



Review

# Integrated Mineral Carbonation of Ultramafic Mine Deposits—A Review

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**Abstract:** Recently, integrated mineral carbonation for CO<sub>2</sub> sequestration has received significant attention due to the high potential for commercialization towards mitigating climate change. This review compiles the work conducted by various researchers over the last few years on integrated mineral carbonation processes in the mining industry, which use ultramafic mine wastes as feedstock for mineral carbonation. Here, we introduce the basic concepts of mineral carbonation including a brief description of the process routes and pre-treatment techniques. We discuss the scope of integrated mineral carbonation process application, and critically review the integrated mineral carbonation process in the mining industry including modified passive carbonation techniques in tailing storage facilities, and ex-situ carbonation routes using fresh tailings. The focus of the discussions is the role of reaction condition on the carbonation efficiency of mine waste with various mineralogical compositions, and the benefits and drawbacks of each integrated mineral carbonation process. All discussions lead to suggestions for the technological improvement of integrated mineral carbonation. Finally, we review the techno-economic assessments on existing integrated mineral carbonation technologies. Research to date indicates that value-added by-products will play an important role in the commercialization of an integrated mineral carbonation process.

**Keywords:** CO<sub>2</sub> sequestration; mineral carbonation; ultramafic tailings; mine waste

## 1. Introduction

The global climate has been warming at a faster pace than in any other period since 1850 [1]. The main cause of climate change is burning fossil fuels to meet global energy demands, which disturbs the balance of the carbon cycle and increases the concentration of greenhouse gases (especially CO<sub>2</sub>) in the atmosphere [2]. The atmospheric CO<sub>2</sub> concentration reached 404 ppm in October 2017—44.3% above pre-industrial levels (280 ppm) [3]. At the same time, the average global temperature shows a warming of 0.8 °C [4]. If no additional efforts are made to constrain or manage anthropogenic greenhouse gas emissions, the CO<sub>2</sub> equivalent concentration is expected to exceed 1000 ppm by 2100, which would result in a global mean surface temperature increase of 2.6 to 4.8 °C [1].

International efforts to mitigate climate change began in earnest in 1992 when the United Nations Framework Convention on Climate Change (UNFCCC) was introduced and adopted. UNFCCC aimed at “stabilized greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system” [5]. The CO<sub>2</sub> emission targets of UNFCCC-involved countries were outlined for the period of 2020 to 2030 in the Paris Agreement [6],

which was negotiated with the awareness that global warming is a serious threat, and its ultimate goal was to limit global warming to less than 2 °C while pursuing efforts to limit the temperature rise to 1.5 °C relative to pre-industrial levels [6]. However, to reach these goals, the global community must find affordable and practical solutions for managing carbon.

Currently, various strategies and solutions have been adopted by countries for the purpose of reducing anthropogenic CO<sub>2</sub> emissions. These approaches include: improving energy efficiency; using energy sources that are less carbon-intensive (e.g., natural gas, hydrogen, and nuclear power) or renewable (e.g., solar, wind, hydropower, geothermal and bio-energy); enhancing biological sinks (i.e., afforestation and reforestation); and CO<sub>2</sub> capture and storage (CCS) [7]. Of these approaches, CCS is the only option that can meet the enormous demand for CO<sub>2</sub> reduction. It is estimated that CCS could reduce 85–90% of CO<sub>2</sub> from large-emission sources and energy-intensive emitters [7]. In the CCS process, CO<sub>2</sub> is concentrated from diluted sources and transported to storage locations. Research on and demonstration of CCS technology has grown significantly in recent years with the hope that it can one day become economically feasible [8].

Since burning of fossil fuels causes a carbon flux from the lithosphere to the atmosphere, strategies and solutions that reverse this process by creating a carbon flux from the atmosphere back to the lithosphere can permanently solve the current anthropogenic CO<sub>2</sub> problem and mitigate global warming. CO<sub>2</sub> sequestration by mineral carbonation is such a process that could permanently store CO<sub>2</sub> and establish a “circular economy” [9]. The mining industry could potentially apply mineral carbonation using ultramafic mine wastes and offset their greenhouse gas emissions. This paper reviews the current research on integrating mineral carbonation in the mining industry. First, the development of mineral carbonation is briefly introduced. Then, the integrated mineral carbonation in mining industry is reviewed, with special focus on modified passive carbonation method and ex-situ mineral carbonation. Finally, a techno-economic assessment is provided.

## 2. Mineral Carbonation

Mineral carbonation has been developed by harnessing the natural weathering process of magnesium, calcium and iron oxide-based silicate minerals, which transforms atmospheric CO<sub>2</sub> into carbonate minerals [10]. Equation (1) shows the simplified reaction process of mineral carbonation.

