Journal of CO2 Utilization 94 (2025) 103063

Contents lists available at ScienceDirect

Journal of CO2 Utilization

journal homepage: www.elsevier.com/locate/jcou

Review Article

Carbon dioxide utilization in mineral processing

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ARTICLE INFO

Keywords:
CO_2 utilization
Leaching
Floatation
Mining wastewater treatment
Mineral carbonation

ABSTRACT

Mining and mineral processing, significant contributors to CO_2 emissions, can reduce their carbon footprint and support a circular economy by integrating CO_2 into various unit operations, offering a promising way to mitigate emissions while enhancing mineral recovery and waste management. This review examines CO_2 utilization in leaching, flotation, wastewater treatment, and CO_2 sequestration via mineral carbonation using tailings. Emerging CO_2-augmented leaching techniques, including CO_2-water, supercritical CO_2, CO_2-ammonia, ionic-liquid, and CO_2-cyanide leaching have shown improved performance for a limited number of minerals at the laboratory scale. Similar to its role in leaching, CO_2 can act as a depressant or pH modifier in flotation, improving process selectivity and efficiency while reducing the need for additional chemicals. For water treatment, CO_2 facilitates the precipitation of heavy metals and contaminants under optimal conditions. CO_2 sequestration through mineral carbonation provides a sustainable method for locking CO_2 in stable mineral forms, utilizing tailings for both waste management and carbon capture. CO_2 utilization in flotation, leaching, and water treatment is currently limited to lab-scale experiments, emphasizing the need for scale-up trials to fully assess technical feasibility and identify potential bottlenecks associated with large-scale implementation. This review also identifies key challenges, including process optimization, scalability, and the need for further technological advancements. Future research should address these barriers to enable the practical integration of CO_2 utilization into mineral processing operations.

1. Introduction

The far-reaching effects of anthropogenic CO_2 emissions on environmental and human health are widely recognized and experienced globally, providing an impetus to mitigate their impacts through various approaches, including reforestation, the use of alternative renewable energy sources, process efficiency improvement, and carbon capture and utilization [1–3]. Early industrial efforts focused on carbon capture and storage (CCS), which mainly comprises storing captured gas in underground geological structures under high temperatures and pressures [4–6]. The buildup of pore pressure is an inevitable geomechanical risk that may trigger the release of CO_2 back into the atmosphere through processes such as caprock failure, reactivation of existing faults, and loss of integrity [7]. However, this approach has since been transformed into relatively more sustainable carbon capture, utilization, and storage (CCUS) to mitigate some of the limitations of the former approach.

Although the mining industry and its auxiliary services contribute at most 7 % of global greenhouse gas (GHG) emissions [8], they are strategically positioned as key players in reducing CO_2 emissions through the CCUS. The mineral processing industry can reduce CO_2 emissions through simultaneous CO_2 utilization in its unit operations and CO_2 sequestration via mineral carbonation. If net-zero CO_2 emissions can be achieved, the industrial sector may become a major contributor to emission trading schemes, eventually becoming a designated CO_2 sink. Therefore, it is useful to assess the progress of the mineral processing industry in terms of CO_2 utilization and sequestration.

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https://doi.org/10.1016/j.jcou.2025.103063
Received 28 August 2024; Received in revised form 30 October 2024; Accepted 10 March 2025
Available online 17 March 2025

Carbon dioxide can be incorporated into leaching, wastewater treatment, and flotation processes as an environmentally benign alternative pH regulator because of its self-buffering effect, which yields precise pH control. In the flotation process, CO_2 can also play a versatile role, depending on the process requirements. For example, it can be used as a bubbling gas, depressant, activator of hydrophilic ores, conditioner, and flocculant, which promote fine particle aggregation [9–11]. CO_2 has been demonstrated to be a highly selective leaching agent because of its high diffusivity, which promotes mass transfer and reaction kinetics while easing the demand for downstream treatment of waste streams [12,13]. When used for treating wastewater produced during mining activities, CO_2 can precipitate certain cations in water as carbonates and bicarbonates [14,15]. One common feature of these processes is that the incorporation of CO_2 results in concomitant mineral carbonation; therefore, in addition to these three processes, mineral carbonation is also reviewed.

The mining industry generates large amounts of mining tailings as waste with appropriate mineralogy and high surface area for enhanced carbon capture [16,17].These vast quantities of tailings are generated from the mineral processing of metallic, diamond, and refractory mineral ores worldwide and can capture considerable amounts of CO_2 from the atmosphere. The benefits of storing carbon dioxide in mine tailings compared with geological formations include lower operating temperatures and pressures, safe storage because of fast reactions, and stable products [18–20]. Thus, mine tailings exhibit a huge potential to supplement geological formations in carbon storage and mitigate challenges related to insufficient storage space and the need for constant monitoring [21].

Recently, many reviews and studies have discussed the utilization and sequestration of CO_2 in mining-related operations. Cheng et al. [22] carried out a two-step leaching and carbonation of activated calcium (Ca) under different conditions to improve the leaching, carbon sequestration and reuse of steel slag. It was observed that the procedure increased the leaching rate by 26.9 % and achieved a capacity of 223.15 kg CO_2/t SS in comparison to conventional methods. Liu et al. [23] studied a path of CO_2 sequestration that offers a solution to low leaching rates by leaching microwave-pretreated magnesium tailings that were impregnated and calcinated. Results showed that pretreatment with impregnation and calcination and an external field has the capacity to promote the leaching process. The CO_2 heterophase absorption and sequestration method effectively fixes CO_2 from the gas phase to the solid phase under normal temperature and pressure conditions, producing a stable carbon sequestration product, $MgCO_3$•3 H_2O, with long-term stability. A novel approach for wastewater treatment that concentrates nutrients via a membrane with a carbonated solvent containing captured CO_2 gas was utilized in forward osmosis as a forward solution to concentrate the nutrient stream as well as to regulate the pH [24].

Wang, [25] conducted a comparative study on the disadvantages and advantages of gas-solid and aqueous carbonation for carbon capture and sequestration, focusing on the reaction kinetics, recovery, and capture efficiency. It was deduced that direct solid-gas carbonation has higher reaction rates and capture efficiencies, whereas direct aqueous carbonation requires less energy. Lin et al. [26] reviewed advancements in mineral carbonation and concluded that magnesium- and calcium-based feedstocks, primarily from industrial waste and mining tailings, are the most studied in this field. They also observed that both direct and indirect carbonation methods hold considerable potential. In an experimental study, Gomari et al. [27] examined the use of steel waste from blast furnaces and ladle slags to directly capture carbon from air and sequestrate it through mineral carbonation. The operational parameters investigated were temperature, reaction time between slag waste and CO_2, pressure and liquid-solid ratio. Results showed that higher CO_2 was sequestrated by ladle slag than by blast furnace slag, with a maximum sequestration of 30 % and 24 % achieved at 90 degrees celsius. Ndlovu et al.[28] detailed advances in mineral carbonation, noting that basalt and ultramafic rock are highly effective for natural carbonation. Moreover, the carbonation of solid waste emanating from the construction industry, mining, and sludge from wastewater with high MgO and CaO concentrations occurring through direct and indirect routes is a promising route for CO_2 sequestration. Pan et al. [29] reviewed the global uptake of CO_2 mineralization and the use of various alkaline solid wastes. It was reported that approximately 4.02Gt of CO_2 could be manged per year through mineralization process leading to 12.5 % reduction in the CO_2 emissions. The main challenges highlighted were the slow kinetics, lack of comprehensive studies on the reaction kinetics, and low carbonation capacity of the feedstocks. Additionally, optimizing the efficiency of the carbonation process is essential, and enhancements, such as microwave radiation and ultrasonic treatment, are essential to improve outcomes. It is important to recognize that high costs significantly hinder the development of this process. Another challenge is that most studies are conducted in laboratories, creating a gap between laboratory results and industrial applications, which hampers the adoption of such methods. Furthermore, many studies utilize pure CO_2, while the research aims to work with flue gas, leading to different reaction dynamics due to the presence of additional components in flue gas.

This review serves as a consolidation of research efforts toward CO_2 incorporation into various unit operations in the mineral processing industry, along with the application of mine tailings in CO_2 sequestration. Additionally, the review focuses on leaching, flotation, and mineral wastewater treatment, which are not as extensively researched as mineral carbonation, examining various findings from publications to illustrate how CO_2 has been utilized in these processes, with a specific emphasis on the positive outcomes achieved in each area. Each section has a dedicated section outlining the research gaps and future directions of springboard innovative net-zero carbon emissions initiatives, aligned with the 2015 Paris Agreement target of reaching net-zero CO_2 emissions by 2050. The current state of the published work indicates that CO_2 use in mineral processing unit operations is still in its infancy, and several factors should be addressed to realize the efficient utilization of CO_2.

2. CO_2 in mineral leaching processes

Leaching processes continue to play a pivotal role in the liberation of valuable metals and minerals from ores for various applications. Leaching involves dissolving target components from solid materials (ore) into a liquid phase, typically using solvents and/or reagents [30]. The commonly used leaching processes are heap leach and tank leach. Fig. 1 shows a schematic of the leaching process. Tank leaching is usually conducted in the presence of adsorbents such as activated carbon for gold; hence, the term “carbon in pulp (CIP) leaching” or “carbon in leach (CIL)”.

The leaching method is extensively employed in hydrometallurgical extraction processes to extract minerals of interest, such as gold (Au) [31], copper (Cu) [32], uranium (U) [33], rare earth elements (REEs) [34], and nickel (Ni) [35]. Traditionally, leaching operations rely on highly reactive and dangerous conventional solvents or reagents such as cyanide (CN-), phosphoric acid (H_3PO_4), thiosulfate ($S_2O_3^{2-}$), sulfuric acid (H_2SO_4), nitric acid (HNO_3), and hydrochloric acid (HCl) to facilitate the dissolution of desired minerals [36]. Although effective, conventional approaches often have significant environmental and safety concerns, including the release of toxic by-products and generation of hazardous waste streams. In recent years, interest in exploring alternative leaching strategies has increased. The aim is not only to improve the efficiency and selectivity of mineral extraction but also to minimize the environmental footprint and health effects, as well as to improve the safety of the process, particularly with respect to the reagents. One promising approach is the use of CO_2 in leaching processes.

Primarily known for its role in climate change as a greenhouse gas (Fig. 2), CO_2 is gaining attention as a potentially versatile and

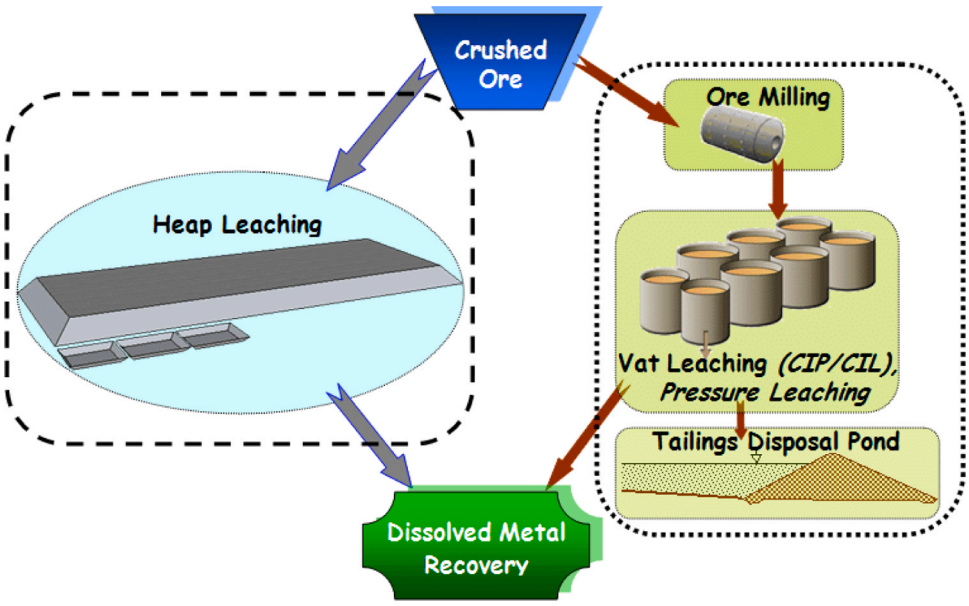

Fig. 1. Schematic of the leaching process.

