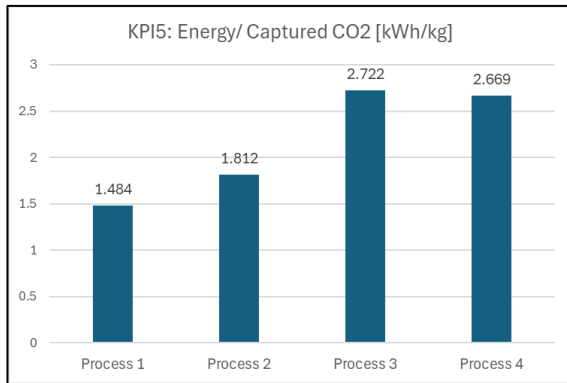


Figure 4.12: KPI5, yearly energy consumption per mass of captured CO₂

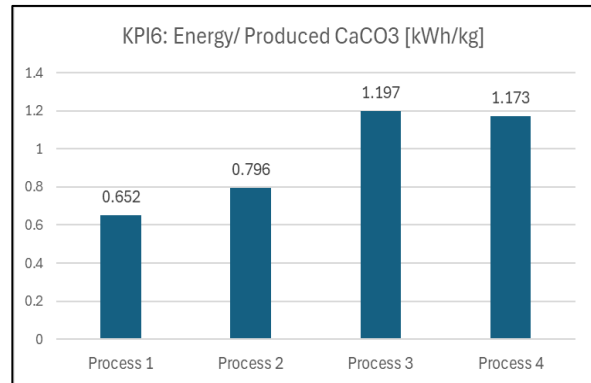


Figure 4.13: KPI6, yearly energy consumption per mass of produced CaCO₃

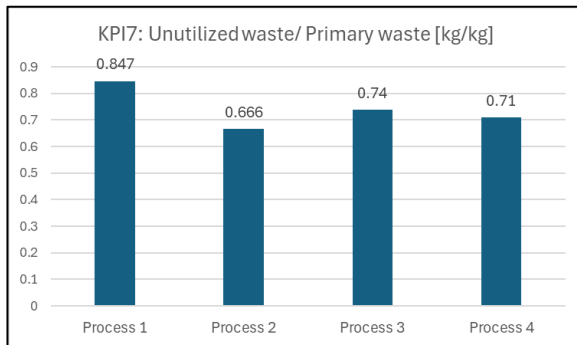


Figure 4.14: KPI7, mass of unutilized industrial waste per mass of primary added industrial waste

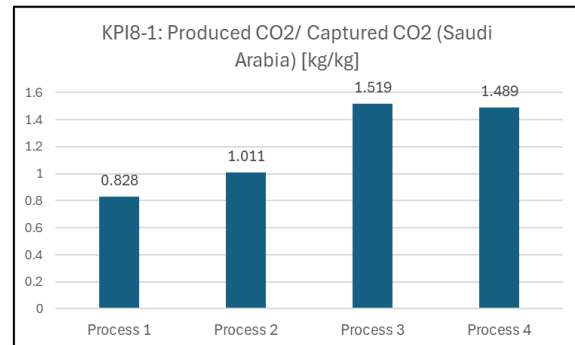


Figure 4.15: KPI8-1, mass of produced CO₂ by the process in Middle East per mass of captured CO₂

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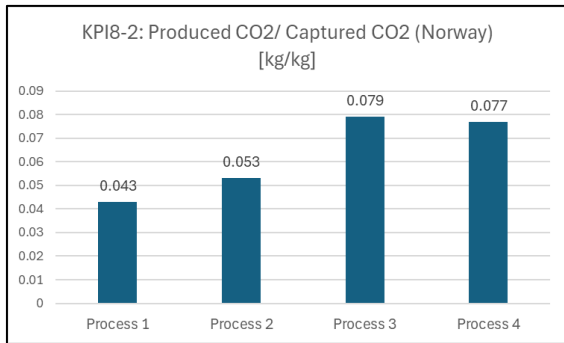


Figure 4.16: KPI8-2, mass of produced CO₂ by the process in Norway per mass of captured CO₂

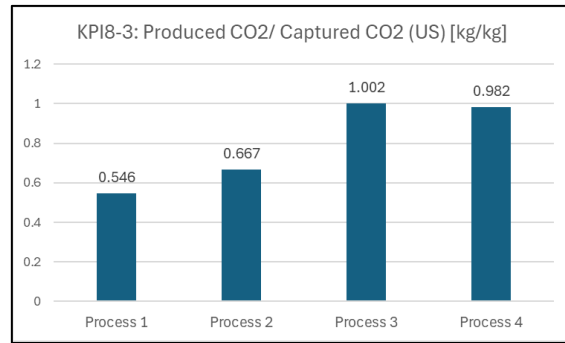


Figure 4.17: KPI8-3, mass of produced CO₂ by the process in the US per mass of captured CO₂

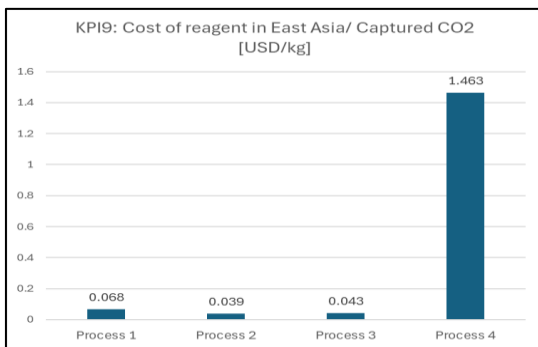


Figure 4.18: KPI9, cost of the reagent supplied from East Asia per mass of captured CO₂

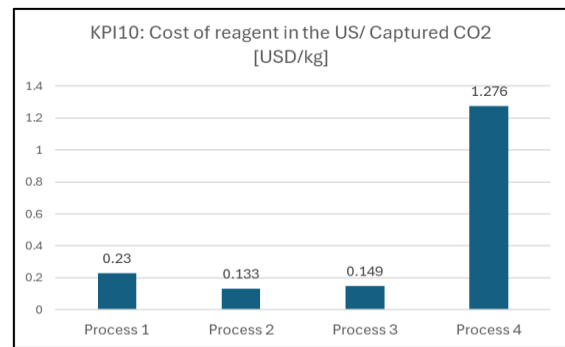


Figure 4.19: KPI10, cost of the reagent supplied from the US per mass of captured CO₂