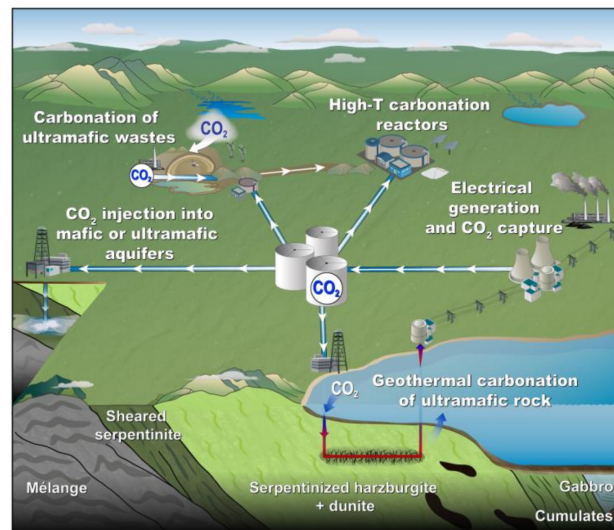


Comparing with other approaches of CCS (e.g., geological storage), mineral carbonation has several advantages and merits. First, mineral carbonation is the only option that can permanently store CO<sub>2</sub> without monitoring [11]. The carbonate products are naturally abundant minerals, which are environmentally friendly and leakage-free for geologic timescales. In addition, the abundance of Mg/Ca-silicates on Earth offers enormous capacity for sequestering CO<sub>2</sub> [12]. There is a wide range of materials that can be used for mineral carbonation, not only naturally occurring formations, such as olivine, serpentine and wollastonite, but also highly reactive wastes comprised of Mg/Ca-rich materials, such as fly ash, iron and steel slags, cement kiln dust, and ultramafic mine wastes [13]. In addition, the carbonation process generates heat, namely exothermic reaction (Equation (1)), and the reactions can be self-perpetuated without additional energy input [14]. Theoretically, the heat of exothermic carbonation can be utilized in the endothermic mineral dissolution, but this has not yet been demonstrated in practice. The only drawback of mineral carbonation is that the reaction kinetics are often too slow and thus research has focused on accelerating carbonation rates.

### 2.1. Process Routes

Mineral carbonation was proposed by Seifritz [15] in 1990. Since this date, numerous researchers have attempted to accelerate the reaction between CO<sub>2</sub> and alkaline minerals in two broad environmental conditions, in-situ and ex-situ [14,16]. In-situ mineral carbonation injects CO<sub>2</sub>

into mantle peridotite and basalts deposits, and the reaction occurs in an underground, possibly hydrothermal, environment. Conversely, ex-situ mineral carbonation is achieved in a separate reactor or an industrial process where the operational conditions are easy to control [17]. Ex-situ mineral carbonation can also apply CO<sub>2</sub> as a carbon source for chemical production (e.g., carbonates), which can be used as building materials, aggregates, and cementitious phases [18,19]. Figure 1 shows a conceptual diagram depicting CCS strategies via mineral carbonation. Developments in process routes have been reviewed in numerous papers related to mineral carbonation [20,21].



**Figure 1.** Conceptual diagram depicting geoenvironmental system designed for CCS process via mineral carbonation. Modified from Power et al. [22].

Ex-situ mineral carbonation has focused on two broad methods: direct and indirect mineral carbonation. Direct mineral carbonation is completed in one single reaction step [23], whereas indirect carbonation introduces an additional step by extracting the reactive compound [20]. Both direct and indirect methods have two different possible routines, namely a dry process and an aqueous process, which have different CO<sub>2</sub> sorption characteristics. In a dry process, CO<sub>2</sub> molecules are chemisorbed to the solid by strong chemical bonds or physisorbed by weaker inter-molecular bonds due to the interactions between CO<sub>2</sub> molecules and a sorbent [24]. In aqueous sorption, CO<sub>2</sub> is dissolved into the solvent first, and then is reacted with olivine or serpentine [25].

At present, mineral carbonation has only reached the demonstration and pilot scales [21]. Although no economically viable mineral carbonation process has been developed so far, some attractive concepts and experimental results suggest the possibility for scaling up and commercializing the carbonation process over the next decade [26]. For example, the Carbfix pilot project in Iceland can carbonate 95% of injected CO<sub>2</sub> in less than two years [27]. The direct aqueous carbonation route proposed at National Energy Technology Laboratory, formerly Albany Research Council in the U.S., has been evaluating the most promising ex-situ process for industrial application to date [23], which is commonly used as a baseline for comparison of other carbonation technologies. The indirect two-step pH-swing carbonation method developed at Finland's Åbo Akademi University has produced results that are able to compete with the direct aqueous carbonation process developed at National Energy Technology Laboratory when using serpentine to trap CO<sub>2</sub> [28]. In cases where geological storage is not applicable, mineral carbonation processes could be an alternative option for the storage of small to medium CO<sub>2</sub> emissions (<2.5 Mt CO<sub>2</sub> per site) [21].

## 2.2. Pre-Treatment

In some cases, the reactivity of Ca/Mg-silicates, particularly serpentine, cannot be enhanced sufficiently by the careful selection of a process route and conditions alone. Pre-treatment options can improve reaction kinetics for some potentially attractive Ca/Mg-silicates by activating the mineral for carbonation [29]. All pre-treatment options either aim to destroy the mineral lattice or create disorder in crystal structure, thus increasing the specific surface area, freeing some of the required cations (e.g., Ca and Mg), and creating an amorphous phase [23]. Pre-treatment can be conducted by thermal [30,31], chemical [32,33], or mechanical means [34–36].

Thermal activation removes chemically-bound water, which can increase the porosity and the resulting surface area. Thermal treatment is suitable only for serpentine group minerals, which contain up to 13 wt % chemically bound water. By heating these to 600–700 °C, the hydroxyl groups of serpentine are removed [37], thereby significantly increasing the carbonation rate. The optimum temperature for thermal activation depends on the serpentine composition. Extremely high temperature will lead to re-crystallization and needs to be avoided for the purpose of activation. There are some thermal activation process other than heat treatment including steam [38] and microwave treatment [11].

Chemical activation aims at polarize and weaken the calcium or magnesium bonds within the silicate structure. As a result, the solubility of magnesium silicate minerals increases, thus overcoming its slow dissolution kinetics. An array of chemicals has been proposed to enhance mineral carbonation. Chemicals that have been used for extracting magnesium or calcium from silicate minerals include inorganic acids (HCl, H<sub>2</sub>SO<sub>4</sub>, and H<sub>3</sub>PO<sub>3</sub>), organic acids (HCOOH, CH<sub>3</sub>COOH, and CH<sub>3</sub>COOH), bases (NaOH and KOH), and salts (NH<sub>4</sub>Cl, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, and NH<sub>4</sub>NO<sub>3</sub>) [20,33]. It is commonly recognized that sulfuric acid is the best extraction agent, and that it can extract more than 70% of the magnesium from serpentine and produce a silica by-product with a high surface area (an increase in surface area from 8 to 330 m<sup>2</sup>/g). For selective leaching, ammonium salts have performed better than sulfuric acid [39]. However, chemical activation tends to form a silica gel, which may trap the required cations needed for mineral carbonation.