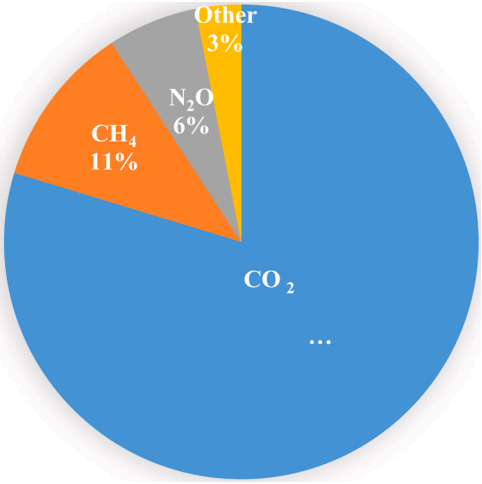

Fig. 2. Overview of greenhouse gas emissions [198].

environmentally benign reagent for various industrial applications, including leaching in mineral processing. CO_2 can be utilized for various applications, such as leaching agents, enhanced oil recovery, pH modifiers, mineral carbonation, and solubility enhancers for targeted minerals [37,38]. The incorporation of CO_2 into the leaching process offers several potential benefits: (i) CO_2 is a natural gas, is abundant, and inexpensive, and can be sourced as a by-product in industrial processes such as power generation and chemical manufacturing [39]. (ii) CO_2-based leaching methods can potentially reduce reliance on hazardous chemicals, thereby minimizing the environmental risks and regulatory burdens associated with conventional leaching agents [40]. Moreover, CO_2 use during leaching can contribute to the overall reduction in carbon emissions by sequestering CO_2 into mineral products or converting it into stable carbonate compounds [40].

Although the concept of taking advantage of CO_2 in leaching processes is still emerging, recent research efforts have shown promising results, demonstrating the feasibility and efficacy of CO_2-based leaching techniques across various mineral systems. However, significant challenges remain, including optimizing the process parameters, addressing scalability issues, and ensuring economic viability on an industrial scale [41]. Arce et al. (2017) investigated the effect of physicochemical properties of Brazilian serpentinites on the leaching process of indirect CO_2 mineral carbonation. This study found that samples with higher concentrations of reactive minerals, such as olivine, exhibited greater leaching efficiency, leading to higher CO_2 sequestration potential. Moreover, finer grain sizes and increased surface area enhance the leaching process by providing more contact points for chemical reactions [42]. Gao et al. (2017) studied the removal of sodium (Na) and calcium (Ca) from Zhundong coal by a novel CO_2-water leaching method. It was found that CO_2-water leaching effectively removed both Na and Ca with improved efficiency compared to water leaching. Higher efficiency was influenced by parameters such as leaching time, temperature, and CO_2 pressure. They further studied the behavior of the leached coal, where they found that its deformation temperature was as high as 1202 °C, higher than those of the raw coal and water leached coal ash, which were 1098 and 1142 °C, respectively. In addition, CO_2 leaching significantly inhibited the agglomeration and sintering of coal ash. Fig. 3 shows SEM images of coal ash samples prepared at different ashing temperatures [43].

In this section, we provide an overview of the advances and current state of research on CO_2 utilization in leaching processes for mineral extraction. We explored the underlying mechanisms/techniques, potential applications and utilization, environmental implications, and future prospects of CO_2-based leaching techniques, drawing insights from recent advances in the field.

2.1. *CO_2 as a primary leaching agent and CO_2 enhanced leaching*

CO_2 is used in traditional leaching methods across various industries owing to its unique chemical properties and environmental advantages. CO_2 is used to enhance leaching processes via various factors, such as i) pH adjustment and acidic leaching by dissolving CO_2 in water to form carbonic acid (H_2CO_3), a weak acid that adjusts and maintains the pH of the leaching solution (acidic conditions are favorable for leaching). This enhances the solubility of the target components in the ore matrix, thereby facilitating their extraction [44]. ii) Selective leaching by controlling factors such as pH and temperature: CO_2 can selectively target specific components while leaving others unaffected. This selectivity enables the extraction of desired elements with minimal interference from unwanted impurities, thereby improving the purity and yield of the desired products [45]. iii) Formation of carbonate complexes, in which

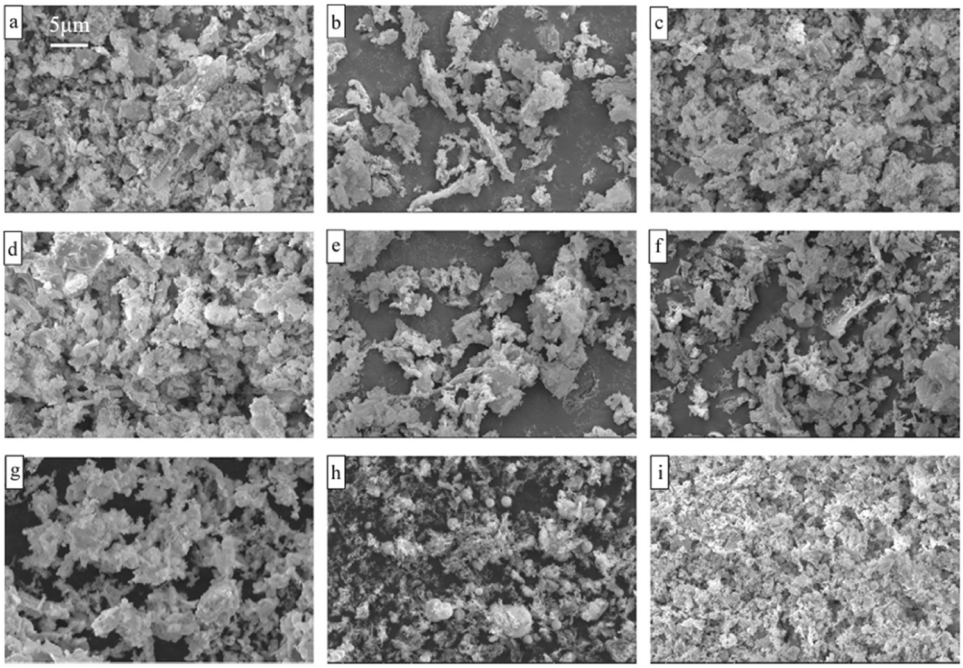

Fig. 3. SEM micrographs of coal ash prepared at different temperatures. a) Raw coal ash at 500 °C. b) Water leached coal ash at 500 °C. c) CO_2-water leached coal ash at 500 °C. d) Raw coal ash at 700 °C. e) Water leached coal ash at 700 °C. f) CO_2-water leached coal ash at 700 °C. g) Raw coal ash at 900 °C. h) Water leached coal ash at 900° C. i) CO_2-water leached coal ash at 900 °C. Reproduced with permission from Elsevier [43].

carbonate ions (CO_3^{2-}) are formed by the reaction of CO_2 with water and act as complexing agents for the metal ions. These carbonate complexes increase solubility, enhance leaching, and promote rapid separation from gangue minerals [46].

Furthermore, CO_2 serves as a beneficial supercritical fluid for leaching applications owing to its unique properties, which include high diffusivity, low viscosity, and adjustable solvation power. Supercritical CO_2 leaching offers several advantages over traditional leaching methods, such as minimal environmental impact, reduced energy consumption, and improved selectivity. Cui et al. (2017) designed a reactor (Fig. 4**)** to study the effects of supercritical CO_2 on the geothermal exploitation of sandstone and carbonate reservoirs [47].

The improved solubility resulting from pH adjustment (acidic) upon CO_2 addition enhances mass transfer. This ensures efficient contact between the leaching solution and solid phase, resulting in faster dissolution kinetics and higher extraction yields. As an example, Fig. 5 shows the mechanisms and kinetics of Zn separation using CO_2 assisted ammonia leaching solution of blast furnace dust, as reported by Luo et al. (2020), where over 91 % leaching was recorded [48]. Another aspect that leads to better solubility, selectivity, and a low chemical environmental footprint is the coupling of ionic liquids with CO_2 for leaching. The proposed system can be tailored and optimized for specific leaching processes [49].

The direct utilization of CO_2 as a leaching agent represents a novel approach for mineral extraction. Unlike traditional leaching agents, such as cyanide and sulfuric acid, which often pose environmental risks due to their toxicity and potential for environmental contamination, CO_2 offers a more environmentally benign alternative [50]. CO_2 high diffusivity allows efficient penetration into mineral matrices, facilitating the dissolution of target minerals. Additionally, CO_2's low viscosity and

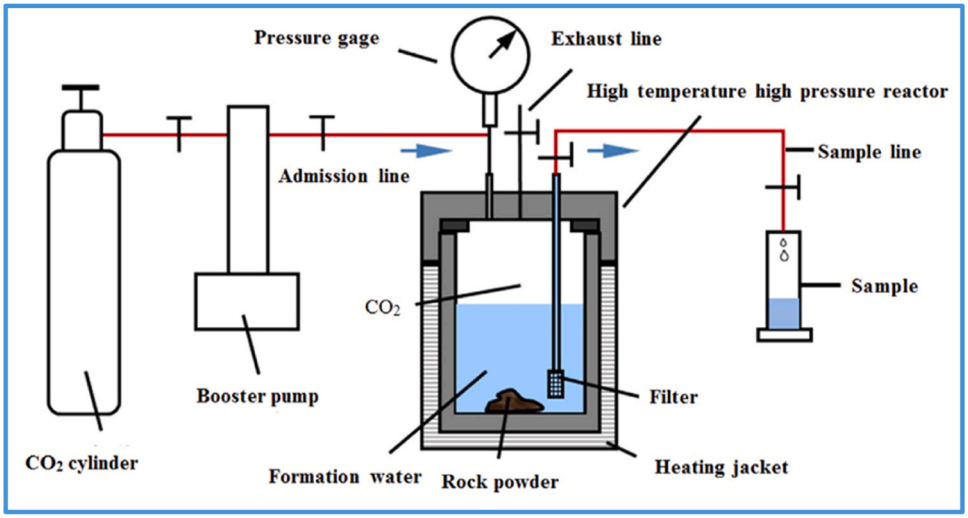

Fig. 4. Schematic of the high-pressure and high-temperature reactor used for CO_2–water–rock geochemical reactions [47].