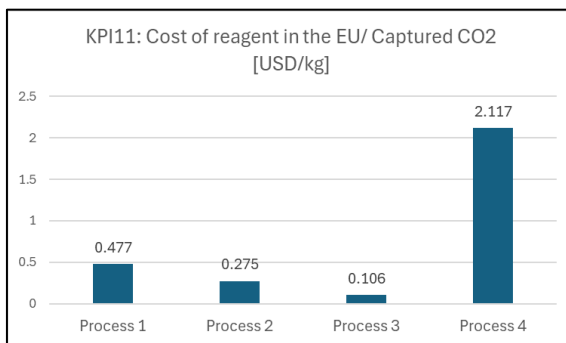


Figure 4.20: KPI11, cost of the reagent supplied from the EU per mass of captured CO₂

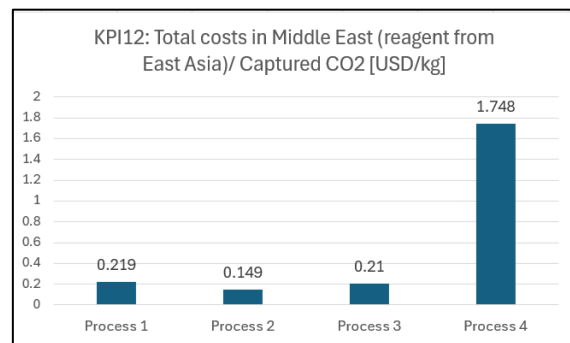


Figure 4.21: KPI12, total costs in Middle East (part of OPEX) with reagent from East Asia per mass of captured CO₂

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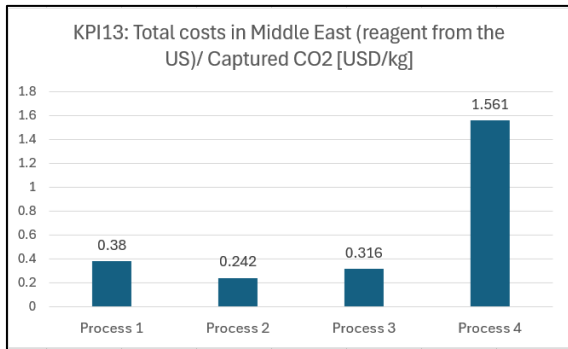


Figure 4.22: KPI13, total costs in Middle East (part of OPEX) with reagent from the US per mass of captured CO₂

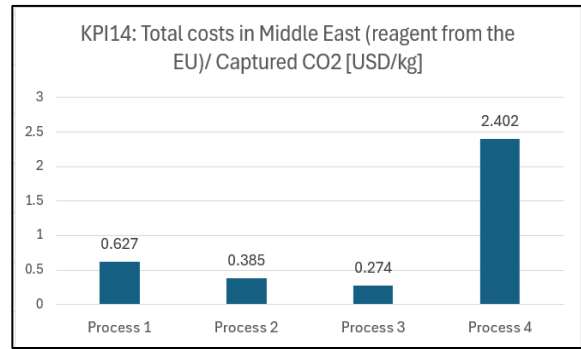


Figure 4.23: KPI14, total costs in Middle East (part of OPEX) with reagent from the EU per mass of captured CO₂

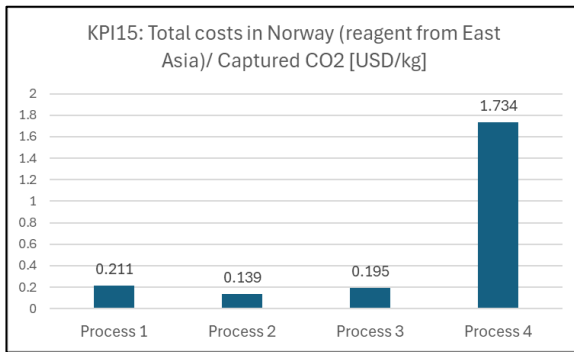


Figure 4.24: KPI15, total costs in Norway (part of OPEX) with reagent from East Asia per mass of captured CO₂

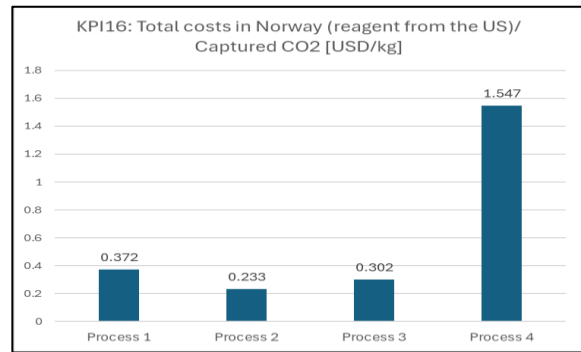


Figure 4.25: KPI16, total costs in Norway (part of OPEX) with reagent from the US per mass of captured CO₂

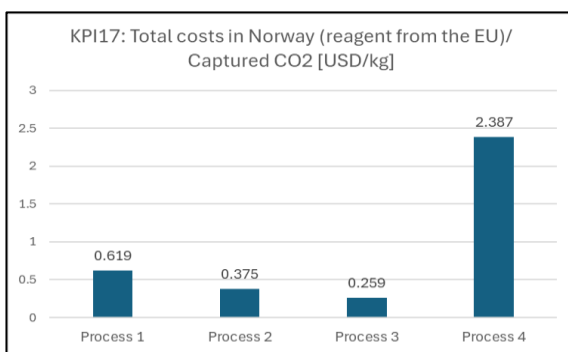


Figure 4.26: KPI17, total costs in Norway (part of OPEX) with reagent from the EU per mass of captured CO₂

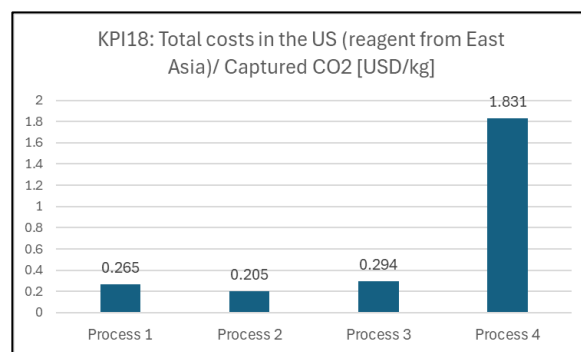


Figure 4.27: KPI18, total costs in the US (part of OPEX) with reagent from East Asia per mass of captured CO₂

