A Review on Integrated Mineral Carbonation Process in Ultramafic Mine Deposit

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Abstract: Dramatical increase in the CO₂ concentration in the atmosphere has led to the climate change, which poses a significant threat to human life on Earth. CO₂ sequestration via mineral carbonation is the one of the most effective method for mitigating global warming, and is the only way that could store CO₂ permanently. In recent years, integrating mineral carbonation via ultramafic mine deposit has received significant attention due to its high potentiality towards commercial application. This review compiles the work conducted by various researchers over the last few years on integrated mineral carbonation processes in mining industry, which use the mine waste materials as CO₂ feedstock for mineral carbonation. This paper initially introduces the basic theory of mineral carbonation, with a brief description of various techniques that enhance the rate of mineral carbonation. The enhanced mineral carbonation strategies include pre-treatment of feedstock by thermal, chemical and mechanical activation, and carbonation in a direct or indirect carbonation routes under gas/solid phase or aqueous phase. This paper then introduces the scope of application of integrated mineral carbonation. This includes the types of mine suitable for integrated mineral carbonation, the properties of mine waste materials preferable for CO₂ sequestration, and the worldwide locations potentially viable for integrated mineral carbonation. Moreover, this paper critically reviews and discusses the integrated mineral carbonation process in mining industry. The integrated mineral carbonation processes include modified passive carbonation techniques at tailing dams, 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 mineralogy, and the drawback of each integrated mineral carbonation process. All the discussions lead to the suggestions on the technology improvement in the integrated mineral carbonation process. Finally, this paper reviews the economical assessments on the existing integrated mineral carbonation process. Literature to date indicates that the value-add by-products (i.e. recovered metals, valuable carbonated products) play an important role in commercialization of an integrated mineral carbonation process.

Keywords: CO₂ sequestration, mineral carbonation, ultramafic mine, mine waste materials, tailings

1. Introduction

Over the past 30 years, the global climate has been warming at a faster pace than in any other period since 1850 (IPCC 2014). The main cause of climate change is enhances the concentration of greenhouse gases (especially CO₂) in the atmosphere via burning fossil fuels to meet the energy demand created by the global economic and population growth (U.S. Energy Information Administration 2016). The atmospheric CO₂ concentration has increased nearly 2 ppm per year since 2000, reaching 402 ppm in August 2016—43.6% above pre-industrial levels (GMD 2016). At the same time, the average global temperature shows a warming of 0.8 °C above pre-industrial levels (Ray 2015). If no additional efforts are made to constrain or manage anthropogenic GHG emissions, the CO₂ equivalent concentration is expected to exceed 1,000 ppm by 2100, which would result in a global mean surface temperature increase of 2.6 to 4.8 °C (IPCC 2014). The CO₂ emission targets of the United Nations Framework Convention on Climate Change (UNFCCC)-involved countries were outlined for the period of 2020 to

2030 in Paris Agreement (UNFCCC 2015). The ultimate goal was to limit global warming to less than 2 °C and to pursue efforts to limit the rise to 1.5 °C relative to pre-industrial levels (UNFCCC 2015). In order to reach these goals, the involved countries must find affordable and practical solutions for managing carbon.

Currently, various strategies and solutions have been adopted by various countries for the purpose of eliminating anthropogenic CO₂ emissions. These approaches include: improving energy efficiency; using energy sources that are less carbon-intensive (i.e. natural gas, hydrogen, and nuclear power) or renewable (i.e. solar, wind, hydropower, geothermal and bio-energy); enhancing biological sinks (i.e. afforestation and reforestation); and CO₂ capture and storage (CCS) (Leung et al 2014). Of these approaches, CCS is the only option that can meet the enormous demand for CO₂ reduction. It is estimated that CCS could reduce 85-90% of CO₂ from large-emission sources and energy-intensive emitters (Leung et al 2014). In the CCS process, CO₂ is concentrated from diluted sources and conveyed to storage.

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Research on and demonstration of CCS technology has grown significantly in recent years with the hope that it can one day become economically feasible (Boot-Handford et al 2014).

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 CO₂ problem and mitigate global warming. CO₂ sequestration by mineral carbonation is such a process that could permanent store CO₂. Mining industry could potentially apply mineral carbonation and contribute to the environmental protection. This paper reviews the current research status of integrating mineral carbonation in mining industry. Firstly, the developed technology of mineral carbonation is introduced. Then, the integrated mineral carbonation in mining industry is reviewed, with special focus on modified passivation method and ex-situ mineral carbonation. Finally, an economic assessment is stated.

2. Mineral Carbonation

Mineral carbonation is the only option that can permanently store CO₂ (Bobicki 2014). Mineral carbonation chemically binds CO₂ in calcium-, magnesium- and iron-rich silicate minerals to fix CO₂ as carbonate minerals (Lackner 2003). This reaction is a naturally-occurring process that slowly binds CO₂ to carbonates over several hundreds to thousands of years. Mineral carbonation has been proposed by (Seifritz 1990). Numerous researchers have attempted to accelerate the reaction between CO₂ and alkaline minerals both in-situ and ex-situ (Sanna et al 2014). The chemical reaction that takes place during carbonation occurs in two broad environmental conditions. In-situ mineral carbonation injects CO₂ into mantle peridotite and basalts deposits, and the reaction occurs in an underground, 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 (Geerlings and Zevenhoven 2013). Ex-situ mineral carbonation can also apply CO₂ as a carbon source for chemical production (i.e. carbonates), which can be used as building materials, aggregations, and cementitious phases (Markewitz et al 2012).