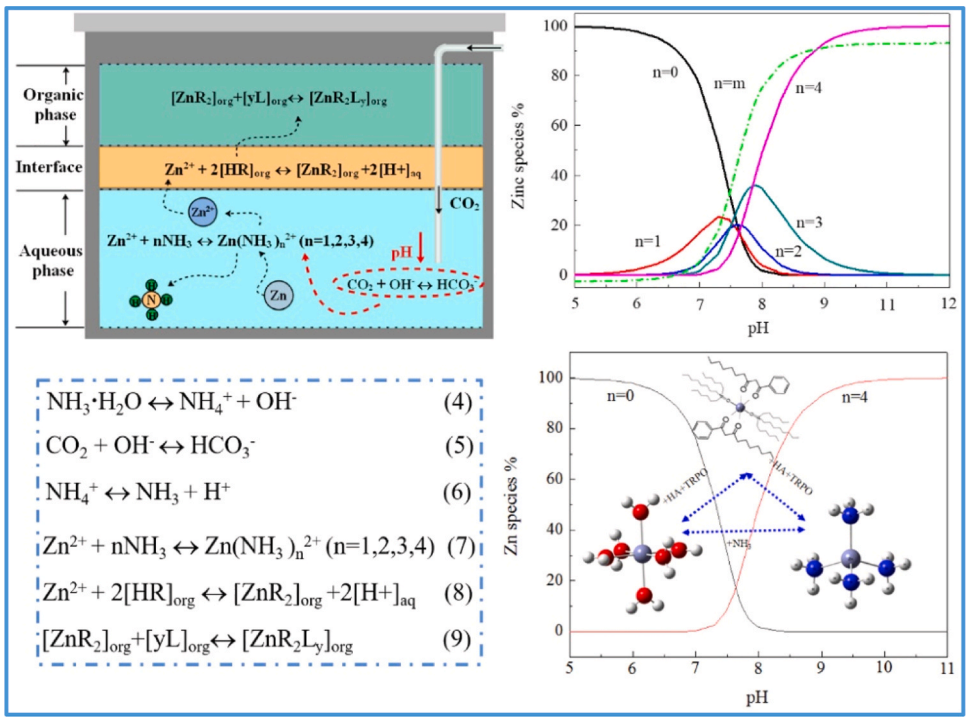

Fig. 5. Mechanisms and kinetics of CO_2 assisted Zn extraction [13].

tunable solvation power provide versatility in adjusting the leaching conditions for optimization [51]. By utilizing CO_2 sourced from industrial processes and power plants or captured from flue gases, mining operations can minimize the environmental footprint while simultaneously extracting valuable minerals, which is called mineral sequestration, as illustrated in Fig. 6 [52,53]. Ongoing research and development efforts are focused on refining CO_2 leaching techniques, optimizing process parameters, and expanding the applicability of this innovative approach across various mineral types and extraction scenarios [54]. For example, CO_2-assisted leaching is an environmentally benign hydrometallurgical approach to extracting lithium from end-of-life lithium-ion batteries, and it is an emerging research area that contributes to the achievement of a circular economy.

Lithium leaching from an otherwise insoluble heat-treated black mass (comprising mostly lithium, nickel, cobalt, graphite, and other impurities) derived from spent lithium-ion batteries was achieved via carbonation with supercritical CO_2 in a lab-scale autoclave, operated at 230°C and 50 bar pressure [55]. This process, known as early-stage lithium recovery, resulted in a leaching recovery of up to 79 %. Other researchers have further optimized the model to function under atmospheric conditions with a continuous flow of CO_2 at moderate temperatures. Neumann and Friedrich (2024) designed experiments using Response Surface Methodology and Central Composite Design. They reported that the optimized conditions yielding the highest lithium recovery of 97.18 %, with a response fit of 93.54 %, were determined at 77°C, 180 minutes, a solid-to-liquid ratio of 10:1 g/L, and a CO_2 flow rate of 6.0 L/min [56]. Although CO_2 flow rate had minimal impact on the leaching rate in this approach, CO_2 plays a crucial role in transforming the lithium phase into easily separable soluble carbonates and bicarbonates according to the following equations [57].

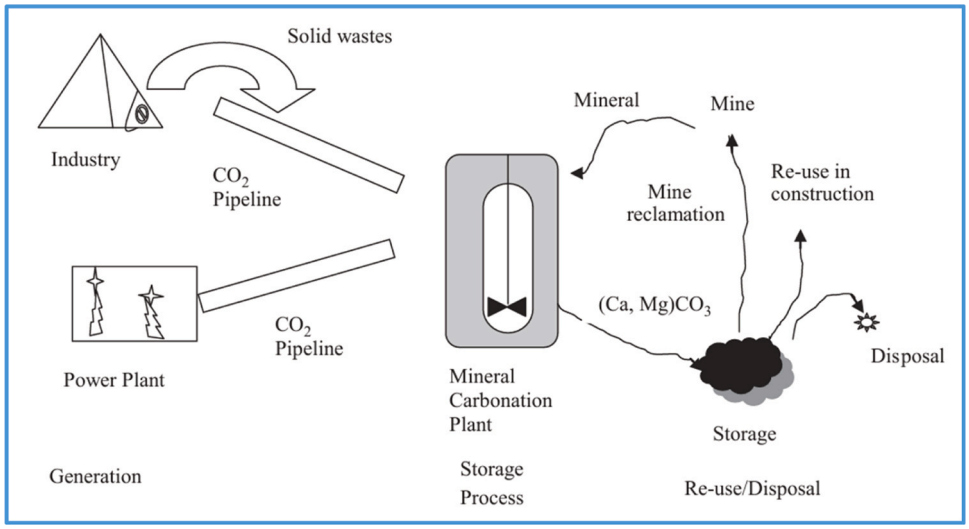

Fig. 6. Illustration of mineral sequestration concept [53].

$$CO_2 + H_2O \rightleftharpoons H_2CO_3 \rightleftharpoons HCO_3^- + H^+ \rightleftharpoons CO_3^{2-} + 2H^+ \quad (2.1)$$

$$Li_2CO_3 + H^+ \rightarrow 2Li^+ + HCO_3 \quad (2.2)$$

$$Li^+ + HCO_3 \rightleftharpoons Li^+ + HCO_3^- \rightleftharpoons LiHCO_3 \quad (2.3)$$

$$Li_2CO_3 + H_2CO_3 \rightarrow 2LiHCO_3 \quad (2.4)$$

Li_2O in the carbonized sample is first transformed into lithium hydroxide (LiOH) and subsequently into highly water-soluble lithium bicarbonate ($LiHCO_3$) according to the following equations:

$$Li_2O + H_2O \rightleftharpoons 2LiOH \quad (2.5)$$

$$LiOH + H_2O \rightleftharpoons Li^+ + OH^- + H_2O \rightleftharpoons LiOH.H_2O \quad (2.6)$$

$$LiOH.H_2O + CO_2 \rightleftharpoons 2Li_2CO_3 + 3H_2O \quad (2.7)$$

$$Li_2CO_3 + 3H_2CO_3 \rightleftharpoons 2LiHCO_3 \quad (2.8)$$

The dissolution rate of Li_2CO_3 in the CO_2-water system increases with both CO_2 pressure and flow rate [58]. When carbonized black mass, consisting of lithium-ion battery (LiB) cathodes and anodes, was processed, high-purity lithium—surpassing the quality of commercial-grade lithium—was recovered at a selective leaching efficiency of 94 % under conditions of a 6 % solid-to-liquid ratio and a CO_2 flow rate of 2 L/min [59]. Feasibility studies on scaling up these promising results are yet to be explored. Prior to the CO_2-H_2O leaching step, supercritical CO_2 and cosolvents can be used to liberate LiB cathode materials (such as $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ and $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$) from polyvinylidene fluoride (PVDF) binders, facilitating their recovery [60]. A recent techno-economic assessment comparing this hydrometallurgical approach with the traditional pyrometallurgical method concluded that the hydrometallurgical approach offers a more robust solution in terms of price fluctuations, which may be triggered by varying cell chemistries [61]. From an economic perspective, it is positioned as the technology of the future. The research discussed above is limited to lab-scale experiments, highlighting the need for scale-up trials to fully assess technical feasibility and identify potential bottlenecks associated with larger-scale implementation.

CO_2+O_2 leaching (HCO_3^- - oxidative leaching) can be applied for the in situ leaching of uranium from low-permeability sandstones. This process can be augmented by ultrasonic or low-frequency vibrations to improve the performance, depending on the type of deposit [62]. This type of leaching is essentially driven by bicarbonate (HCO_3^-). For example, in the case of uranium leaching, the role of CO_2 is demonstrated by the following equations [63,64]

$$2UO_2 + O_2 \rightarrow 2UO_3 \quad (2.9)$$

$$UO_3 + 2HCO_3^- \rightarrow \left[UO_2(CO_3)_2\right]^{2-} + H_2O \quad (2.10)$$

$$\left[UO_2(CO_3)_2\right]^{2-} + HCO_3^- \rightarrow \left[UO_2(CO_3)_3\right]^{4-} + H^+ \quad (2.11)$$

In a related study, the use of the TOUGHREACT model demonstrated that CO_2 enhanced HCO_3^--assisted oxidative dissolution and the mobility of uranium [65]. The dissolution rate could be further accelerated in the presence of Fe^{3+} ions [66]. While both studies investigated uranium mobility under geological CO_2 storage conditions, it is reasonable to infer that CO_2 plays a significant role in the leaching of uranium. The utilization of CO_2-augmented leaching can be extended to scandium extraction. Scandium extraction from landfilled bauxite residue was significantly improved (40–45 %) under Na_2CO_3 leaching and ultrasonic conditions, with CO_2 playing a critical role in facilitating carbonation within the reactor, which operated in the optimal pH range of 9.5–10.0 [67].

A thorough comparison of CO_2-aided leaching and conventional leaching agents provided important insights. Although traditional leaching is at the forefront of its effectiveness in mineral extraction, it has significant environmental and health drawbacks [54,68,69]. CO_2, on the other hand, has the potential to achieve comparable effectiveness with substantially reduced environmental impact. The advantages of using CO_2 as a leaching agent are thus evident. The low toxicity reduced chemical environmental footprint, low greenhouse gas emissions, and potential for enhanced selectivity make it an ideal option for sustainable mining [70,71]. However, despite these advantages, challenges still exist, including the need for specialized equipment and potential energy requirements for CO_2 capture and utilization with minimum loss. Achieving the desired extraction rates using CO_2 may also pose a challenge compared with conventional agents, which require optimization and improvement [72,73].

2.2. CO_2-induced pH modification in leaching

When CO_2 is introduced into a leaching solution, it undergoes chemical reactions that alter its pH. The extent of pH modification depends on several factors such as the CO_2 concentration, temperature, and initial pH of the solution [74]. The pH plays a crucial role in leaching processes as it influences the solubility of minerals and the kinetics of chemical reactions; thus, as CO_2 is introduced or removed from a solution, the pH levels fluctuate, which controls the leaching kinetics and metal recovery [75]. Changes in pH can significantly affect the metal dissolution rate and overall leaching efficiency. At low pH (acidic conditions), metal ions are typically more soluble, leading to faster leaching kinetics and higher metal recovery. However, excessively low pH levels can also result in the formation of insoluble metal hydroxides or carbonates, thereby reducing the leaching efficiency [75,76]. Conversely, under alkaline conditions (high pH), metal solubility may decrease, slow the leaching rate, and potentially limit metal recovery [77,78]. Therefore, the maintenance of optimal pH levels is essential for maximizing the leaching performance and metal extraction efficiency.