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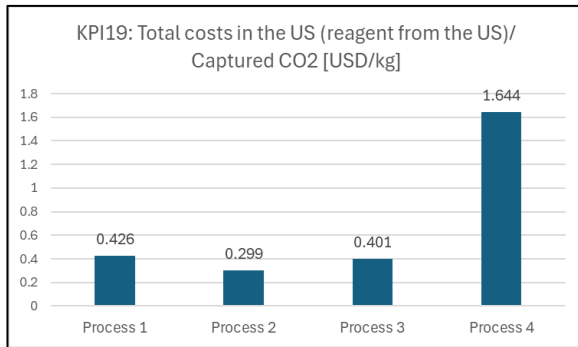


Figure 4.28: KPI19, total costs in the US (part of OPEX) with reagent from the US per mass of captured CO₂

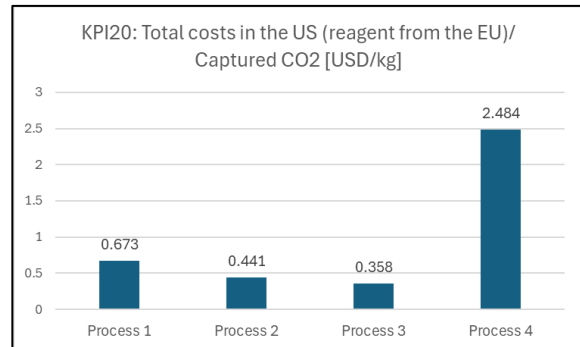


Figure 4.29: KPI20, total costs in the US (part of OPEX) with reagent from the EU per mass of captured CO₂

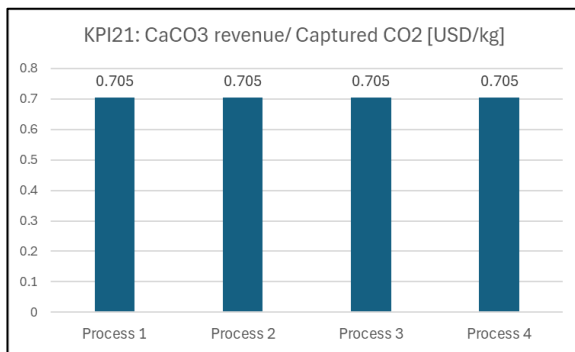


Figure 4.30: KPI21, revenue from sales of CaCO₃ per mass of captured CO₂

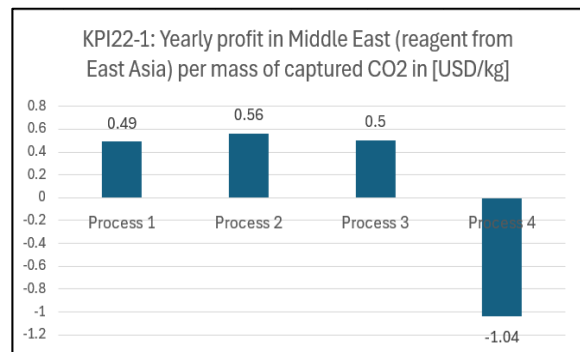


Figure 4.31: KPI22-1, yearly profit in Middle East with reagent supplied from East Asia per mass of captured CO₂

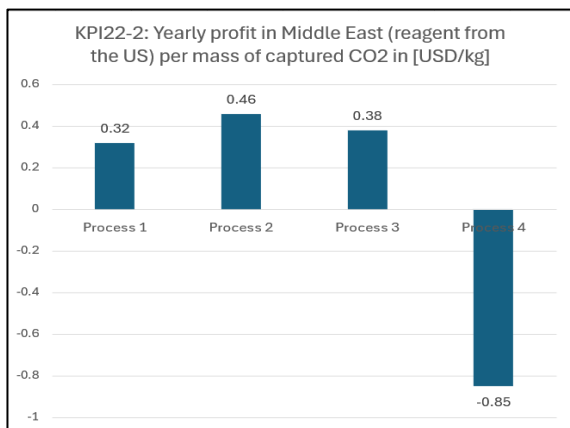


Figure 4.32: KPI22-2, yearly profit in Middle East with reagent supplied from the US per mass of captured CO₂

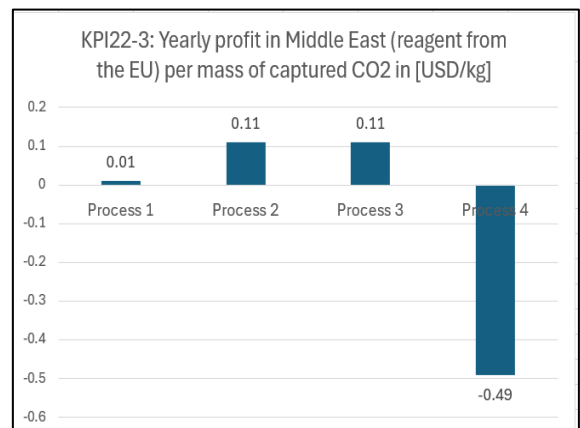


Figure 4.33: KPI22-3, yearly profit in Middle East with reagent supplied from the EU per mass of captured CO₂

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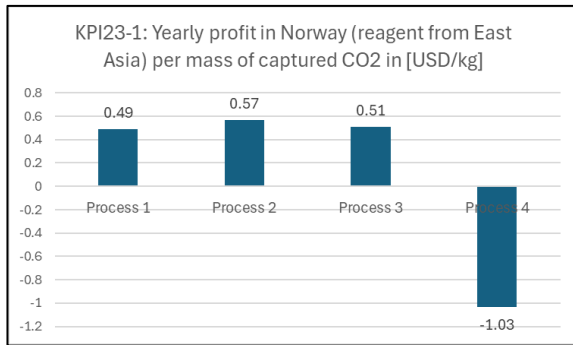


Figure 4.34: KPI23-1, yearly profit in Norway with reagent supplied from East Asia per mass of captured CO₂

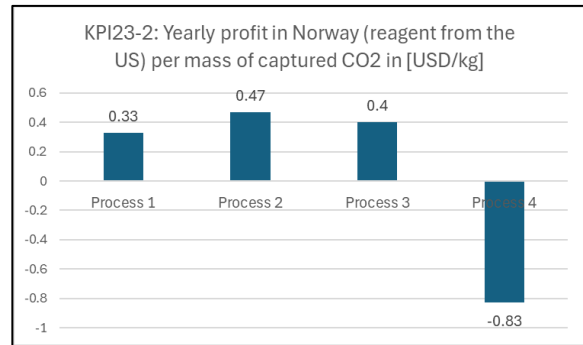


Figure 4.35: KPI23-2, yearly profit in Norway with reagent supplied from the US per mass of captured CO₂