Mechanical activation via high-energy attrition grinding not only decreases particles size and increases the surface area of minerals, but also introduces imperfections into the crystal lattice [40,41]. The mechanical activation of serpentine often focuses on the structural changes caused by losing –OH molecules [36,42]. Studies on the use of mechanical activation on ultramafic rocks are constantly expanding. They have focused on comparing individual mineral (e.g., olivine or serpentine) [36,43] responses to the grinding energy input [44,45] and milling instruments (e.g., planetary mills, vibration mills, attrition mills, or tumbling mills) [34,40], as well as milling atmosphere (e.g., N<sub>2</sub>, CO<sub>2</sub> or air) [46], solutions (e.g., water, alcohol or acid) [43,47], and temperatures [48].

The combination of two pre-treatment methods, such thermal-chemical activation [49], chemical-mechanical activation [50], thermal-mechanical activation [51] can achieve unexpected activation effects. Except for the above mentioned pre-treatment methods, some novel processes for enhancing the rate of mineral carbonation have been proposed for waste materials [52]. For example, co-utilization with wastewater (or brine solution) [53]; biological enhancement using enzymes and microorganisms [54]; reactor integration, such as rotating packed bed [55]; ultrasonic vibration [56]; and electrolysis treatment [57]. Although these novel treatment methods could not activate materials up to the level that the former three techniques could reach, they are promising for the industrialization application as Ca/Mg containing waste materials (e.g., steel slag, fly ash, ultramafic tailings, etc.) are already related to industrial operations (e.g., mines), which could dramatically reduce the cost of mineral carbonation.

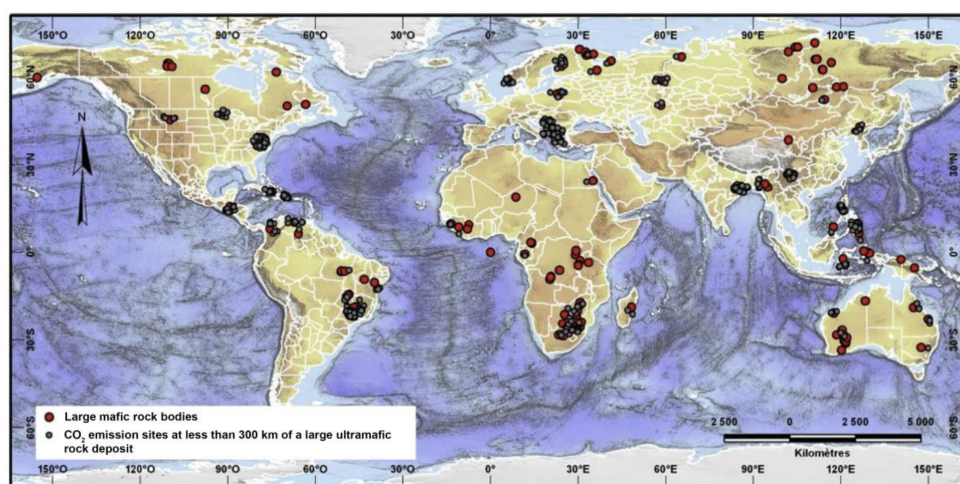
## 3. Integrated Mineral Carbonation in Mining

As a carbon-intensive industry, mining is facing ever-increasing pressure to reduce CO<sub>2</sub> emissions as a result of emerging carbon management policies [58]. Integrated mineral carbonation processes

in the mining industry that use mine wastes (e.g., waste rock, tailings, and slag) to trap CO<sub>2</sub> [59] are advantageous for their ability to manage CO<sub>2</sub> emissions and remediate potentially hazardous materials (e.g., chrysotile) [60]. By using mine wastes, especially tailings, many of the costs associated with the comminution required for mineral carbonation are absorbed as part of conventional ore processing. This has a large impact on the costs associated with altering tailings to a carbonate form. There is also the possibility that carbonation of mine wastes could create profit through carbon credits, which opens the opportunity to turn marginal projects, such as low-grade nickel deposits, into economically viable operations [61,62].

### 3.1. Suitable Mine for Mineral Carbonation

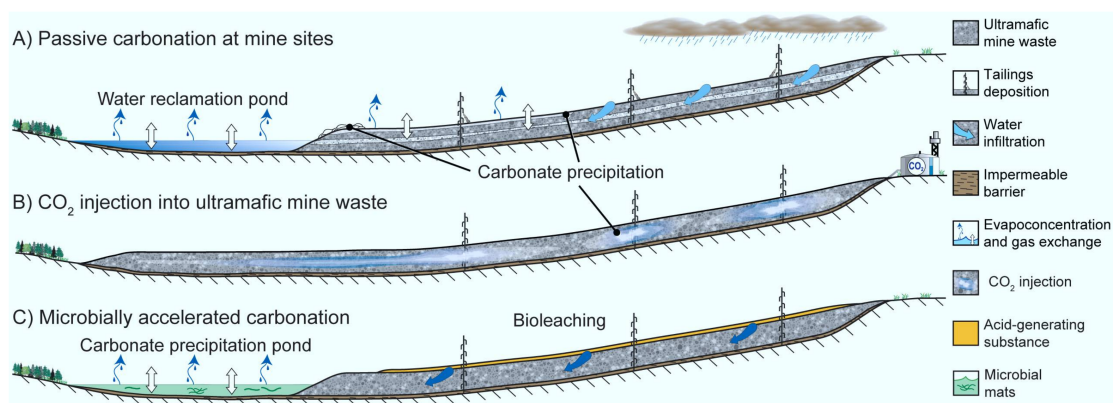
The ultramafic rock-hosted ore deposits that are favored for integrated mineral carbonation technologies include chrysotile [63], nickel [64–66], chromium [67,68], diamond [69] and platinum group element (PGE) mines [70,71]. These mine types are abundant worldwide and their CO<sub>2</sub> storage potential is large [61]. Figure 2 shows the potential of large and super-large ultramafic mine sites that are suitable for the integrated mineral carbonation process [72]. Notably, the annual amount of mining tailings produced globally could potentially offset approximately 1.5% of annual global CO<sub>2</sub> emissions [73]. The principle minerals being targeted for CO<sub>2</sub> sequestration in the ultramafic mine wastes are olivine and serpentine group minerals.