Numerous case studies and experimental investigations have demonstrated the effects of CO_2-induced pH changes on the leaching performances of various mineral systems. For example, research on gold leaching has shown that CO_2 addition to cyanide leaching solutions can decrease the pH and enhance gold dissolution rates and overall recovery [79]. Similarly, studies on Cu and Ni leaching have illustrated the impact of pH modification on metal solubility and leaching kinetics. The experimental data from these studies provide valuable insights into the relationship between CO_2-induced pH changes and leaching efficiency, highlighting the importance of pH control in optimizing metal recovery processes [80,81].

In summary, the successful development and deployment of CO_2-augmented leaching techniques requires the integration of CO_2 capture technologies directly into leaching facilities, enabling on-site capture and utilization. Additionally, the co-location of CO_2 capture facilities with mineral processing plants can streamline logistics and reduce transportation costs. These integrated approaches leverage the synergies between CO_2 capture and mineral leaching operations, thereby contributing to more sustainable and environmentally friendly mining practices. Table 1 lists some studies on the use of CO_2 in mineral processing.

Research Gaps:

- Enhancing the efficiency and scalability of CO_2-based mineral processing techniques.
- Catalyst design, reactor engineering, and process optimization can be used to improve the economic and environmental performance of these technologies.
- Under-utilizing CO_2 emitted from industrial sources as feedstock to reduce greenhouse gas emissions while producing valuable minerals.

Table 1
Summary of studies on the use of CO_2 in mineral leaching.

Ref.	Investigation	Process	Findings
[199]	Generalized strategy for recycling noble metals and fabricating SiO_2-based nanocomposites	CO_2-assisted weathering of steel slag-derived calcium silicate hydrate	Structure and negative surface charges enable steel slag-derived amorphous calcium silicate hydrate (CSH) to effectively retain incoming metal ions via chemisorption. The decalcification reactions of metal-bearing CSH resulted in the successful recovery of noble metals (Ag, Au, Pd).
[200]	Recovery of degraded $LiCoO_2$	CO_2-assisted low-temperature thermal reduction	Selective recovery of Li reached 94.21 %. Spatial isolation of $MgCO_3$ in the bottom layer, graphite in the middle layer, and $LiCoO_2$ in the top layer in the same reactor avoids complex. $LiCoO_2$ is directly regenerated by CoO/Co_3O_4 and Li_2CO_3. Regenerated $LiCoO_2$ exhibited good electrochemical performance with a discharge capacity retention of 94.0 % after 300 cycles of separation.
[42]	Effect of the physicochemical properties of Brazilian serpentinites on the leaching process	Indirect CO_2 mineral carbonation	S-GO is approximately 50 % more reactive than S-MG, and that S-MG is limited by low accessible surface and the high mineralogical complexity. Leaching confirmed the reactivity of Mg and Fe extraction.
[201]	Early-stage recovery of lithium from spent batteries	CO_2-assisted leaching	Results demonstrate a maximum leaching efficiency of lithium with optimized parameter set, achieving 97.18 % recovery with a fit response of 93.54 %.
[202]	Precipitation of rare earth elements from acid mine drainage	CO_2 mineralization	90 % Al and over 85 % of the REEs were recovered at pH < 7, while Fe precipitation was suppressed (i.e., 65 %). The precipitation pattern of REE hydrooxides followed the tetrad classification of the coordination number of the corresponding elements. The precipitation pattern of REE-carbonates followed the hydration energy trend of these elements.
[52]	Steel slags and leaching behavior variability in view of industrial mineral carbonation	Effective CO_2-specific sequestration	Separate leaching processes may need to be developed for slags with largely distinct mineralogical compositions and structural features.
[203]	Extraction of lithium and cobalt from $LiCoO_2$	Leaching using p (VBPDA-co-FDA) copolymers in supercritical CO_2	Supercritical CO_2 extraction conditions were optimized to improve metal extraction from $LiCoO_2$ cathode material, leading to 75 % extraction efficiencies of Li and Co at 60 °C and 250 bar.
[204]	Steelmaking slag with NH_4Cl as the leaching solution	Indirect CO_2 mineral sequestration	Rapid reaction rate with an apparent activation energy of 11.8 kJ/mol, indicating a diffusion-determined core-shrinking reaction during carbonation.
[205]	Extraction of selected rare earth elements from anthracite acid mine drainage	Supercritical CO_2 via coagulation and complexation	Overall, the REE extraction efficiency increased with the atomic number of the REEs. The average overall REE extraction efficiencies were 41.8 %, 40.1 % and 58.2 % for Ce, La and Nd, respectively.
[206]	Changes in mineralogical and leaching properties of converter steel slag	Accelerated carbonation at low CO_2 pressure	Sequestered CO_2 and the resulting pH reduction occurred within 24 h and pH reduction led to an increased vanadium leaching.

- Promote the adoption of CO_2-based processing technologies for sustainable mineral extraction and the formulation of regulatory policies that target carbon emission reduction.
- Identification of suitable mineral deposits, development of efficient processing techniques, and improvement of catalyst materials (with improved activity, selectivity, process intensification, and stability) for CO_2 mineralization reactions tailored to specific mineral compositions and conditions to enhance process efficiency and mineral recovery rates.

3. Utilization of CO_2 in flotation processes

Owing to the depletion of high-grade ore on the earth's surface, the mining sector is forced to process low-grade and complex ores with high clay content, which require many processing steps before attaining the desired product. This has also forced processing steps to ground the ores to finer particle sizes, introducing a large amount of clay content [82]. Valuable froth flotation has been employed to separate the clay and concentrate. Conventionally, flotation separates particles based on differences in their ability to attach to air bubbles in a slurry [83]. The particles that attach to the air bubbles are hydrophobic, adhere to the bubble, and rise to the surface, forming the froth layer, whereas hydrophilic gangue material remains in the pulp [84,85]. However, it should be noted that flotation has key limitations due to its dependence on the size of fine particles and the bubbles of the forming agent. Moreover, particle-bubble collision, bubble stability, and particle

attachment are critical in the flotation process. Therefore, the flotation recovery is dependent on these three factors[86]. Bubble size and abundance have also been identified as key factors in the rate of mineral recovery; hence, foaming gas is imperative in the floatation process. A lot of research has been focused on measuring the sizes and analysis of the bubble structure, distribution, and quality[87,88]. Another aspect which is imperative in the flotation process is the optimization of the grinding size to control the chemical-physical interaction between the surface chemistry of both the particle and the bubble and the multiphase fluid mechanics, which are governed by the engineering constraints on flotation circuits design [89].

Traditionally, air is used to create froths in flotation cells. However, recent research has shown an increase in the use of different types of gases to improve the process efficiency. The following gases have been used in flotation processes: nitrogen (N_2), oxygen(O_2), air, sulfur dioxide (SO_2), CO_2, and H_2S [90–92]. Incorporating CO_2 into the flotation process has the advantages of mitigating adverse climate effects and improving mineral processing beneficiation. Removal of atmospheric CO_2, decarbonization, and retrofitting of industrial CO_2 emitters with carbon capture utilization and storage (CCUS) systems are imperative for climate stabilization. In addition, CO_2 use, and reuse are crucial because of the uncertainties in the permanent sequestration of landforms. These strategies are in line with reducing the greenhouse gas effect and managing environmental pollutants using an economic and energy-based approach [93–95]. Numerous studies have been conducted on the utilization of CO_2 in flotation, either as a bubbling gas, pH regulator, or depressant [96–98]. In solution, CO_2 gas dissolves to form carbonate species, such as carbonate (CO_3^{2-}), bicarbonate (HCO_3^-), and carbonic acid (H_2CO_3). These carbonate species buffer the pH of the system, influencing mineral adsorption [99]. The findings of these studies provide irrefutable evidence that CO_2 efficiently enhances separation during the flotation process [100].

3.1. Mechanism of CO_2-aided flotation

The reported kinetics of CO_2-assisted flotation were documented by Wu et al. (2024) in their study on pyrite-assisted CO_2 flotation [101]. Different parameters were examined for the recovery and flotation process.

CO_2 content: The recovery of pyrite increased from 10.39 % to 89.44 % as CO_2 was increased from 0 to 10 mL/min. The recovery then dropped to 68.35 % when the CO_2 flow rate was further increased to 25 mL/min. Hence, an optimum amount of CO_2 activated pyrite, whereas excessive CO_2 reduced the activation effect.

pH: High pulp pH depresses pyrite, leading to low recovery. The addition of CO_2 effectively reduced the pulp pH owing to the dissolution of CO_2 forming carbonic and bicarbonate acids. A lower pH facilitated xanthan adsorption on the surface and the formation of dixanthogen. The flotation of pyrite is effective in neutral or slightly acidic environments of xanthate, where dixanthogen is stable.

Dissolved oxygen: The amount of dissolved oxygen in the pulp decreased as the CO_2 concentration increased, and the recovery of pyrite decreased when the oxygen content was less than 6.2 mg/L at a CO_2 concentration of 10 mL/min. The adsorption of xanthane on the pyrite surface is an electrochemical process involving the anodic process of xanthate oxidation to dixanthogen, as shown in (Eq. 3.1) and the cathodic reduction of O_2 in (Eq. 3.2). Moreover, pyrite flotation is dependent on the content of O_2 in solution, with excessive CO_2 is unfavorable for cathodic O_2 reduction, leading to the oxidation of the collector on the mineral surface, resulting in hydrophobic products and a decrease in the flotation process.

$$2X^- \rightarrow X_2 + 2e^- \quad (3.1)$$

Where X^- and X_2 are the xanthate ion and dixanthogen, respectively.

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \quad (3.2)$$

Stirring speeds: Recovery of pyrite was increased until a certain level and then decreased with increasing stirring rate. It can be concluded that an optimum stirring rate is essential for balancing the attachment and detachment rates of mineral particles in the flotation cell.

Electrochemical measurements: Electrochemical reactions are abundant in flotation cells depending on the oxidation-reduction conditions. For the CO_2-pyrite system, the reaction pathway for the pyrite-lime-CO_2 system occurs in the absence of additives. The reaction is described by Eqs. 3.3 and 3.4. The term S^0 represents an intermediate product, i.e., polysulfide (FeS_n) and metal-deficient sulfides ($Fe_{1-X}S_2$). These sulfur-containing compounds improve the hydrophobicity of pyrite, making it easier for the mineral to float. In comparison with other pyrite systems, it was deduced that CO_2 treatment suppressed the oxidation of pyrite [102]

$$FeS_2 \rightarrow Fe^{2+} + 2S^0 + 2e^- \quad (3.3)$$

$$E_h = 0.34 + 0.00295\lg[Fe^{2+}]$$

$$FeS_2 + 3H_2O \rightarrow F(OH)_3 + 3H^+ + 2S^0 + 3e^- \quad (3.4)$$

$$E_h = 0.59 - 0.0059pH$$

3.2. Experimental studies of CO_2 in flotation

Most sulfide mineral ores are hydrophobic in anaerobic environments, and their hydrophobicity is reduced by oxidation at the ore surface. These surface properties have been intensively investigated using various methods to understand the oxidation process [103,104]. The oxidation process in ores, such as pyrite, produces ferric hydroxide, which causes the surface to become hydrophilic. Oxidation of fresh pyrite occurs during the combustion of oxygen dissolved in pulp before flotation. These reactions adversely affect flotation and hence the recovery of sulfide minerals in the feed [105,106]. The incorporation of CO_2, N_2, or other activators can interact with mineral surfaces and alter their properties, leading to improved selectivity in the flotation process. This can be accomplished by modifying the mineral surface chemistry, resulting in improved mineral recovery. Somasundaran et al. (1985) examined the electrokinetics, solubility, spectroscopy, and flotation of apatite and calcite in electrolyte solutions. It was observed that the solution interactions of these systems could modify the surface characteristics of minerals [107]. Hence, understanding mineral-solution chemical equilibria in different physicochemical systems is imperative for ore processing, factoring in the existence of atmospheric CO_2 gas.