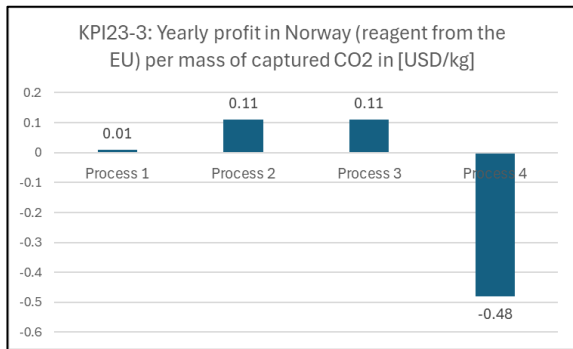


Figure 4.36: KPI23-3, yearly profit in Norway with reagent supplied from the EU per mass of captured CO₂

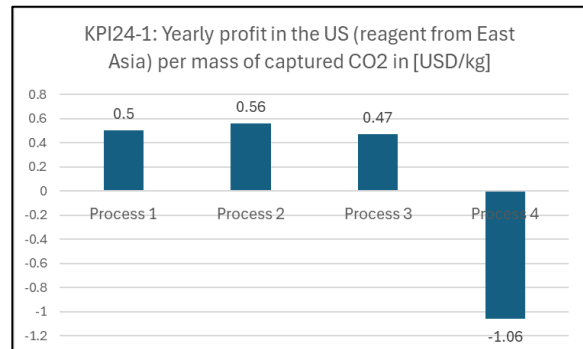


Figure 4.37: KPI24-1, yearly profit in the US with reagent supplied from East Asia per mass of captured CO₂

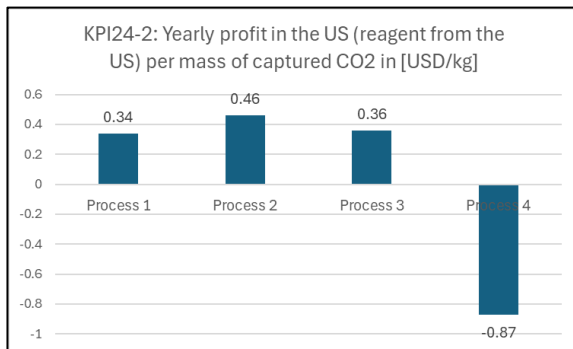


Figure 4.38: KPI24-2, yearly profit in the US with reagent supplied from the US per mass of captured CO₂

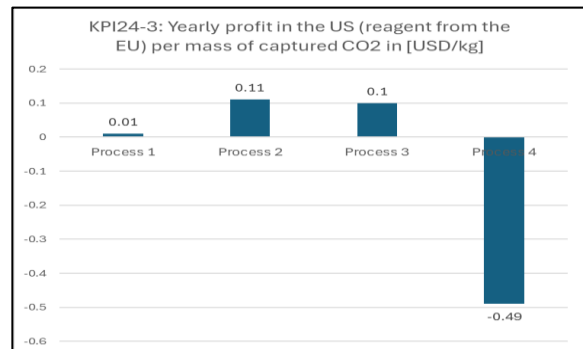


Figure 4.39: KPI24-3, yearly profit in the US with reagent supplied from the EU per mass of captured CO₂

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Regarding KPI1 (Figure 4.8), process 2 has the most amount of captured CO₂ per mass of industrial waste (0.33 kg/kg). The reason is the high amount of initial Ca in industrial waste (approximately 0.51 kg/kg). The second highest belongs to process 4, and the third highest belongs to process 3 with 0.287 and 0.257 kg/kg. Although the industrial waste in process 4 and 3 do not have as much amount of Ca as process 2, their higher extraction efficiency makes them good competitors. The lowest amount of captured CO₂ per mass of the waste is for process 1 with 0.151 kg/kg due to its low extraction efficiency.

The trend is similar to KPI1 for KPI2 in Figure 4.9 where the amount of produced CaCO₃ per mass of industrial waste is calculated. Since the produced CaCO₃ follows the rate of captured CO₂, process 2 has the highest mass of CaCO₃ per mass of waste (0.751 kg/kg), which is followed by process 4, 3, and 1 (0.653, 0.505, and 0.344 kg/kg, respectively).

So, in terms of the capacity to capture CO₂ and produce CaCO₃, process 2 is the best option, followed by processes 4 and 3, in that order.

When it comes to KPI3 and KPI4 in Figure 4.10 and Figure 4.11 where the consumption of the reagent per mass of captured CO₂ and produced CaCO₃ is shown, process 4 has dramatically higher consumption of reagent compared to other processes. Process 4 consumes 3.113 kg of reagent per each kilogram of captured CO₂ and 1.368 kg per each kilogram of produced CaCO₃ while these numbers are 0.491 kg/kg and 0.216 kg/kg per captured CO₂ and produced CaCO₃ for process 2. The reason for such high consumption in process 4 is its comparably lower solid to liquid ratio which results in more volume of water and more mass of reagent to reach the required concentration. So, process 2 and 1 consume the least amount of reagent to reach the declared extraction efficiencies.

The total energy consumption of each process per mass of captured CO₂ and produced CaCO₃ are calculated in KPI5 and KPI6 and shown in Figure 4.12 and Figure 4.13. Process 3 has the highest energy consumption per mass of captured CO₂ with 2.722 kWh/kg and per mass of produced CaCO₃ with 1.197 kWh/kg. The second place belongs to process 4 with 2.669 and 1.173 kWh/kg per captured CO₂ and produced CaCO₃, respectively. Processes 2 and 1 seem to be more energy efficient with 1.812 and 1.484 kWh/kg of captured CO₂ and 0.796 and 0.652 kWh/kg of produced CaCO₃. The reasons of high energy consumption for process 3 are mostly high heating and cooling demands due to temperature difference in the system, and the reasons for process 4 are mostly the agitators and centrifuges consumptions due to higher flow rate in this process. So, process 1 and then 2 are the most optimized ones in terms of energy consumption.

KPI7 represents the mass of unutilized waste per mass of primary waste added to the process. It means the mass of the solid residues exiting the process per the mass of the waste added to the process for leaching. The results are shown in Figure 4.14 which depicts that the solid residues coming out of the four processes are somewhere between 0.66 kg and 0.85 kg per kilogram of added waste.