**Figure 2.** Large ultramafic rock-hosted ore deposits lying less than 300 km from a CO<sub>2</sub>-emission site. Reprinted with permission from Bodéan et al. [74]. Copyright 2014 Elsevier Ltd.

### 3.2. Modified Passive Carbonation Method

Weathering and carbonation of ultramafic mine wastes may occur passively, within tailings storage facilities at both closed and active mines without human mediation [75]. Passive carbonation relies on the exposure of tailings to the Earth's atmosphere, hydrosphere, and biosphere, thereby allowing natural weathering and gradual carbonation (Figure 3). Field studies have demonstrated that certain mines can offset approximately one-tenth of their annual CO<sub>2</sub> emissions through passive carbonation of tailings [76]. These offsets are achieved through the precipitation of hydrated Mg-carbonate minerals including lansfordite [MgCO<sub>3</sub>·5H<sub>2</sub>O], nesquehonite [MgCO<sub>3</sub>·3H<sub>2</sub>O], dypingite [Mg<sub>5</sub>(CO<sub>3</sub>)<sub>4</sub>(OH)<sub>2</sub>·5H<sub>2</sub>O], and hydromagnesite [Mg<sub>5</sub>(CO<sub>3</sub>)<sub>4</sub>(OH)<sub>2</sub>·4H<sub>2</sub>O]. These minerals are commonly seen as efflorescent crusts associated with the tailings and are stable for long periods [77–79].



**Figure 3.** A schematic of three mine tailings facilities depicting: (A) passive carbonation under conventional tailings management; (B) CO<sub>2</sub> injection; and (C) biological approaches incorporating bioleaching and microbial carbonate precipitation. Modified from Power et al. [22].

Passive carbonation rates in ultramafic tailings storage facilities are highly variable and are controlled by a variety of factors including: (1) tailings mineralogy and physical properties; (2) mineral processing and mine design; (3) tailings handling and storage (e.g., subaerial versus subaqueous); and (4) local climate. Passive carbonation similar to other forms of carbonation may be limited by: (1) CO<sub>2</sub> supply; (2) mineral dissolution; or, in rare cases, (3) carbonate precipitation [80]. For example, passive carbonation at the Mount Keith Nickel Mine in Western Australia is limited by CO<sub>2</sub> supply as evidenced by experiments mimicking pore water chemistry and stable carbon and isotopes of carbonates from the site [76]. Rate-limiting processes may also work somewhat in concert to limit carbonation. For instance, tailings at the Diavik mine are mainly stored sub-aqueously, which limits CO<sub>2</sub> ingress that in turn limits carbonic acid generation that would aid mineral dissolution [69]. Thus, rate limitations are typically a function of tailings mineralogy and storage conditions. It is necessary to determine the rate limitation to devise an appropriate acceleration strategy to optimize carbonation [80].

Carbonation of mine tailings that only contain relatively less reactive silicate minerals (e.g., olivine and serpentine group minerals) compared to oxide and hydroxide minerals will tend to be limited by mineral dissolution. Mg-silicate minerals such as forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) and serpentine group minerals [Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>] have pH-dependent dissolution rates that are much faster than other silicate minerals, yet their dissolution at Earth's surface conditions is still sluggish [22,81]. There are few economically viable options for accelerating mineral dissolution of silicate minerals. One possibility is the application of acid-generating materials that are colonized by *Acidithiobacillus* spp., which can dramatically enhance magnesium leaching from silicate minerals [82,83] (Figure 3). Grinding to a finer grain size to increase metal recovery would also accelerate mineral dissolution by creating greater surface area [84–86]. Enhancing mineral dissolution rates to increase the flux of available cations remains the greatest challenge in implementing mineral carbonation technologies at mine sites.

One alternative strategy is referred to as “enhanced weathering”, which is a geoengineering approach that relies on mining, pulverization, and distribution of ultramafic and mafic rock to remove CO<sub>2</sub> from the atmosphere as dissolved inorganic carbon (e.g., HCO<sub>3</sub><sup>−</sup>) by accelerating natural weathering [87,88]. This strategy “accepts” that silicate dissolution is inherently slow, but relies on extended “reaction times” in the environment, e.g., dissolution continues until a mineral grain is fully dissolved. This strategy could be implemented with the use of preexisting ultramafic mine tailings, which would eliminate the need for superfluous mining.