Orlich et al. (2017) patented a novel technique for the flotation of sulfide and carbonate ores in acidic solution using CO_2 as a bubbling gas. Furthermore, it has been reported that the use of CO_2/N_2 gas mixture flotation and solution conditioning accomplished pH control, thereby improving the recovery for flotation [108]. Aresta et al. (2003) reported the feasibility of using CO_2 as a pH regulator in flotation processes, leading to improved mineral recovery and reduced environmental footprint [109]. In addition, Wang et al. (2020) investigated the effects of CO_2 injection on bubble-particle attachment in flotation, highlighting its potential to enhance flotation kinetics and selectivity [110]. Sun et al. (2009) explored the impact of high-intensity conditioning (HIC) on the aggregation of fine particles and their flotation characteristics in a CO_2-saturated slurry with a specified bubble size [111]. Further, we examined the influence of dissolved gas, xanthate, and agitation speed on particle aggregation and flotation performance. The results revealed that the dissolved gas in the pulp combined with HIC promoted fine particle aggregation, thereby improving the flotation behavior. The concentration of dissolved air imposes a constraint on the quantity of bubbles produced by hydrodynamic cavitation, which is aimed at improving fine particle aggregation and flotation kinetics. Given that the solubility of CO_2 in water exceeds that in air, saturation with CO_2 before

high-intensity conditioning can amplify hydrodynamic cavitation, possibly boosting fine particle aggregation. Table 2 summarizes the key studies on the use of carbon dioxide in flotation processes.

Wani et al. (2021) investigated the utilization of captured CO_2 in the processing of nickel. CO_2 was used as a conditioning agent before froth flotation, which resulted in the conversion of monohydroxide complexes to carbonates. This resulted in an increase in the electrostatic repulsion between the gangue and valuables, resulting in improved froth flotation recovery [112]. Khan et al. (2021) investigated the conversion of serpentine to magnesite by CO_2 (ex situ mineral carbonation), which prevented the slime coating of pentlandite, the main nickel-bearing mineral, and improved the recovery of nickel and the grade of the concentrate [113]. In this study, micro-flotation improved the nickel recovery from 61.2 % to 87.4 %, whereas the concentration grade increased from 20.6 % to 24.7 %. Moreover, carbonation as a means of assisting nickel (Ni) separation from low-grade ultramafic ores has been investigated because of the difficulty in recovering Ni from low-grade fines on an industrial scale. The low-grade serpentine ore was carbonated for 24 h at 185 ° C and 15 MPa, and the resulting conversion of serpentine to magnesite was 22 % in the presence of inorganic salts and 16 % in the absence of inorganic salts. The resultant compound was then introduced to a Denver cell for froth flotation investigation, which resulted in carbonated ore in the presence of inorganic salts with 29 % higher Ni recovery and 0.15 % higher nickel grade than uncarbonated ore [113]. Wang et al. (2021) used pressurized CO_2 to precipitate nickel sulfide from a solution of magnesite ($MgCO_3$), nickel sulfide, and carbonate at a neutral pH from olive ore with 32 % conversion [114].

3.3. Future research on CO_2 utilization in flotation

In summary, although the use of CO_2 in flotation has not been extensively researched, literature sources show that nickel ultramafic low-grade fines and uranium-carbonate phosphate processing are two large-scale mining operations that use CO_2 for carbonation of mineral ore, resulting in improved yields. It is important to further investigate the potential benefits and limitations of this process.

The carbon dioxide-aided flotation process provides an alternative pathway for greenhouse gas control, where CO_2 can be recovered and reused in a closed process. When reuse is not possible, CO_2 escapes into the atmosphere. However, in the case of nickel flotation, there is a partial reduction of CO_2 released into the atmosphere owing to surface reactions occurring during the process.

It is imperative to focus on the following to better comprehend the application of CO_2 in flotation.

- **Carbon dioxide-responsive flotation Reagents**: The development of flotation chemical reagents that are compatible and responsive to different CO_2 concentrations. This aims at controlling the capacity and improving the flotation process. Moreover, it can be investigated as a frothing agent during flotation.
- **Application of CO_2 in the flotation of Carbonaceous Materials**: Exploring CO_2-assisted flotation on carbonaceous materials, such as graphite and coal. Owing to the nature of these materials, they may interact differently than traditional flotation substances, leading to an efficient separation pathway.
- **Scale –Up studies** – The use of CO_2 in large-scale flotation studies is limited to nickel ores in Brazil. Hence, its widespread application at the industrial scale is premised on a techno-economic analysis of the process to determine the cost–benefit analysis.
- **Modelling and Simulation**: Computational models and simulations are used to understand the micro-scale interaction of carbon dioxide with different ore minerals, as well as to optimize the operational parameters.

Table 2
Studies carried out in flotation using carbon dioxide.

Refs.	Nature of study	Conditions and Additives	Operation scale	Study Outcomes
[113]	Utilization of carbonation to enhance Ni beneficiation via froth flotation from low-grade ultramafic ores containing carbonation-reactive silicates	Carbonation-185 °C, 15 MPa, and 24 h Reagents for carbonation – $NaHCO_3$ and NaCl Collector – potassium amly xanthate Frother – polypropylene glycol pH-HCl and $NaCO_3$ Bubble generator - air	Denver cell	Ni-Fe sulfides particles available after carbonation provided a route for direct flotation of carbonate ore. Carbonation-assisted serpentine flotation is a promising two-stage industrial process
[8]	Utilization of carbon dioxide in the mineral processing of ultramafic nickel ores	pH modifier – NaOH Collector – potassium amly xanthate Zeta potential –	Laboratory scale	Incorporation of CO_2 in the froth flotation improved nickel recovery and grade by 10 % and 4 % respectively, while storage of CO_2 was achieved. Also, MgO was supressed thereby improving cumulative Ni grade
[98]	Adsorption of sulfosuccinate collector on apatite and carbonates in a phosphate ore, in the presence of CO_2	Collector-Sulfosuccinate Bubble generator – CO_2 pH modifier – NaOH	Lab scale and Microflotation	Selectivity in the experiment was enhanced by the use of CO_2 as a means of bubble generation instead of air through the enhancement of the. sulfosuccinate collector.
[207]	Calcite flotation from apatite of a uranium-carbonate phosphate ore. Sample size was 100 % passing 3.36 mm.	Collector-Saponified coconut fatty acid pH regulator & bubble generation – CO_2	Bench and pilot scale	P_2O_5 recovery was 91 %, P_2O_5, SiO_2, Al_2O_3 were 29 %, 11 %, 13 %. CO_2 can be used as a depressant in place of organic acids as apatite depressant. Concentration of apatite was achieved without the need to remove generated slimes, which is not observed in the current practice of apatite flotation plants
[208]	Investigation of the effects of CO_2 & N_2 on	CO_2 for pH control and bubbling gas	Microflotation pyrite	Use of CO_2 reduces the amount of acid

(continued on next page)

Table 2 (*continued*)

Refs.	Nature of study	Conditions and Additives	Operation scale	Study Outcomes
	pyrite surface properties and flotation outcomes.	Collector – potassium amyl xanthate (PAX)		required for the process when compared with the N_2 system. It also controls the pH of the solution and inhibits oxidation while increasing the recovery of pyrite particles
[108]	Patent of a processing of carbonate & sulphide (Carlin ores)	CO_2 as the flotation gas	Pilot scale	Auriferous pyrite flotation recovery of 51.3 %, 36.7 % and 38.6 % and 51.3 % in the presence of high, medium and low carbonate feed.
[209]	Flotation for Apatite concentration of the Santa Quiteria carbonaceous uranium-phosphate 96 % passing 150 μm (>3.36 mm) 100 % passing	Carbonic gas for flotation Calcite collector – coconut soap Apatie collector - sulphosuccinate	Pilot scale	P_2O_5 overall recovery was 68 % from a feed of 16.8 %
[210]	Use of micro-sized carbon dioxide gas bubbles in flotation instead of air bubbles.	Bubble generator – air/CO_2 Coagulant – polyaluminum chloride (PAC). Coagulation pH-Sodium hydroxide (NaOH). pH-was slightly decreased by CO_2 from 7.0 to 6.0	Bench-scale	Air bubble sizes decrease as the pressure increases but no change was observed under constant pressure. Whereas, CO_2 bubble size increased at higher pressures. Since the pressure of CO_2 bubble collector is lower than that of the air bubbles in the flotation process, the CO_2 bubbles offer an advantage of lower operating costs.
[211]	To analyse the effects of CO_2 on apatite flotation from Catalao siliceous carbonate phosphate ore	Collector – sulphosuccinate, Depressant - corn starch Bubble generation – CO_2	Flotation experiments machines	Apatite was collected in the froth, carbonate minerals remain in the sink. HCO^- is produced from the reaction between CO_2 and the carbonate mineral it competes with anionic collector species to decrease the

Table 2 (*continued*)

Refs.	Nature of study	Conditions and Additives	Operation scale	Study Outcomes
				hydrophobicity on the surface state for calcite and dolomite
[212]	Flotation of siliceous carbonate phosphate ore form Araxa carbonatite based on calcite/ dolomite	pH modifier & bubble formation – CO_2 Mineral Collector – coconut soap Depressant – corn starch Collector – fatty acid & sulphosuccinate	Pilot scale	Flotation was improved due to the utilization of CO_2 gas

4. Carbon dioxide in wastewater treatment

The utilization of carbon dioxide in wastewater treatment from mineral processing operations is an emerging field of study that exploits the properties of CO_2 and its reactivity to manage wastewater. In this context, CO_2 can be used for pH adjustment, carbonate and bicarbonate formation, metal solid precipitation, and advanced oxidation processes (AOPs). The origins of wastewater produced during mineral processing and mining operations are presented in Table 3. Depending on the source point, the effluent may contain increased levels of dissolved metals, acids, and alkalis [115].

Traditional physicochemical treatments such as neutralization, sedimentation, and coagulation/flocculation, which are most commonly used in wastewater treatment processes, are inefficient, particularly for wastewater containing recalcitrant pollutants. Improved treatment of biological pollutants has been achieved with the application of technologies such as advanced oxidation processes (AOPs) in the mineral industry [116]. Seawater with industrial wastewater was utilized to precipitate $MgCO_3$ and $NaHCO_3$ using captured CO_2 and NaOH as an absorbent[117]. This innovative approach to managing mining wastewater involves using the excess CO_2 gas produced as waste. This underscores the different strategies required for incorporating CO_2 into the treatment of mining wastewater.

4.1. Application of CO_2 in pH adjustment

Carbonic acid is a naturally occurring substance that helps regulate pH levels. When carbon dioxide is added to wastewater, it produces benign byproducts that successfully regulate the pH under slightly acidic conditions. Carbon dioxide has been observed to be more effective for pH regulation than convection pH regulators such as sulfuric acid (H_2SO_4) in wastewater treatment [115]. Carbon dioxide gas is deemed innocuous because it does not induce a decrease in pH beyond a specific threshold, unlike sulfuric acid or other substances. Injecting CO_2 generates carbonic acid, which is weaker than the sulfuric acid and chlorine used in chemical treatments [118]. This slows down the reduction in the pH of wastewater, thereby making it easier to control [119]. It has been observed that not all carbon dioxide wastewater treatment options exhibit equal efficiency. Existing methods for applying CO_2 in wastewater management are less efficient because they add carbon dioxide

Table 3
Sources of wastewater generated in mineral processing [115].