KPI8-1, KPI8-2, and KPI8-3 show the carbon footprint of the four processes in Middle East, Norway, and the US, respectively. As shown (and expected) in Figure 4.15, Figure 4.16, and Figure 4.17, all four processes produce the highest amount of CO₂ per mass of captured CO₂ in Middle East and the lowest amount in Norway. Among all four processes, processes 1 and 2 produce the least amount of CO₂ in each of three chosen countries and the reason is their

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lowest energy consumption compared with processes 3 and 4. Processes 2, 3, and 4 produce more CO₂ than captured if the plant is in Middle East. Process 3 in the US produces more CO₂ than captured too and process 4 in the US is quite on the border of producing the same amount of CO₂ as it is captured. KPI8 is one of the most important KPIs to be considered and to be checked in the early stages of concept studies of the project since it has the potential to stop the whole project.

Talking about the economics of the processes and the cost of reagents supplied from East Asia, the US, and the EU, KPI9, KPI10, and KPI11 provide valuable information in Figure 4.18, Figure 4.19, and Figure 4.20. As can be expected from KPI3, process 4 has the highest price of consumed reagent per mass of captured CO₂ regardless of the origin and type of the reagent since its consumption is much higher than the rest of the processes. Meanwhile, if supplied from East Asia (figure 4.18), the cost of the reagent per mass of the captured CO₂ is below 0.1 USD/kg for processes 1, 2, and 3. If supplied from the US, this number is below 0.3 USD/kg for processes 1, 2, and 3, and finally, if supplied from the EU, it will be below 0.5 USD/kg for the mentioned processes.

KPIs 12, 13, and 14 (Figure 4.21, Figure 4.22, Figure 4.23) are showing the total costs which is a part of the OPEX in Middle East in three different scenarios: reagent supplied from East Asia, the US, and the EU. The reason to mention that the numbers are part of the OPEX is that there are more costs to be considered in the OPEX such as the maintenance of the plant, the transportation costs for inputs and outputs of the system, and manpower costs which have not been taken into account in the current study. The reason is that these costs include many uncertainties and ambiguities which need to make many assumptions. These assumptions decrease the accuracy of the results in the study. Hence, it was decided that it is worth to have more accurate results for limited parameters than inaccurate and uncertain results for numerous parameters. As shown in the above-mentioned figures, process 4 has the highest total costs per mass of captured CO₂ and if the reagent is supplied from the EU, the total costs are considerably higher for all processes.

Figure 4.24, Figure 4.25, Figure 4.26, Figure 4.27, Figure 4.28, and Figure 4.29 (KPIs 15, 16, 17, 18, 19, and 20) show the same logic if the plant is located in Norway and in the US and the results have the same trend as KPIs 12, 13, and 14.

Comparing the total costs per mass of captured CO₂ of Middle East, Norway, and the US regardless of the origin of the reagent, Norway has comparably lower total costs per mass of captured CO₂ and the reason is the lower price of electricity in this country.

Looking into the KPI21 and Figure 4.30, it is not surprising that the revenue from sales of CaCO₃ is the same for each process since the captured CO₂ is the same in all processes and the amount of CaCO₃ will be the same too.

KPIs 22, 23, and 24, represented by Figure 4.31 to Figure 4.39, depict the yearly profit of each process in Middle East, Norway, and the US each one with three different origins for the reagent to be supplied from and per mass of captured CO₂. As is obvious, process 4 has minus profit in all cases meaning that this process is not profitable. The reason is that its total costs are much more than its revenue. Even in the US, where the captured CO₂ has a tax income added in the revenue, process 4 is still unprofitable.

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The rest of the processes are profitable in all the cases. Interestingly, the amounts of the profits for each process in each country are quite close to each other when the country of origin for the reagent is the same. Choosing process 2 in Middle East, Norway, and the US as an example, the yearly profit per mass of captured CO₂ is 0.56 USD/kg in Middle East, 0.57 USD/kg in Norway, and 0.56 USD/kg in the US if the reagent is supplied from East Asia.

Although there is a tax income in the US for the captured CO₂, this will not change the results considerably since the electricity cost in the US is much higher compared to Norway and Middle East.

5 Conclusion and recommendation for further investigation

In this thesis, an in-depth study of the literature between 2020 and 2024 about the aqueous indirect mineralization of CO₂ was conducted, whereby twenty-seven processes which were studied in the laboratory were recognized and studied in detail. The details of these processes were gathered in a table providing a holistic overview of each process and the values of the parameters impacting the process.

Then, out of the processes in the literature, four of them namely process 1, 2, 3, and 4 which were using different leaching reagents, industrial wastes, and operational conditions were chosen for scale-up calculations.

Using the reactions ruling the processes, the results from the laboratory work in the papers, and the necessary assumptions and simplifications, the scale-up mass balance calculations were conducted for each of four processes and the flowrates of the streams and the required mass of the reagents in the processes were calculated.

Using the flowrates and by recognizing the nature of the processes and their probable needs, the required equipment such as tanks, reactors, pumps, etc. were chosen and sized, and PFDs of each process were designed.

Based on the PFDs, knowing the masses and flowrates and operational temperatures of each process, and making some assumptions due to lack of some information such as head loss in the system, the energy balance calculations were performed and the energy consumption of each equipment and the whole process for all four processes were calculated.

After that, three countries (Middle Eastern countries, Norway, and the US), each one as a representative of a region in the world, were chosen as the probable location of the plant to study its economy and environmental impact.

Using the data of energy consumption, the amount of produced CO₂ by each process in each of the mentioned countries was calculated to see the environmental impact of the processes and their CO₂ footprint.

Next, by assuming three different areas namely East Asia, the US, and the EU as the supplying origin of the reagents and by finding the electricity cost in the countries mentioned as the location of the plant, the total costs of the processes (part of OPEX) in different scenarios were calculated and by finding the price of the produced CaCO₃ and the tax income of capturing CO₂, the revenue of the processes in the same scenarios was calculated making it possible to see if they are profitable.

Finally, some KPIs were defined and calculated based on the results from the above-mentioned calculations to compare the four processes.

Based on the KPIs, process 2 is the best in terms of capturing CO₂ per mass of waste (0.33 kg/kg). Process 4 and 3 are in the second and third place with 0.28 and 0.25 kg/kg and process 1 is the last one with 0.15 kg/kg. In terms of the produced CaCO₃ per mass of waste, the trend

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is the same with 0.75 kg/kg, 0.65 kg/kg, 0.58 kg/kg, and 0.34 kg/kg for processes 2, 4, 3, and 1, respectively.

Considering the consumption of reagent per mass of captured CO₂, process 2 has the lowest reagent consumption with 0.5 kg/kg and process 4 owns the highest with 3.1 kg/kg.

Talking about the energy consumption per mass of captured CO₂, the best process is process 1 with 1.48 kWh/kg and the second-best process is process 2 with 1.8 kWh/kg.