Certain mine tailings contain highly reactive phases such as oxide and hydroxide minerals. For instance, the Mount Keith tailings contain a minor abundance of brucite [Mg(OH)<sub>2</sub>], which has a pH-dependent dissolution rate that is orders of magnitude faster than olivine or serpentine

group minerals [22]. In laboratory experiments, brucite carbonation remains CO<sub>2</sub>-limited even when exposing it to CO<sub>2</sub>-rich fluids [89,90]. Thus, acceleration can be achieved by increasing the rate CO<sub>2</sub> supply. Firstly, a CO<sub>2</sub> source must be identified, whether only atmospheric CO<sub>2</sub> is available or a point source of CO<sub>2</sub>-rich gas (e.g., flue gas from diesel-burning generators on site). In the former case, passive carbonation can be accelerated through simple tailings modification, such as increasing the surface area of tailings, optimizing tailings distribution, and enhanced aeration of the tailings (e.g., tilling) [75]. These tailings management practices increase the diffusion of atmospheric CO<sub>2</sub> into tailings pore waters, which allows for greater carbonation. These practices contradict conventional tailings management that generally aims to limit tailings exposure to the atmosphere. However, ultramafic mine tailings are usually benign (e.g., very low acid-generating potential/high neutralization potential) and pose no substantial risk to the environment with the possible exception of asbestos and chromite mine wastes. Alternatively, point sources with CO<sub>2</sub>-rich gasses could be utilized (Figure 3) [89]. Some mines that are off-the-grid will generate electricity using fossil fuel-fired generators or will be within a reasonable distance to other point sources of CO<sub>2</sub> (Figure 2). These CO<sub>2</sub>-rich gasses could be injected into tailings, incorporated into ore processing, or used in specially designed reactors following ore processing [91]. CO<sub>2</sub> supply could be further enhanced by accelerating the hydration of dissolved CO<sub>2</sub> gas to form a hydrated phase (e.g., HCO<sub>3</sub><sup>-</sup>). For instance, using carbonic anhydrase catalyzes the hydration of aqueous CO<sub>2</sub> [54,90].

In general, carbonation in ultramafic mine wastes is not limited by carbonate precipitation. The kinetic reactions are sufficiently fast that there generally is no need for acceleration. For example, nesquehonite has little kinetic inhibition and will begin precipitating soon after a solution becomes saturated [92]. However, precipitation of magnesite (MgCO<sub>3</sub>), the stable form of magnesium carbonate, is kinetically inhibited at Earth's surface conditions [92]. Thus, magnesite formation in ultramafic tailings does not occur even if tailings pore waters are supersaturated with respect to magnesite, which is often the case at ultramafic mines. Overcoming this kinetic inhibition would allow for greater carbonate formation. For example, utilizing surfaces with high carboxyl site densities has been shown to accelerate magnesite formation in the laboratory at room temperature within 10 s of days [93]. Other biological approaches could be harnessed to increase carbonate deposition in mining environments. For example, cyanobacteria can mediate Mg-carbonate precipitation in the natural environment (Figure 3) [60,94]. Moreover, the by-products of carbonation, such as photoautotrophic biomass, can be harvested as biofuel [94]. Alternatively, seeding with carbonate promotes mineral nucleation [95]; and periodically adding small amounts of water keeps partial pore saturation at optimum levels [96,97], which promotes the conversion of CO<sub>2</sub> gas to a hydrated form.

The main advantage to these modifications that accelerate passive carbonation is that they operate at ambient temperature and pressure conditions, and therefore the energy consumption should be low. While these strategies do not aim to achieve complete carbonation of the tailings, there is the potential for substantial offsets at a cost that aligns with current carbon prices and are thus likely to be economically feasible. Importantly, not all tailings will be suitable for these strategies; for example, tailings that are highly carbonated [82] or those that contain abundant sulfides [98]. Careful selection of the mine site and carbonation process are necessary first steps.

### 3.3. Ex-Situ Mineral Carbonation

In an active mining operation, it is possible to apply the ex-situ carbonation to fresh tailings, which are more active for carbonation than weathered ones due to minimal authigenic carbonate content [82]. Accelerated mineral carbonation includes feedstock pre-treatment (e.g., thermal, chemical and/or mechanical activation), followed by a direct or indirect carbonation process in gas–solid phases or aqueous phases at high temperature and high pressure. These advanced technologies have been adopted to estimate the suitability of mine waste materials as feedstock for CO<sub>2</sub> sequestration (Table 1).

**Table 1.** Summary of current studies on ex-situ mineral carbonation of mine waste materials.

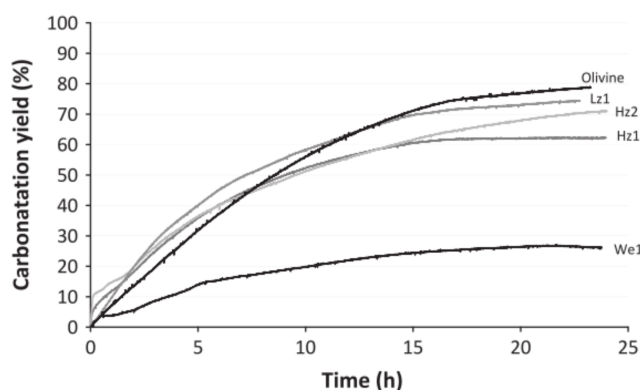
Mining Deposits	Majority Minerals	CO <sub>2</sub> Source	Rock Pretreat	Carbonation Method	Ref.
American Chrome, QC	Serpentine	Flue gas	TA	DC gas–solid	[99]
Old ilmenite mine, QC	Anorthite	Flue gas	NA	DC gas–solid DC aqueous	[100]
Okanogan nickel deposit, WA; Thompson nickel bell, MB	Serpentine	Pure CO <sub>2</sub>	TA, CA	DC aqueous	[64]
American Chrome, QC	Serpentine	Flue gas	TA	DC aqueous	[67,101]
Black Lake mine, QC	Serpentine	Flue gas	TA, MS	DC gas–solid	[102]
Lonmin Platnum mine, South Africa	Enstatite, plagioclase feldspar	Pure CO <sub>2</sub>	CA	IDC aqueous	[70]
Nickel Slag, New Caledonia	Olivine, serpentine	Pure CO <sub>2</sub>	TA, CA, MA	DC aqueous	[74]
Turnagain deposit, BC	Olivine, serpentine	Pure CO <sub>2</sub>	NA	DC aqueous	[103]
Turnagain deposit, BC	Olivine, serpentine	Pure CO <sub>2</sub>	MA	DC aqueous, DC gas–solid	[104,105]
Troodos ophiolite complex, Cyprus	clinopyroxene, anorthite	Pure CO <sub>2</sub>	MA	DC gas–solid	[106]
Black Lake mine, QC; Dumont Nickel project QC	Chrysotile, lizardite	Flue gas	NA	DC gas–solid	[66]
Thetford Mines, QC	Chrysotile	CO <sub>2</sub> mix	TA	DC gas–solid	[63]
Hitura nickel mine, Finland	Serpentine	Pure CO <sub>2</sub>	CA	IDC aqueous	[107]