Category	Sources
Processing wastewater	Concentrate wastewater, Tailing wastewater and Ore flushing water
Other wastewater	Equipment with water, Drainage of wet dust cleaning apparatus Floor flushing water, Mine acid wastewater

into the water and release gas into the atmosphere [120,121]. The reaction of H_2O with CO_2 to form H_2CO_3, which then dissociates into H^+ and HCO_3^- ions governs the pH changes of the solution with the changes in the H^+ ion concentration [122]. A study on pulsed bed treatment acid mine drainage (AMD) using CO_2 as a means of pH control to precipitate Fe^{3+}, Al^{3+}, and Mn^{2+} ions from a solution demonstrated that CO_2 effectively raises the pH to the desired level of approximately 5.85 – 6.09. Moreover, numerical modeling has shown that CO_2 requires an increase in the acidity of the AMD solution [123].

4.2. Precipitation of metal ions

AMD is the predominant form of wastewater generated in mines [124]. It contains sulfide minerals and ontains elevated levels of dissolved metals, which vary depending on the preceding processes [125]. Power stations are notorious for producing aluminosilicate alkaline solutions, usually referred to as coal fly ash (CFA). The introduction of CO_2 into the solution resulted in the formation of $CaCO_3$ as Ca^{2+} ions reacted with CO_2. This procedure enhances the management of CFA, improves AMD quality, and manages CO_2 emission. One study discovered that by introducing CO_2 into the leachate, the carbonation process not only further neutralizes acid mine drainage (AMD), but also effectively stores CO_2 [126]. Nyambura et al. (2011) utilized CO_2 gas to react with a CFA/brine slurry, from which coal-fired power plants contained CaO, which could be converted to solid benign calcite. A maximum of 71.84 kg of CO_2 can be sequestered per ton of FA [127]. Furthermore, Park et al. (2014) experimentally carried out the reaction of Ca $(OH)_2$ slurry with CO_2 in the presence of 30 wt% MEA (mono-ethanolamine) solution, the MEA converted CO_2 to ionic CO_2 with $CaCO_3$ produced as well as $Ca(OH)_2$ being identified in the products [128]. Bullock et al. (2021) compiled a database of diamond tailings, metal tailings, and silicate-based mining waste studies on CO_2 utilization for the management of these waste materials with the precipitation of metal ions [129]. Habte et al. (2020) investigated the removal of two metal ions, cadmium (Cd(II)) and lead (Pb(II)), using $Ca(OH)_2$ and CO_2 gas, which controlled the pH and produced carbonate ions in solution. More than 99 % metal ion removal through precipitation was achieved using both the metal ions. The resulting products are $PbCO_3$ (cerussite) and $CdCO_3$ (otavite). It was also observed that the removal of metal ions from the solution could occur through adsorption on the surface of calcium carbonate [130]. Brush et al. (2006) reported the precipitation of carbonate minerals from MgO in a waste isolation pilot plant (WIPP) by the U.S. Department of Energy's (DOE). MgO is composed of periclase (pure crystalline MgO that reacts with CO_2 and H_2O to form $Mg(OH)_2$) and hydromagnesite ($Mg_5(CO_3)_4(OH)_2$ 4 H_2O) as an intermediate product, with $MgCO_3$ as the final product [131].

4.3. Carbonate formation

The formation of metal carbonates during mineral processing is also known as mineral carbonation. This process involves the reaction between CO_2 and minerals such as serpentine and olivine to produce stable carbonates [132] [14]. During mineral carbonation, CO_2 undergoes a chemical reaction with calcium (Ca) or magnesium-rich (Mg)-rich minerals, resulting in the formation of carbonate compounds [133–135].This reaction also releases heat [136]. The reactions are shown in Eqs. 4.1–4.4:

$$\text{Wollastonine}: CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2 \quad \Delta H = -90kJ/mol \tag{4.1}$$

$$\text{Olivine}: Mg_2SiO_4 + CO_2 \rightarrow 2MgCO_3 + SiO_2 \quad \Delta H = -89kJ/mol \tag{4.2}$$

$$\text{Serpentine}: CaO + CO_2 \rightarrow CaCO_3 \quad \Delta H = -178kJ/mol \tag{4.3}$$

$$Mg_3SiO_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + 2SiO_2 + 2H_20 \quad \Delta H = -64kJ/mol \tag{4.4}$$

The conversion of CO_2 to solid inorganic carbonates is thermodynamically favorable [14,135,137]. By forming solid carbonates, this process immobilizes elements that may be harmful to the environment, such as Cr, Zn, Cu, and Pb. These elements precipitate out of the solution as carbonates, thereby reducing their hardness and removing them from wastewater. By trapping these elements at this stage, this method allows the recycling of alkaline materials and decreases the expenses associated with landfilling [138,139]. Dri et al. (2013) investigated the calcium carbonation of ground-graduated blast furnace slag (GGBS), steel slag (SS), and phosphorus slag (PS) with a focus on the solid-to-liquid ratio (S/L). It was observed that the carbonation efficiency was inversely proportional to the S/L ratio due to the dilution effects. The main product obtained was $CaCO_3$ with trace amounts of $CaSO_4$ and SiO_2 [140]. The carbonation of phosphorus slag to produce high-purity $CaCO_3$ was carried out using a novel procedure that incorporated acid leaching in the presence of CO_2 [141]. The use of CO_2 for the treatment of metal-oxide-bearing industrial waste streams involves the formation of carbonates and solid products. It has been reported that magnesium and calcium ions are sufficient to react with CO_2 in solution [142].

Mineral carbonation effectively absorbs carbon dioxide (CO_2), whereas the industrial applications of CO_2 and the products of CO_2 sequestration prevent its reintroduction into the environment. However, despite its high cost, this technique will continue to be widely used because it guarantees the long-term storage of CO_2 [134,135,143]. Table 4 presents a concise overview of important studies on the use of carbon dioxide in mineral wastewater treatment.

4.4. Research trend in utilization of CO_2 in mining wastewater treatment

- Incorporation of Photocatalysis: Utilization of numerous catalysts, including activated carbon, to enhance the utilization of CO_2 in treating mining waste with the use of solar energy to improve the process is a promising avenue that can be researched.
- Electrochemical approach: The use of electrochemical means to facilitate the conversion of CO_2 into valuable chemicals while simultaneously affecting wastewater treatment can be investigated further.
- Hybrid systems: The development of numerous systems that incorporate various biological treatment methods into CO_2 usage in wastewater treatment can be explored.
- Biocarbonation: There is a need to explore the use of microorganisms to improve the carbonation process and hence the efficiency of CO_2 use.
- Environmental impact assessment It is imperative to increase research focus on the evaluation of the environmental impacts of using CO_2 in the wastewater treatment, with a special focus on greenhouse gas emission mitigation.

5. Mineral tailings carbonation

Tailing carbonation is a spontaneous process involving the leaching of cations (e.g., Mg^{2+}, Ca^{2+} and Fe^{2+}) from mine tailings, followed by an exothermic reaction with dissolved CO_2 (HCO_3^-) to form solid carbonates [144,145].

This process occurs naturally when atmospheric CO_2 dissolves in water during rainfall, forming carbonate and bicarbonate ions that leach divalent metal ions from the minerals at low pH and react with CO_2 at high pH to produce carbonate minerals (natural weathering) [146,147]. Carbonates with different thermodynamic stabilities are formed depending on the temperature, CO_2 partial pressure, and water availability. The mechanism of aqueous carbonation is schematically summarized in Fig. 7. The process involves [1] the dissolution of divalent

Table 4
Research conducted on mineral processing wastewater using carbon dioxide.

Ref.	Impurities targeted.	Use of CO_2	Findings
[213]	Various industrial chemicals	Mineral Carbonation	The challenge with CO_2 utilisation in managing waste streams is short storage times but mineral carbonation offers long-term CO_2 solutions with the need to make the overall CO_2 sequestration economically. In addition, the use of CO_2 as a feedstock in chemical production is detailed.
[115]	Ammonia plant wastewater with heavy metal	pH adjustment	The use of Carbon dioxide (CO_2 instead of sulphuric acid (H_2SO_4) in a modified wastewater treatment scheme. CO_2 reduced pH to 7.56 and heavy metals to 1.49 ppm (neutralized the solution) while H_2SO_4 reduced the pH to 8.77 and heavy metals to 2.90 ppm.
[133]	Ca/Mg bearing alkaline solids and heat	Mineral carbonation	Carbon dioxide fixation in the form of inorganic carbonates, often known as 'mineral carbonation and CO_2 industrialization as a feedstock for carbon-containing compounds.
[214]	Ni/Al_2O_3 and $Ni/MgAl_2O_4$ catalyst	pH Control	Carbon dioxide rapidly decreases elevated pH levels to 5.9 – 6.4 that are the requirements for RO membrane requirements. Since it is not stored as an acid solution, it is regarded as safer than sulfuric acid. Moreover, CO_2 exhibited a self-buffering effect as it tends towards neutral levels that enables control of the end point without overshooting into undesirable low pH levels.
[124]	Alkaline oxides such as calcium oxide (CaO) and magnesium (MgO)	Mineral carbonation	Physico-chemical properties of fly ash, its effects on carbonation, kinetics of improving CO_2 mineral carbonations were detailed. These improvements were specifically applicable to cement and concrete properties. Proposal for combinations of various waste streams to reduce processing costs and recovery of elements such as Al, Na, and Fe before carbonation
[126]	FeS_2 and alkaline aluminosilicate known as fly ash in acid mine drainage. Additive added were Lime and $Al(OH)_3$	Precipitation of $CaCO_3$	Mineral carbonation enhances the disposal process of fly ash waste material, and aids in managing CO_2 emissions from coal power plants. Solid products were utilised in back filling of mine voids
[125]	Acid mine drainage with coal fly ash with Al, Fe, Ca, Mg, sulfate	pH Control	Major components removed were Al, Fe, Ca, and Mg. Removal efficiency of 98.4 % was achieved. The residue of this process is suited for backfilling of mine voids. Furthermore, in this study a jet loop reactor pilot plant was shown to be suitable for the treatment of AMD with coal fly ash to produce clear water.
[215]	Ca/Mg bearing alkaline solids	Mineral carbonation	The influence of iron and silica on the rates of carbonates formation and mechanisms need further as this has impacts on the understanding of CO_2 storage in geological settings
[138]	Waste residue from stainless steel slags	CO_2 carbonation though using continuous casting (CC) and argon oxygen decarburization	Thin- film deemed more effective means of carbonation for slag as it eliminates processing steps and reduces energy demand. Mineral carbonation proceeded with reduction in basicity with slurry carbonation found to be commercial viable for carbonation conversion and optimal treatment.
[216]	Ca/Mg bearing alkaline solids	Mineral carbonation, pH adjustment. Ca ions precipitate	The transformation of CO_2 into a precipitated mineral carbonate through a mineral carbonation route is Carbon capture and storage (CCS) is viewed as a viable solution for addressing carbon emissions.
[217]	Cementitious materials (SCM), and basic oxygen furnace slag (BOFS)	High-gravity carbonation	BOFS was the nuclei for the precipitation of the hydrate products. Compressive strength, autoclave expansion and sulfate resistance identified as critical parameter for products.
[218]	Iron and steel slag from blast furnace, basic oxygen furnace slag, ladle refining furnace slag and electric arc furnace,	CO_2 for carbonation	Physico-chemical properties of slag were outlined and means of accelerating the carbonation of mineral carbonation. Alkaline waste carbonation data

(*continued on next page*)

Table 4 (*continued*)

Ref.	Impurities targeted.	Use of CO_2	Findings
			is vast for lab scale operations and scale up is imperative for reactor design instrumentation control and process control models.