The produced CO₂ per mass of captured CO₂ by the processes is different in each country from less than 0.08 kg/kg for all four processes in Norway to even more than 1 kg/kg for some of the processes in Middle East and the US. But in each country, the highest amount belongs to process 3 and the lowest amount belongs to process 1.

Finally, considering Middle East, Norway, and the US as the location of the plant and assuming East Asia, the US, and the EU as the origins of the reagents to be supplied from, all of the processes are profitable in a year except process 4 which has always more costs than revenue making it an unprofitable process.

Recommendation for further investigation

There are always some assumptions and simplifications in each study due to the lack of sufficient supporting data, time restrictions of the study, or to reduce the degrees of freedom which make the calculations and simulations more complicated and reduce the accuracy of the results. These assumptions and simplifications could be considered as areas for further analysis and investigation.

This study is not an exception too. There are some inputs for the calculations such as the conversion of Ca, the purge percentage, the head loss for the pumps, the specific heat capacities, the heat transfer coefficient, densities, the effect of agitation speed etc. which have been assumed or neglected, but their effect on the mass, energy, and economic results could be investigated more through sensitivity analysis if being supported by real data.

Moreover, when it comes to economic assessment and cost calculations, there are some simplifications in OPEX calculations that have ignored a part of OPEX such as the manpower costs, maintenance costs, and logistics costs for which it was possible to find data, but it could result in inaccurate outcomes since it was based on assumptions. These could also be an area for further studies.

Finally, if any of the four processes are to be chosen for making a real plant, a multi-criteria decision analysis/ making (MCDA, MCDM) could be conducted based on the KPIs.

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Appendices

Appendix 1: Industrial waste characteristics and pretreatment

Paper No.	Metal	Source	Composition	Grinding/Milling	Particle Size
1 [60]	Mg, Ni	Laterite Ore Copperas	MgO: 37.11%,	Yes, crushed	less than 150 micrometer
2 [61]	Ca	Biomass ash	CaO: 36.7%	N/A	N/A
3 [62]	Ca	Kambara reactor desulphurization slag	CaO: 72.04%	Crushed	74-150 micrometer
4 [63]	Ca	Steel slag	CaO: 45.4%	Crushed and ground	38-75 micrometer
5 [64]	Ca	BOF LFS EAFS	BOF. CaO: 46.2% LFS. CaO: 48.29% EAFS. CaO: 40.8%	N/A	50-74 micrometer
6 [46]	Ca	BFS	CaO: 39.74%	N/A	N/A
7 [65]	Ca	Coal fly ash	Ca: 3.44%	N/A	N/A
8 [66]	Ca	Calcium carbide slag	N/A	N/A	125 micrometer
9 [67]	Mg	Serpentine	N/A	N/A	less than 75 micrometer
10 [68]	Ca	Ladle furnace slag	CaO: 46.73%	Ground	68 micrometer
11 [69]	Ca, Mg	Iron ore mining waste	Ca: 1.45%, Mg: 0.63%	Crushed and sieved	less than 10 micrometer
12 [70]	Ca	Basic oxygen furnace slag (BOF)	CaO: 40%	N/A	17.3 micrometer
13 [71]	Ca	Steel slag	CaO: 46.7%	Crushed and sieved	Less than 53 micrometer
14 [72]	Ca	Calcium silicate	N/A	N/A	less than 20 micrometer
15 [73]	Ca	Phosphogypsum (PG)	N/A	Ground	less than 150 micrometer

16 [74]	Ca	Paper sludge ash (PSA)	Ca: 67.2%	Sieved	Avg: 17.5 micrometer
17 [75]	Ca	BOF	CaO: 37.68%	No grinding to reduce energy consumption.	75 to 4000 micrometer
18 [76]	Ca	Biomass bottom ash (BBA) Biomass fly ash (BFA) Fly ash from refuse-derived fuel incineration (RFA) Stainless steel slag (SSS)	BBA: 23.3% Ca BFA: 16.1% Ca RFA: 23.5% Ca SSS: 43.2% Ca	N/A	N/A
19 [77]	Ca	Grits, made during the kraft pulp production process	Ca: 346200 mg/kg	N/A	Less than 0.5 mm
20 [78]	Ca	BFS	CaO: 40.56%	Ground	Less than 150 micormeter
21 [79]	Ca	CaO	N/A	N/A	N/A
22 [80] [81]	Ca	CKD	CaO: 46.4%	No need	23.6 micrometer
23 [82]	Ca	Red mud	CaO: 14.21%	N/A	N/A
24 [83]	Ca	Recycled concrete aggregate (RCA)	CaO:27.8%	N/A	Less than 4 mm
25 [84]	Mg and Fe	Mining residue	Fe ₂ O ₃ : 22.9%,MgO: 9.3%	Samples blended	65 micrometer
26 [85]	Ca	Waste concrete	Ca: 59.4%	N/A	less than 150 micormeter
27 [45]	Ca	Recycled concrete fines (RCF)	CaO: 71.11%	Crushed and ground by ball mill	less than 75 micrometer

Appendix 2: Leaching process details

Paper No.	Reagent	Concentration	Solid/Liquid	LT	LP	Leaching agitation	Leaching Time	Leaching Efficiency/Capacity
1 [60]	Distilled water	N/A	1:10	80 C	Ambient (N/A)	N/A	60 min	86% and 82% for Mg and Ni
2 [61]	Glycine	2 mol/L	300 g/L	25 C	Ambient	80 rpm	60 min	19.90%
3 [62]	NH ₄ Cl	0.5 mol/L	5 g per 500 mL	25 C	Ambient (N/A)	500 rpm	120 min	89.76%
4 [63]	NH ₄ Cl HCl	2 mol/L	5 g per 100 mL	25 C	Ambient (N/A)	500 rpm	30 min	appx. 28%
5 [64]	NH ₄ Cl CH ₃ COOH	1 mol/L	2 g per 250 mL	15 C	Ambient (N/A)	500 rpm	120 min	BOF in NH ₄ Cl: 68% LFS and EAFS in NH ₄ Cl <10% BOF in CH ₃ COOH: 86% EAFS in CH ₃ COOH: 52% LFS in CH ₃ COOH: 61%
6 [46]	HCl	4 mol/L	10 g per 125 mL	80 C	Ambient (N/A)	600 rpm	120 min	91%
7 [65]	HNO ₃	0.3 mol/L	20 g per 400 mL	25 C (N/A)	Ambient (N/A)	350 rpm	150 min	72.60%
8 [66]	Ammonium acetate	1 mol/L	N/A	40 C	Ambient (N/A)	600 rpm	30 min	79.20%
9 [67]	NaHSO ₄	1.4 mol/L	50 g/L	70 C	Ambient	N/A	60 min	48%
10 [68]	NaHSO ₄	110 g/L	1 g per 7 mL	25 C	Ambient (N/A)	450 rpm	90 min	N/A
11 [69]	Oxalic acid Formic acid Acetic acid Hydrochloric acid	Mentioned for leaching efficiency.	15 g per 150 mL	25 C	Ambient	Orbital shaker, 220 rpm	100 min	Oxalic acid. 16% Ca at 0.25 mol/L acid 54% Mg at 0.25 and 0.75 mol/L acid HCl. 98% Ca at 1.5 mol/L acid 6.8% Mg at 1 mol/L acid Formic acid. Appx 25% Ca at 1.5 mol/L acid Appx 1.5% Mg at