Note: TA is thermal activation, CA is chemical activation, MA is mechanical activation; MS is magnetic separation, NA is not applied, DC is direct carbonation, IDC is indirect carbonation.

Current investigations into integrated mineral carbonation processes mostly focus on mine waste that contains serpentine and brucite [11,91]. Currently, mine waste materials consisting of partially-serpentinized olivine draws the most attention [108]. Styles et al. studied the behavior of various ultramafic rock compositions in acid leaching in the presence of ammonium bisulfate. The Mg leaching extent after 1 h was: lizardite (78%) > serpentinized peridotites (65%) > forsterite (55%) > antigorite (40%) > pyroxene- and amphibole-rich rocks (25%) [109]. Bodénan et al. have examined the reactivity of variably serpentinized olivine in a direct aqueous carbonation process. They found that the degree of serpentinization made no difference on the performance of carbonation when an attrition stirrer was applied during carbonation (Figure 4) [74]. Li and Hitch found that the reactivity of partially-serpentinized olivine is higher than that of pure olivine in a direct aqueous carbonation process, due to large surface area and poorly ordered structures induced by serpentinization [84–86]. As partially-serpentinized olivine is the most abundant form of ultramafic rock in nature [109], it is of great significance and provides the potential for ex-situ mineral carbonation to become widely employed in the mining industry. Thus, accurate mineralogical characterization of mine waste materials is necessary for process development.

The integrated mineral carbonation process proposed for carbonation of ultramafic mine wastes takes advantage of the technologies developed for ex-situ mineral carbonation of natural occurring ultramafic rock and innovates according to its own properties. The most common methods for mine waste pre-treatment are thermal activation using calcination [67], microwave heating [11], steam mediation [110], and chemical activation using organic or inorganic acids [11]. These pre-treatment methods greatly accelerate serpentine carbonation. However, pre-treatment methods, such as thermal activation and chemical activation by organic ligands, may not appreciably accelerate the reaction rate of olivine [111]. Li studied the mechanical activation on mine waste materials consisting of partially-serpentinized olivine. Regardless of milling conditions, forsterite, the olivine mineral variety in the mine waste, was found to be the main mineral being mechanically-activated and carbonated. It was determined that lizardite, a hydrated magnesium silicate also common in ultramafic-hosted mineral deposits, acted as catalyst by assisting forsterite reaching high levels of activation. This condition generated a greater CO<sub>2</sub> conversion to carbonate than that of pure olivine with the equal specific milling energy input [112].



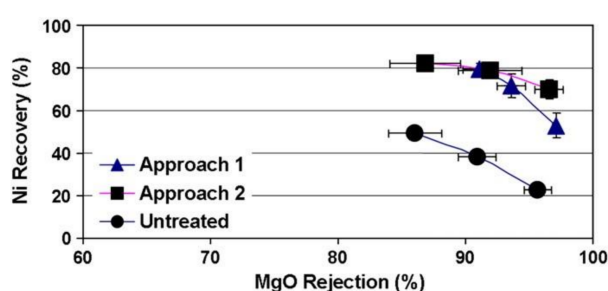


**Figure 4.** Kinetics of carbonation of various partially-serpentinized olivine as a function of time ( $T = 180\text{ }^{\circ}\text{C}$ ,  $P_{\text{CO}_2} = 2\text{ MPa}$ ,  $90\text{ g/L}$ ) with a continuous passivation layer exfoliation scheme. Hz is Harzburgite, We is Wehrlite, and Lz is Lherzolite. The ratio of serpentine to olivine from high to low is:  $\text{Hz2} > \text{Hz1} > \text{We1} > \text{Lz1} > \text{Olivine}$ . Reprinted with permission from Bodénan et al. [74]. Copyright 2014 Elsevier Ltd.