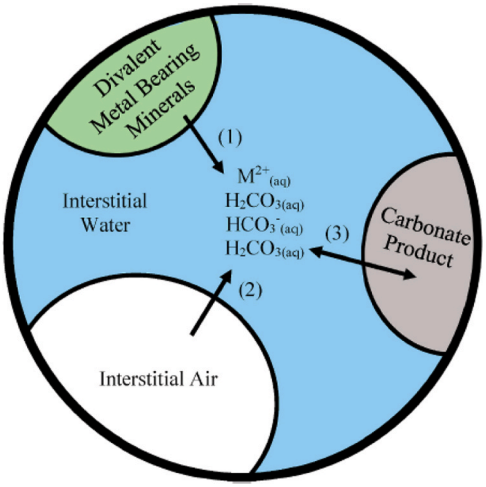

Fig. 7. Conceptual model of mineral carbonation in ultramafic tailings[179].

metals into interstitial water, [2] the dissolution of CO_2 into interstitial water, and [3] the precipitation of carbonate products.

5.1. *Suitable tailings for carbon storage*

Therefore, it is important to develop a global database of tailings based on their reaction kinetics and storage capacity. Tailings suitable for carbon storage should combine small particle sizes and high mineral-reactive content for fast reactions [148]. Based on these aspects, we discuss suitable tailings.

5.1.1. *Grain sizes*

Minerals should be sufficiently small to have high surface areas for fast carbonation. For the same mineral, fine-grained material underwent faster dissolution than the coarser particles. Compared to geological rocks, tailings already possess high surface areas and therefore require no further milling. This is because most of the samples were ground to less than 50–200 μm during mineral processing [149–151]. Finer-grained mine tailings, such as PGMs, diamonds, and asbestos (75 μm), exhibit faster dissolution kinetics than coarser tailings, such as Cr, Ni, Cu, and Ti (100–250 μm) [129].

5.1.2. *Chemistry and mineralogy*

The reaction kinetics depend on the mineralogy of the rock. The dissolution kinetics of different minerals determines their reactivity and suitability [18]. Tailings with high quantities of silicates exhibit slower kinetics than oxides and hydroxides owing to passivation [152]. Thus, tailings with brucite minerals are more kinetically favorable for carbonation, even without optimizing the conditions, than tailings with silicate minerals that require activation [153]. Ores from NiS [154,155] asbestos [156,157] and kimberlite diamond [158,159] exhibit fast natural weathering because of the presence of fast-reacting brucite in the tailings. Fig. 8 shows the importance of chemical composition and temperature on the dissolution rates of the selected minerals.

However, the main limitation is that the brucite content is lower in

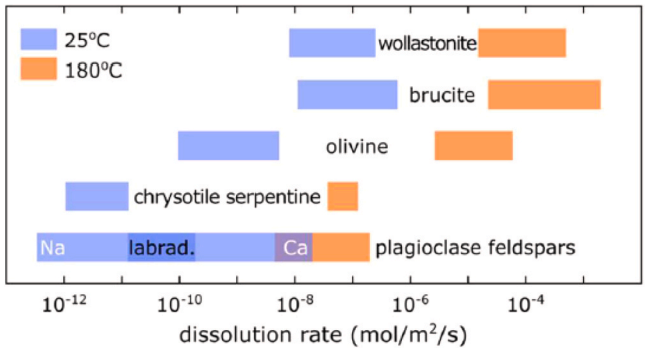

Fig. 8. Dissolution rates of magnesium- and calcium-rich minerals at 25°C (blue) and 180°C (orange) [18].

tailings than in silicates, which generally exist in abundance in the Earth's crust. Laboratory experiments and reactive transport modeling have been developed and used to investigate and quantify the rate of mineral carbonation in mine tailings [160–162]. For example, the shrinking core model is commonly used to estimate the weathering rates of different minerals and determine the dissolution times of minerals of a particular size [129,163,164]

The carbon dioxide storage quantity of a mineral depends on its chemical composition, specifically the amounts of Mg, Ca, and Na present, because once these elements have reacted, the mineral will be saturated with CO_2 [152]. Mg-bearing minerals are found in high quantities in ultramafic and mafic (sedimentary) deposits below the Earth's surface, and Mg is more abundant than Ca or Na. Ultramafic (igneous) rocks are more suitable than mafic (sedimentary) rocks because of their high Mg content. Bullock et al. (2022) studied and classified mineral kinetics and storage capacity, and found that out of 31 minerals, only 12 were suitable for capturing more than 100gCO_2/kg [165]. Research on the most suitable mineral tailings for carbon dioxide storage generally accepts those associated with forsterite olivine (Mg_2SiO_4), serpentine ($Mg_3Si_2O_5(OH)_4$), wollastonite ($CaSiO_4$), and brucite-layered mafic intrusions because of their abundance and reactivity [165–167] (Table 5), although further research is still necessary to identify others. Tailings from Fe, Sn, and U mining occur in large volumes, but their mineralogy is unfavorable for mineral carbonation [129, 168].

5.2. *Global production and storage capacity of mine tailings*

In addition to the chemical reactivity and composition demonstrated in Section 5.1, the annual volumetric production rates and abundance of kinetically favorable minerals are other requirements for successful mineral carbonation. In addition, mining sites should be close to and accessible to the main CO_2 emission sites. A survey on the global production and storage capacity of mine tailings is not yet conclusive, and further research is still necessary. The storage capacity depends on the identification of all suitable tailings and actual production rates. Bodénan et al. (2014) matched accessible mining waste with large-scale CO_2 emitters using a geographical information system (GIS) [169]. This section reviews the global production and storage potential of known tailings currently being produced.

5.2.1. *Global production*

The global annual production of mine tailings is approximately 420 million tons, which is expected to increase drastically because of the depletion of high-grade ores [170]. In addition, historical tailings stored in dams and stockpiles, which have accumulated over several years, have the potential to offset many emissions [171]. The quantity of existing ultramafic tailings accumulated over a period of time approaches 10 billion tons, whereas the fraction already carbonated by

Table 5
Promising minerals for mineral carbonation [219,220].

Mineral	Formula	Application	Ref.
Olivine	$(Mg, Fe)_2SiO_4$	High energy mills for structural changes	[221]
		Mineral sequestration of CO_2	[82]
		Manufacturing of magnesite and silica	[83]
		Study of Mg-rich silicates carbonation at 400 and 500 °C and 1 kbar	[222]
		Effect of temperature, pH, CO_2 pressure and salinity on the olivine dissolution kinetics	[223]
		Influence of attrition milling on carbon dioxide sequestration on magnesium–iron silicate	[224]
Serpentine	$Mg_3Si_2O_5(OH)_4$	Long term CO_2 storage	[225]
		Enhancing process kinetics for mineral carbon sequestration	[92]
		Promote CO_2 sequestration by mineral carbonation	[94]
		Direct flue gas CO_2 capture and sequestration	[95]
		Enhancing Mg extraction from lizardite-rich serpentine	[192]
Serpentinite		Enhanced CO_2 mineral sequestration	[18]
		Carbonation of magnesium silicate	[97]
		Effect of liquid recirculation on CO_2 sequestration and carbonate precipitation	[96]
		CO_2 sequestration following acid leaching	[98]
		Preparation of magnesium hydroxide for CO_2 mineral carbonation	[99]
Wollastonite	$CaSiO_3$	Carbonation of Ca bearing silicates	[226]
		Chemically accelerated mineral carbonation	[227]
		Wollastonite in situ mineral carbonation for CO_2 sequestration	[228]
		Energy-efficient mineral carbonation of $CaSO_4$ derived from wollastonite	[229]
		CO_2 mineral sequestration	[230]

natural weathering is unknown [171]. Currently, approximately 30 countries produce tailings suitable for carbon capture [165]. South Africa produces more suitable tailings than other countries, mostly from PGMs and diamond mining. Global PGMs production is projected to contribute approximately 2.1Gt of tailings by 2030 [129]which may lead to an accumulation of approximately 41Gt by 2050 and 145Gt by [163,165].

5.2.2. Storage capacity

We now examine the storage capacity of tailings produced annually worldwide. Currently, most tailings capture and store CO_2 through natural weathering, and their storage capacity is low. The estimated storage capacity of tailings is 93(natural)-465(enhanced) Mt/y [165].It is projected that approximately 1.5 % of global CO_2 emissions are passively offset by mine tailings [165]; passive mineral carbonation has been observed in many mine tailings worldwide. This has been characterized by studying the surface and depth of tailing stockpiles in the laboratory, field experiments, and observations [172–174]. However, natural weathering rates are very low when compared with carbon dioxide production to achieve appreciable carbon dioxide removal (CDR) on timescales relevant to climate change mitigation [175–177].

It is expected that 10Gt/year by 2050 and 20Gt/year by 2100 CO_2 will be removed from the atmosphere [165,178]. The total cumulative CDR from 2030 to 2100 is expected to reach 33GtCO_2 and this, 60 % of which will be removed by PGM tailings produced in South Africa, Russia, and North America [165]. Among suitable tailings, PGMs are the highest contributor, with an annual production estimate of approximately 2.1Gt in 2030, contributing to > 60 % of the total storage potential [165]. Many countries that use active mining have the potential to capture atmospheric CO_2. There have been successful projects on carbon storage in mineral tailings, including Baptiste Nickel, Dumont Nickel, and Crawford Nickel [152,179,180], as shown in Fig. 9. Improvements in carbon capture can also be achieved with the use of historic tailings and acceleration of chemical reaction rates. Thus, large-scale implementation of carbon dioxide removal strategies should be developed, and the availability of cost-effective methods for carbon storage in tailings will unlock this potential.

In addition to permanent storage as carbonates, mine tailings can also be used for carbon capture. This approach involves capturing CO_2 from the atmosphere to form carbonates and recycling them through calcination to release CO_2 before compressing and transporting it through pipes for permanent storage, for example, in geological formations, as shown schematically in Fig. 10 [18,19]. It has been estimated that if one-quarter of global magnesite reserves are mined and used for CO_2 capture before calcination and MgO recycling, approximately 1Gt of CO_2 can be removed in one cycle [18].This approach can also be used to obtain high-purity CO_2 for injection into the bulk mine tailings for permanent carbonation storage.

5.3. Enhancing CO_2 storage in mine tailings

The aim of enhanced weathering is to achieve the same results as natural weathering with much faster kinetics. Laboratory experiments and field-scale studies have proven that the mineral carbonation of mine tailings can be enhanced [181,182]. The feasibility of implementing most of these suggestions is in its infancy, and these suggestions have not yet been implemented in the field. For example, approaches such as tailing spreading are limited by the area footprint and are not yet practical. Challenges in implementation exist because the laboratory environment is very different from the field environment, particularly the scale of fresh tailing production. We present promising approaches for enhancing the carbonation of tailings in mines on a large scale.