								1.5 mol/L acid Acetic acid. 20% Ca at 1.5 mol/L acid 3.5% Mg at 1.5 mol/L acid
12 [70]	NH ₄ Cl	2 mol/L	15 g per 300 mL	80 C	Ambient (N/A)	750 rpm	10 min	60.30%
13 [71]	CH ₃ COOH	1 mol/L	5 g per 100 mL	25 C	Ambient (N/A)	500 rpm	30 min	Original slag: 59.28% Processed slag: 84.46%
14 [72]	N,N-dicarboxymethyl glutamic acid (GLDA)	0.1 mol/L	4.64 g per 100 mL	50 C	Ambient (N/A)	Yes	20 min	24.04%
15 [73]	NaCl solution	3 mol/L	1 g per 50 mL	30 C	Ambient (N/A)	Yes	60 min	49.42%
16 [74]	Citrate IDA	0.1 mol/L 0.5 mol/L	1g per 50 mL	25 C	Ambient	250 rpm	60 min	23.5% for Citrate 20.4% for IDA
17 [75]	NH ₄ NO ₃ NH ₄ Cl	3 mol/L	1:10 mass ratio	25 C	Ambient (N/A)	400 rpm	90 min	75% for NH ₄ NO ₃ 69% for NH ₄ Cl
18 [76]	HNO ₃	0.1 mol/L	5 g per 50 mL	25 C (N/A)	Ambient (N/A)	Stirred	1440 min	N/A
19 [77]	CH ₃ COOH	2 mol/L	30 g per L	45 C	Ambient (N/A)	N/A	120 min	77% for CH ₃ COOH
20 [78]	H ₂ SO ₄	2 wt%	1:2 mass ratio	60 C	Ambient (N/A)	N/A	40 min	100%
21 [79]	Artificial sea water and sucrose (1:2 molar ratio of sucrose to Ca)	N/A	1 g per 50 mL	25 C (N/A)	Ambient (N/A)	250 rpm	60 min	Maximum (appx 7000 mg/L)
22 [80] [81]	Citrate Malonate Adipate	0.1 mol/L	Molar. 1:1 Ca and citrate 1:2 Ca and malonate 1:2 Ca and adipate	23 C	Ambient	250 rpm	60 min	42% for citrate 23.1% for malonate 20.8% for adipate
23 [82]	HCl	1 mol/L	21 g RM per 300 mL	80 C	Ambient	600 rpm	120 min	85%
24 [83]	NH ₄ NO ₃	2 mol/kg	S/L: 20%	25 C	Ambient (N/A)	N/A	180 min	appx 90%

25 [84]	NH ₄ HSO ₄	1.5 mol/L	40 g per 200 mL	61 C	Ambient (N/A)	250 rpm	120 min	28 % Fe 41 % Mg
26 [85]	HCl	0.5 mol/L	S/L. 1:15 (w/v)	25 C	Ambient	N/A	30 min	94%
27 [45]	NH ₄ Cl	2 mol/L	1 g per 10 mL	85 C	Ambient (N/A)	500 rpm	60 min	65.70%

Appendix 3: Mineralization process details

Paper No.	Reactor type	Mineralization agitation	Mineralization time	Mineralization gas type, concentration and flow rate	MT	MP	Mineralization product and its purity	Mineralization efficiency/capacity	Reagent recovery/recycling
1 [60]	Batch with stirring	Yes	120 min	(NH ₄) ₂ CO ₃ as CO ₂ source	30 C	Ambient (N/A)	MgCO ₃ and NiSO ₄	291 kg CO ₂ per ton laterite ore	N/A
2 [61]	Glass bubble column with stirring	100 rpm	60 min	Humidified CO ₂ at 100 ml/min	25 C	Ambient	Vaterite CaCO ₃	76.80%	N/A
3 [62]	Batch	300 rpm	120 min	CO ₂ at 15 Ml/min	40 C	Ambient (N/A)	99% calcite and vaterite	396 kg CO ₂ per ton slag 794 kg calcite and vaterite per ton of slag	N/A
4 [63]	Batch	N/A	20 min	CO ₂ at 0.1 L/min	25 C (N/A)	Ambient (N/A)	Calcite CaCO ₃	appx. 75%	N/A
5 [64]	N/A	N/A	N/A	N/A	25 C (N/A)	Ambient (N/A)	N/A	N/A	N/A
6 [46]	Batch	Ultrasonic with power of 200 W	15 min	CO ₂ at 1 L/min and concentration of 70%	20 C	Ambient (N/A)	90% Vaterite CaCO ₃	N/A	Yes
7 [65]	Batch	400 rpm	30 min	Na ₂ CO ₃ as CO ₂ source	25 C (N/A)	Ambient (N/A)	94.6% calcite CaCO ₃	Ca conversion : 97.3% CO ₂ conversion: 92.9% 11k g CO ₂ absorbed by 1 ton fly ash	Yes
8 [66]	Batch	Stirred	N/A	CO ₂ at 500 mL/min	25 C	Ambient (N/A)	98% single vaterite phase	CO ₂ uptake 23.5% in slag	N/A