Amongst various ex-situ mineral carbonation routes, the direct carbonation method is commonly applied in the current study of the integrated mineral carbonation process (Table 1). The direct carbonation route is simple and easy to operate in industry. It provides the potential to store  $\text{CO}_2$  directly from a flue gas [102]. This will save the  $\text{CO}_2$  capture and concentration cost, which take up to 9–30% of the total cost of the integrated mineral carbonation process [62,113]. Li and Hitch [104,114] studied the direct gas–solid carbonation of mine waste using carbon dioxide adsorption isotherm at  $25\text{ }^{\circ}\text{C}$ . They systematically analyzed the effect of surface area, micropore volume, and weight ratio for Mg to Si, and forsterite crystal structure disorder on the  $\text{CO}_2$  adsorption capacity (or  $\text{CO}_2$  sequestration efficiency). However, they only used pure  $\text{CO}_2$  for the study and the process is not optimized. Veetil et al. [102] studied the direct gas–solid carbonation of activated serpentine mining residue using a simulated cement flue gas. They optimized the operation temperature ( $T$ ),  $\text{CO}_2$  partial pressure ( $P_{\text{CO}_2}$ ) [102], and the carbon dioxide–water vapor ratio ( $P_{\text{CO}_2}:P_{\text{H}_2\text{O}}$ ) [99]. However, the  $\text{CO}_2$  sequestration efficiency only reached  $0.07\text{ g CO}_2/\text{g residue}$  after carbonation for 1 h at the optimum condition ( $T = 200\text{ }^{\circ}\text{C}$ ,  $P_{\text{CO}_2} = 0.47\text{ MPa}$ ,  $P_{\text{CO}_2}:P_{\text{H}_2\text{O}} = 1.8$ ), due to the poor gas–solid interaction. Pasquier et al. [67] pursued the direct aqueous carbonation of thermal-activated serpentine mining residue in a stirred reactor using a simulated cement flue gas at low temperature and pressure. The  $\text{CO}_2$  sequestration efficiency was enhanced to  $0.28\text{ g CO}_2/\text{g residue}$  after carbonation for 15 min at the optimum condition ( $T = 22\text{ }^{\circ}\text{C}$ ,  $P_{\text{CO}_2} = 0.19\text{ MPa}$ ,  $P_{\text{CO}_2}:P_{\text{H}_2\text{O}} = 1.8$ ) [101]. Kemache et al. [115,116] tested the direct aqueous carbonation process at pilot scale. Although they obtained a low process efficiency compared to that in the laboratory, it was a meaningful step for mineral carbonation research under real conditions. They suggest optimizing the heat-treatment and the carbonate precipitation to enhance the pilot scale process efficiency [115]. Adding a liquid recirculation process in the pilot scale plant seems to be a promising strategy and reaches a  $\text{CO}_2$  sequestration efficiency of  $0.47\text{ g CO}_2/\text{g residue}$  [117], which doubled the value obtained from the optimal conditions in the laboratory scale ( $0.28\text{ g CO}_2/\text{g residue}$ ) [101].

Some integrated mineral carbonation processes adopt the indirect aqueous carbonation process to add value to the process through recovering valuable metals and producing pure carbonate, a potential value-added product. Teir et al. [107] studied the indirect aqueous carbonation of serpentine rock from a nickel stockpile through a pH-swing process using  $\text{HCl}$  (or  $\text{HNO}_3$ ) and  $\text{NaOH}$  to adjust the pH. They successfully separated the iron oxide (88 wt %) and amorphous silica (82 wt %), and produced a high purity carbonate product (99 wt % hydromagnesite). However, this process did not recover the nickel content, which remains in the carbonated products. Recovery of nickel before carbonation is recommended [107]. Uddin et al. [50] used a combination of chemical ( $\text{H}_2\text{SO}_4$ ) and mechanical

(grinding) treatment to disintegrate the chrysotile fibers in low grade nickel ore, which reduced the pulp viscosity and enhanced the nickel recovery in downstream flotation. At the same time, the generated MgO-rich tails are potential candidate for CO<sub>2</sub> capture and sequestration. Figure 5 shows the relationship between nickel recovery and MgO rejection before and after pretreatment [50]. For the same purpose, Bobicki et al. [118,119] destroyed the serpentine content in low grade nickel ore using microwave treatment. They found that this treatment converted serpentine in ultramafic nickel ores to olivine, which improved the grindability of ores and reduced the viscosity of the ore slurries. Thus, both ore processing and mineral carbonation were improved from the microwave treatment [11]. Similarly, Mayer et al. [70] studied the indirect aqueous carbonation of platinum group metal (PGM) tailings by pH-swing process, which used both organic and inorganic acid for cation extraction. They suggested that the process would be coupled with PGM recovery in the tailings to increase the economic viability of mineral carbonation [70]. The experience in mineral carbonation of nickel ore would provide guidance to the PGM mines.



**Figure 5.** Nickel recovery vs. MgO rejection: treated (approaches 1 and 2) vs. untreated ore. Approach 1: 2-h aging and frother (DF250) addition; Approach 2: 5-min aeration, no frother for cons 1 and 2. Reprinted with permission from Uddin et al. [50]. Copyright 2012 Elsevier Ltd.

#### 4. Techno-Economic Analysis

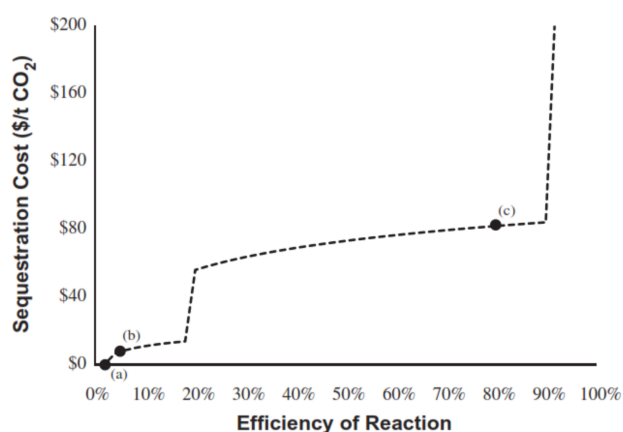
One of the major challenges for CCS is to reduce its cost for large scale application. The life cycle of CCS includes CO<sub>2</sub> capture, CO<sub>2</sub> transportation, CO<sub>2</sub> storage and utilization. Life cycle assessment on the cost of the full chain of CCS process is commonly adopted to evaluate the feasibility of a CCS project [28,74,120,121]. Table 2 lists the cost of the CCS technology as of 2014; however, it should be noted that the cost is expected to decrease as technology improves. The most cost efficient form of ex-situ mineral carbonation is still approximately six times higher compared to geological storage.