5.3.1. Integrated reactor design

Operating new mines with tailings that demonstrate potential for carbon capture should consider integrating their processing mills with reactors for the carbonation of tailings. Proper energy and mass balances should be performed such that the energy from mill/carbonation can be harnessed back into the reactor to enhance the weathering reaction of tailings [183–185]. Mines should properly design their reactors for enhanced weathering of fresh tailings when dumped by machinery. Regulations will continue to be stringent in the future so that incentive and profits for carbon capture will become common.

5.3.2. Expose fresh tailings to fast carbonation

To quickly carbonate large quantities of tailings, fresh tailings from the mineral processing plant mill should be spread in thin layers, made porous in grates, stirred, purged with CO_2, and sprinkled with water before they are buried beneath new tailings [180]. The main challenge encountered in spreading them is that they require very high surface areas to achieve high storage rates [152,180]. To mitigate this challenge, carbon capture and injection of pure CO_2 rather than natural atmospheric CO_2 should be the norm [169,186,187].The dissolution of mine tailings with acids is another potential strategy for the rapid reaction of silicate minerals [173,184,186].An example of the proposed design for tailings carbonation with CO_2 injection is shown in Fig. 11.

5.3.3. Chemical, thermal and mechanical activation

Large fractions of tailings suitable for carbonation are in the form of passive silicates with high Mg content and require temperature, mechanical, and chemical activation to accelerate carbonation. The passivation layers can be removed to expose fresh surfaces for reaction by mechanical exfoliation (e.g., stirring the bed) or chemical reactions [188–190]. Preheating treatment can also be used to convert passive silicates to reactive minerals that are suitable for mineral carbonation;

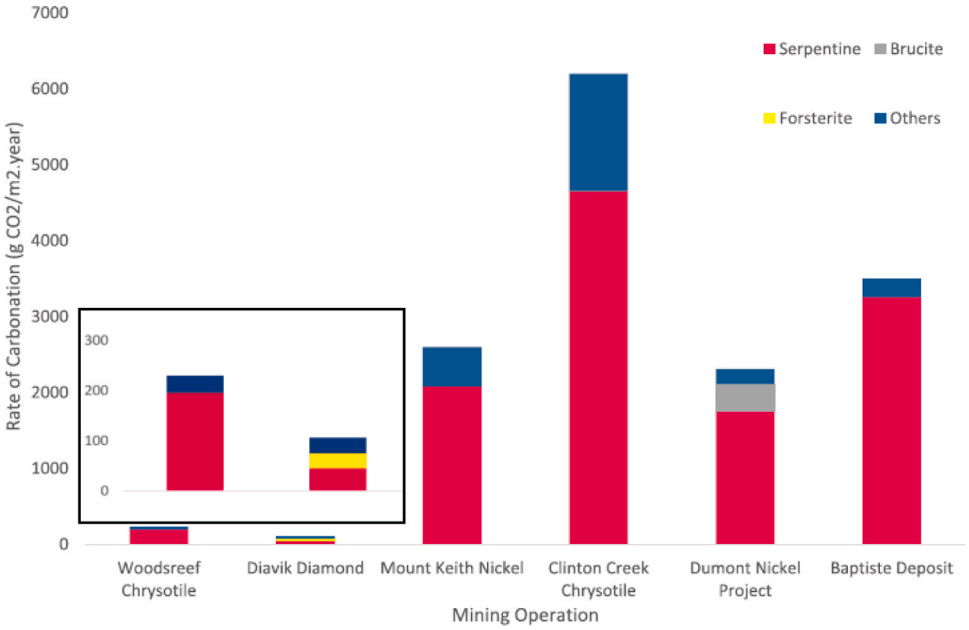

Fig. 9. Passive carbonation rates and compositions of major mineral phases in major ultramafic mine residues [172].

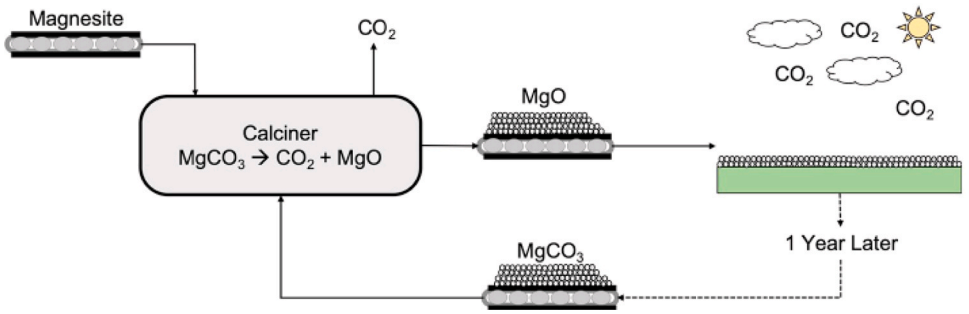

Fig. 10. Schematic of potential use of mine tailings to capture carbon dioxide from the atmosphere [19].

for example, serpentine forms MgO, which readily reacts without passivation [191–193].

5.3.4. *Microbiological approaches*

Recently, there has been much interest in tailing carbonation enhancement by introducing microorganisms; this process was found to be successful in the laboratory [194,195]. Biofilms (groups of microbes), including cyanobacteria and ureolytic bacteria, extract nutrients from tailings (dissolving them) by breaking down minerals to increase the alkalinity required to precipitate carbonates [17,181,196]. In addition, the weathering process forms porous polymeric substances, which are important nucleation sites for carbonation and soil rejuvenation. Thus, there is a huge potential for using these microbes, even leachates from mine tailings in bioreactors, to accelerate carbonation [195,197]. However, the main challenge is to scale up from the laboratory to the mine level, which should be guided by the carbonation level.

In summary, mining tailings have huge potential for offsetting greenhouse gas emissions. This can be achieved through the proper design of reactors in mines with tailings suitable for carbon storage. Much research effort is still needed to quantify all tailings with the capability to store stored dioxide. Some suitable mine tailings are far from carbon-emitting sites; these tailings can be effectively used for carbon capture from the atmosphere to obtain pure CO_2 for injection into bulk tailings. There is a challenge in the large-scale implementation of this technology: research should move from a laboratory-to-field scale. The main approaches with high potential include [1] direct air capture of atmospheric carbon CO_2 and mineral carbonation using CO_2-rich gases, [2] thermal activation using exothermic energy generation from mineral carbonation, and [3] the use of microorganisms to accelerate weathering.

6. Future perspectives and research directions

CO_2-based mineral processing technologies have immense potential for mitigating carbon emissions while simultaneously improving the extraction technology of valuable minerals. Several trends and challenges may shape the future development and adoption of these technologies. Further R&D should focus on enhancing the efficiency and scalability of CO_2-based mineral processing techniques. Catalyst design, reactor engineering, and process optimization can significantly improve the economic and environmental performances of these technologies. Using the CO_2 emitted from industrial sources as feedstock for mineral processing could play a significant role in reducing greenhouse gas emissions while producing valuable minerals.

Adoption of CO_2-based processing technologies hinges on market demand for sustainable mineral extraction and regulatory policies targeting carbon emission reduction. Stakeholders could incentivize adoption through subsidies or regulatory frameworks to reduce costs and enhance economic viability. Reducing capital and operating costs, improving energy efficiency, and identifying value-added products to enhance the economic viability of these processes can encourage the use of this technology. Research efforts should focus on identifying suitable

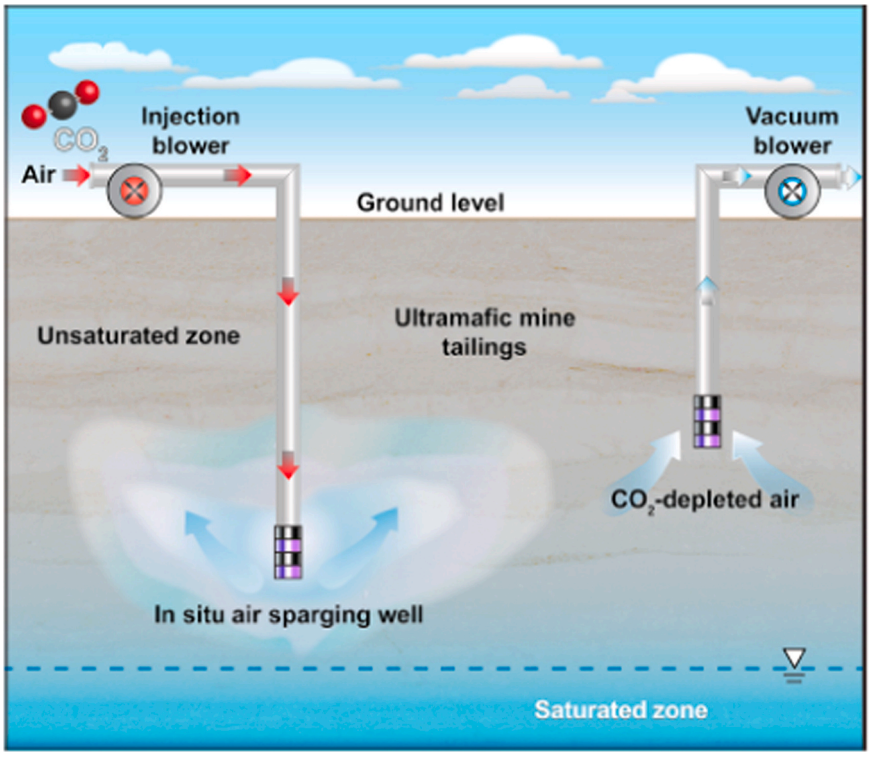

Fig. 11. Tailings Carbonation with Stirring and CO_2 Injection [180].

mineral deposits, developing efficient processing techniques, and improving catalyst materials (with improved activity, selectivity, process intensification, and stability) for CO_2 mineralization reactions tailored to specific mineral compositions and conditions to enhance process efficiency and mineral recovery rates.

Research gaps exist in the understanding of the long-term environmental consequences of this technology. Consequences, such as potential changes in soil and water quality, biodiversity, and ecosystem services. Advanced characterization techniques, life cycle assessment, techno-economic analysis, and collaborative research initiatives can enable a better understanding of CO_2-based mineral processing systems, guide decision-making and technological development efforts, and foster interdisciplinary collaboration to address key research gaps and challenges.

7. Conclusions

The drive to achieve carbon neutrality has garnered considerable attention and support from various industrial and research sectors. The captured CO_2 has been expected to have two destinations; sequestration in geological landforms and usage in various industrial and waste management processes. Due to risks of leakages of the stored CO_2 in various landforms, research has shifted toward CO_2 use as a more lasting solution for captured CO_2. As highlighted in this review, the mining sector, which is a major CO_2 emitter, can play a crucial role in the uptake and use of captured CO_2. Processes such as flotation, wastewater treatment, leaching, and mineral carbonation can enhance productivity while maintaining a low CO_2 footprint. Among these, mineral carbonation has emerged as the most promising method for utilizing CO_2. It is essential for key stakeholders to adopt an initiative to manage mine tailings through carbonation to enhance the reaction process. Additionally, investing in research in this sector is vital for achieving meaningful results.

CRediT authorship contribution statement

Ndlovu Phakamile: Writing – review & editing, Formal analysis, Conceptualization. **Chabalala Mandla:** Writing – original draft, Visualization, Methodology. **Rampou Mohau:** Writing – original draft. **Khumalo Siphesihle:** Writing – original draft. **Fashu Simbarashe:** Writing – review & editing, Formal analysis, Conceptualization. **Hungwe Douglas:** Writing – review & editing, Supervision.

Declaration of Competing Interest

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

Data availability

No data was used for the research described in the article.

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