9 [67]	Batch	800 rpm	N/A	Na ₂ CO ₃ as CO ₂ source	80 C	Ambient (N/A)	MgCO ₃	CO ₂ sequestration capacity of 77.4% Carbonation efficiency: 92%	Yes
10 [68]	N/A	600 rpm	120 min	CO ₂	25 C	Ambient (N/A)	CaCO ₃	Sequestration: 9.3%	Yes
11 [69]	Batch	250 rpm	60 min	CO ₂ 99.8% pure	25 C	Ambient	CaCO ₃ and MgCO ₃	131.58 kg CO ₂ in 1 ton waste	N/A
12 [70]	Batch	N/A	10 min	CO ₂ at 0.5 L/min	25 C (N/A)	Ambient (N/A)	98% homogeneous vaterite	99.5% carbonation ratio 420 kg CaCO ₃ and 184 kg CO ₂ per ton of BOFS	Yes
13 [71]	N/A	N/A	N/A	N/A	25 C (N/A)	Ambient (N/A)	N/A	N/A	N/A
14 [72]	Batch	N/A	70 min	Na ₂ CO ₃ as CO ₂ source at 0.3 mol/L	80 C	Ambient (N/A)	Aragonite and calcite	Carbonation ratio: 72.5%	Yes
15 [73]	N/A	N/A	60 min	CO ₂ at 80 mL/min	30 C	Ambient (N/A)	Pure calcite or vaterite	Carbonation ratio: 96.31% 115 kg CO ₂ and 262 kg CaCO ₃ per ton of PG	Yes
16 [74]	N/A	300 rpm	Until PH = 7	99.9% pure CO ₂ at 0.1 L/min	25 C	Ambient	Citrate: 80.1 % pure calcite IDA: 95.4 % pure calcite	Citrate: 142.2 kg CO ₂ and 323.2 kg CaCO ₃ per ton PSA IDA: 99.4 kg CO ₂ and 225.8 kg CaCO ₃ per ton PSA	N/A
17 [75]	Batch	N/A	15 min	CO ₂ at 150 cm ³ /min	25 C	6.5 bar	99.8% pure CaCO ₃	CaCO ₃ yield: 80%	Yes

18 [76]	Batch	N/A	10080 min	CO2	25 C (N/A)	Ambient (N/A)	N/A	BBA: 2.11 mol CO2/kg BFA: 0.99 mol CO2/kg RFA: 1.74 mol CO2/kg SSS: 2.54 mol CO2/kg	N/A
19 [77]	Batch	N/A	N/A	Pure CO2	30 C	30 bar	Mainly calcite and aragonite	Carbonation efficiency: 74% 460 kg CO2 per ton grits	N/A
20 [78]	Batch	N/A	N/A	CO2	25 C (N/A)	Ambient (N/A)	CaCO3	36 kg per ton of BFS	Yes
21 [79]	Batch	200 rpm	N/A	99.9% pure CO2 at 0.15 L/min	25 C (N/A)	Ambient (N/A)	94.6% content of vaterite under 2.4 micrometer.	N/A	N/A
22 [80] [81]	Batch	250 rpm	when PH = 10.5	Both pure and mixed CO2	23 C	Ambient	98% pure calcite	88.9% carbonation and 430 kg/t CaCO3 for citrate 95.5% carbonation and 255 kg/t CaCO3 for malonate 96.6% carbonation and 232 kg/t CaCO3 for adipate	Yes
23 [82]	Batch	250 rpm	N/A	Flue gas: 10% CO2 and 90% N2 Na2CO3 used as CO2 source at 20 mL/min	25 C	Ambient (N/A)	98% pure CaCO3	Carbonation efficiency= 32.71% 34 kg CO2per ton red mud	Yes

24 [83]	N/A	N/A	N/A	N/A	25 C (N/A)	Ambient (N/A)	N/A	39 kg CO2 per ton RCA	Yes
25 [84]	Batch	250 rpm	120 min	Flue gas: 10% CO2	80 C	Ambient (N/A)	Carbonates	110 kg CO2 per 1 ton residues	N/A
26 [85]	Batch	N/A	30 min	CO2 at 0.1 L/min	25 C	Ambient	99% pure CaCO3	CaCO3 synthesis efficiency: 96%	N/A
27 [45]	Batch	N/A	30 min	N/A	25 C	0.025 Mpa	97.8% pure vaterite	270 kg CO2 and 615 kg vaterite per ton of RCF	Yes

FMH606 Master's Thesis

Title: CO₂ capture through mineralization of CaO-containing by-products

USN supervisor: Lars-André Tokheim

External partner: Caox (Milana Ayzenberg)

Task background:

The global antropogenic CO₂ emissions are currently 37 Gt/y, and the CO₂ concentration in the atmosphere is increasing. A reduction of at least 40 % is required not to increase the global mean temperature by more than 1.5 °C.

45 % of the global emissions are due to energy production in thermal power plants, 8 % comes from cement production and 8 % originates from steel production. Hence, reducing the CO₂ emissions from these sectors will make a significant contribution to reaching the climate goals.

Some industrial by-products/waste streams contain CaO. Such streams may be used for CO₂ capture through mineralization ($\text{CaO} + \text{CO}_2 \rightarrow \text{CaCO}_3$). In some cases, the carbonated reaction product may be applied as a useful product/additive in other processes. This concept can be seen as a path towards a more circular economy.

Large amounts of CaO-rich materials will be required to capture CO₂. Examples of such materials are fly-ash, cement kiln dust and blast furnace slag. Lab research has been conducted in the field, and several publications on mineralization are available. However, it appears that more knowledge is required, in particular the enrichment part of process.

Task description:

The task may include some of the following sub-tasks (subject to change based on discussions between the student, the supervisor and the external partner):

- Make a literature study on experience from application of CaO-rich by-products for mineralization
- Study in detail how CaO may be enriched based on raw waste streams
- Describe the composition of the enriched stream
- Quantify how much CaO-rich waste is required per ton of captured CO₂
- Make a mass and energy balance of a pilot-scale process
- Make a process flow diagram for the pilot-scale process
- Simulate the pilot-scale process
- Describe the logistics of an industrial plant

The results from this thesis will be used by Caox (<https://caox.no/index.html>) in their technology development process.

Student category: PT or EET

Is the task suitable for online students (not present at the campus)?: Yes


Practical arrangements:

The project will be conducted in close cooperation with the technology teams at Caox (Bergen, Norway), the University of Newcastle upon Tyne, UK, and industrial partners. During the project, you will be part of an international cross-disciplinary team with the opportunity to learn and contribute to the deployment of technology for CO₂ capture and mineralization from hard-to-abate industries. The team will provide support throughout the project. Teams/Zoom will be used for project meetings.

Supervision:

As a general rule, the student is entitled to 15-20 hours of supervision. This includes necessary time for the supervisor to prepare for supervision meetings (reading material to be discussed, etc.).

Signatures:

Supervisor (date and signature): 31 January, 

Student (write clearly in all capitalized letters): AMIRHOSSEIN GHAZI

Student (date and signature): 11 Feb. 