**Table 2.** The current cost of each CCS component.

CCS Component	Technology	Cost (\$/t CO <sub>2</sub> )	Ref.
CO <sub>2</sub> capture <sup>a</sup>	Post-combustion (coal-fired)	34	[7]
	Pre-combustion (coal-fired)	23	[7]
	Oxy-fuel (coal-fired)	36	[7]
	Post-combustion (gas-fired)	58	[7]
	Pre-combustion (gas-fired)	112	[7]
	Oxy-fuel (gas-fired)	102	[7]
CO <sub>2</sub> transportation	Railway	12.64	[7]
	Ship	7.48	[7]
	Pipeline	7.05	[7]
CO <sub>2</sub> storage & utilization	Geological storage <sup>b,c</sup>	8	[21]
	In-situ mineral carbonation <sup>c</sup>	17	[21]
	Ex-situ mineral carbonation	50–300	[21]

Note: <sup>a</sup> includes CO<sub>2</sub> compression to 10 MPa, <sup>b</sup> does not include monitoring costs, <sup>c</sup> includes CO<sub>2</sub> transportation cost.

The economic analysis on the integrated mineral carbonation process in mining industry was first studied by Hitch and Dipple [122], whose model shows the tradeoff of carbonation cost versus carbonation efficiency (Figure 6). In a case study of Turnagain Nickel Deposit located in northern British Columbia, Canada, the theoretical carbonation potential was estimated to be 81.9 Mt CO<sub>2</sub> over a 29-year mine life [61]. The integrating mineral carbonation process yielded a net present value of \$131.5 million at a discount rate of 8%, with an operating cost of \$82.5/t CO<sub>2</sub> [122]. A carbonation efficiency of 60% could be achieved using mechanical activation as a pre-treatment method and the mine can potentially sequester 14.6 Mt CO<sub>2</sub>/year using their mineral wastes during the life of mine at an operating cost in the range of \$104–107/t CO<sub>2</sub> [113]. Pasquier et al. [123] investigated an integrated mineral carbonation process using southern Québec mining wastes to trap CO<sub>2</sub>, applied heat treatment to activate the waste materials, adsorbed CO<sub>2</sub> directly from flue gas, and recovered the by-products (pure carbonate). The mine could sequester 0.39 Mt CO<sub>2</sub>/year. The global process cost is estimated at \$144/t CO<sub>2</sub>, and the generated revenues is \$644/t CO<sub>2</sub>. The payback period is 1.4 years [123]. From an economic point of view, this process is highly beneficial. However, these estimations were based on electricity generated by inexpensive hydroelectric, which is not applicable for other projects that use coal or natural gas as power supplies.



**Figure 6.** Conceptual model of mineral carbonation cost versus efficiency for various methods of mineral carbonation. Corroborating data points: (a) natural alteration; (b) heap leaching; and (c) autoclave reaction. Reprinted with permission from Hitch and Dipple [122]. Copyright 2012 Elsevier Ltd.

Table 3 lists the costs of some projects using updated CCS technology. From an economic perspective, integrated mineral carbonation within the mining industry [122] has the potential to compete with geological storage methods [124]. Although the cost of mineral carbonation is reduced via integrating into the mining industry, its cost is still too high for application given current carbon prices. For example, the California cap and trade price is only approximately \$15/t CO<sub>2</sub>. Revenue that is generated from value-added products (e.g., pure carbonate) and through enhanced metal recovery during carbonation (e.g., Ni) may allow for carbonation processes to be economically viable. The pure carbonates produced through indirect mineral carbonation method are of high value and easy to use. However, the carbonate products generated from direct mineral carbonation process, which are mixtures of carbonates, amorphous silica, and unaltered silicates are hard to use. Further study on the potential application of carbonation products would add value to integrated mineral carbonation process and may lead towards economically viable technologies.

**Table 3.** The cost of selected CCS projects.

CO <sub>2</sub> Source	CO <sub>2</sub> Capture	CO <sub>2</sub> Transportation	CO <sub>2</sub> Storage	Cost (\$/t CO <sub>2</sub> Avoided)	Ref.
NGCC <sup>a</sup>	Post-combustion	Pipeline	Geological storage	59–143	[124]
SCPC <sup>b</sup>	Post-combustion	Pipeline	Geological storage	46–99	[124]
IGCC <sup>c</sup>	Pre-combustion	Pipeline	Geological storage	38–84	[124]
NGCC	Post-combustion	Pipeline	EOR <sup>d</sup>	10–112	[124]
SCPC	Post-combustion	Pipeline	EOR	(5)–58	[124]
IGCC	Pre-combustion	Pipeline	EOR	(16)–46	[124]
Oil & gas	Not given	Pipeline	Mineral carbonation	28–238	[122]
NGCC	Not applied	Pipeline	Mineral carbonation	120–159	[125]

Note: <sup>a</sup> NGCC is the natural gas combined cycle plant; <sup>b</sup> SCPC is the supercritical pulverized coal burning plant; <sup>c</sup> IGCC is the coal-based integrated gasification combined cycle system; <sup>d</sup> EOR is the geological storage with enhanced oil recovery credits.

## 5. Conclusions

Integrated mineral carbonation in the mining industry has been widely studied in recent years. Research has demonstrated that there is great potential for commercial applications. The modified passive carbonation method in tailings storage facilities are relatively inexpensive and may benefit tailings reclamation. However, conversion rates are relatively slow and not all tailings are suitable for this method. The ex-situ carbonation of mine waste mostly focuses on serpentine group minerals given their abundance. The carbonation of partially weathered olivine needs to be investigated further to develop more effective carbonation routes for various ultramafic mines. The techno-economic analysis of integrated mineral carbonation shows great potential for commercialization, especially when the carbonate products can be utilized. A cost model for adopting integrated mineral carbonation process in an area using power generated from coal or natural gas is required to advance carbonation technologies worldwide.

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