At present, mineral carbonation has only reached the demonstration and pilot scales (Sanna et al 2014). Although no economically and financially 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 (Doucet 2011). For example, the Carbfix pilot project in Iceland can carbonate 95% of injected CO₂ in less than 2 years (Matter et al 2016). The direct aqueous carbonation route proposed at National Energy Technology Laboratory (NETL), formerly Albany Research Council (ARC) in the U.S., (O'Connor et al 2005) has been evaluating the most promising ex-situ process for industrial application, and this process is commonly used as a base line against which other methods of carbonation are compared. The indirect two step

pH-swing carbonation method developed at Finland's Åbo Akademi University (ÅAU) has produced results that are able to compete with the NETL process when using serpentine as CO₂ feedstock (Nduagu et al 2012). In cases where geological storage is not applicable, mineral carbonation processes could be an alternative option for the storage of small to medium CO₂ emissions (< 2.5 Mt CO₂) (Sanna et al 2014). Figure 1 shows an example of the CCS process via ex-situ mineral carbonation.

Figure 1. Material fluxes and process steps associated with the mineral carbonation of silicate rocks or industrial residues. (Source: IPCC 2005)

2.1 Process routes

Mineral carbonation has focused on two broad methods: direct and indirect mineral carbonation. Direct mineral carbonation is completed in one single reaction step. Indirect carbonation introduces an additional step by extracting the reactive compound. Both direct and indirect methods have two different possible routines, namely a dry process and an aqueous process, which have different CO₂ sorption characteristics. In a dry process, CO₂ molecules are chemisorbed to the solid by strong chemical bonds or physisorbed by weaker inter-molecular bonds due to the interactions between CO₂ molecules and a sorbent (Kwon et al 2011). In aqueous sorption, CO₂ is dissolved into the solvent first, and then is reacted with olivine or serpentine (Guthrie et al 1999).

2.2 Pre-treatment

Sometimes Ca/Mg-silicates, particularly serpentine, cannot be enhanced sufficiently by the careful selection of a process route and process conditions alone. Pre-treatment options can improve reaction kinetics for some potentially attractive Ca/Mg-silicates by activating the mineral to overcoming its slow dissolution kinetics. Pre-treatment can be conducted by thermal, chemical, or mechanical means (Power et al 2013b).

Thermal treatment removes chemically-bound water, which can increase the porosity and the resulting surface area. Thermal treatment is suitable only for serpentine group minerals, which contains up to 13 wt% chemically bound water. By heating it to 600-650°C, the hydroxyl groups of serpentine are removed (Gualtieri et al 2012). Significant improves the carbonation rate of serpentine. There are some thermal activation other than heat treatment, for example,

steam treatment (O'Connor et al 2001a), microwave treatment (Bobicki 2014).

Chemical activation aims at polarizing and weakening the magnesium bonds within the magnesium silicate structure. An array of chemicals have been proposed to for chemical activation include inorganic acids (acetic, hydrochloric, phosphoric and sulfuric acid), organic acids (HCOOH, CH₃COOH, CH₃COOH), bases (sodium hydroxide, KOH), and salts (NH₄Cl, (NH₄)₂SO₄, NH₄NO₃) (Azdarpour et al 2015). It is commonly recognized in the literature that sulfuric acid is the best extraction agent, and that it can extract more than 70% magnesium from serpentine and produce a silica by-product with a high surface area (an increase in surface area from 8 m²/g to 330 m²/g). For selective leaching, ammonium salts have performed better (Wang and Maroto-Valer 2011).

Mechanical activation via high-energy attrition grinding increases the surface area of minerals, and introduces imperfections into the crystal lattice (Baláz et al 2008). Studies have focused on comparing individual mineral (i.e. olivine or serpentine) responses to the grinding energy input, milling instruments (i.e. planetary mills, vibration mills, attrition mills, or tumbling mills), milling atmosphere (i.e. N₂, CO₂ or air), milling solutions (i.e. water, alcohol or acid), and milling temperatures (Li and Hitch 2017b).

The combination of two pre-treatment methods, such thermal-chemical activation (O'Connor et al 2001b), chemical-mechanical activation (Uddin et al 2012), thermal-mechanical activation (McKelvy et al 2005) can achieve unexpected activation effects. Some novel process for enhance the rate of mineral carbonation has proposed for waste materials, include co-utilization with wastewater (or brine solution), biological enhancement, and reactor integration (Figure 2). Although these novel treatment methods couldn't activate materials up to the level that the former three techniques could reach, they are promising for the industrialization application as waste materials are applied and industrial ready instruments are used, which reduce the carbonation cost dramatically (Pan et al 2015).

Figure 2. Different approaches to process enhancement of ex-situ direct carbonation for alkaline solid wastes (Pan et al 2015)

3. Integrated Mineral Carbonation in Mining

As a carbon-intensive industry, mining is facing ever-increasing pressure to reduce CO₂ emissions as a result of emerging carbon management policies (Ballantyne 2010).

Integrated mineral carbonation processes in the mining industry, which is using mine wastes (i.e. waste rock, tailings, slag) as CO₂ feedstock, is advantageous for its ability to manage CO₂ emissions and remediate potentially hazardous materials (i.e. chrysotile) in mine wastes (McCutcheon et al 2014). By using mine wastes, especially tailings, many of the costs associated with the comminution required for mineral carbonation are absorbed as part of the traditional process of metallic mineral extraction. This has a large impact on the costs associated with altering tailings to a carbonate form. With a mineral carbonation process added in, some marginal projects, such as low-grade nickel deposits, may potentially lower their cut-off grades in order to become economically feasible (Hitch and Dipple 2012).

3.1 Suitable mine for mineral carbonation

The ultramafic-hosted ore deposits that are favored in the integrated mineral carbonation process include chrysotile mines, nickel mines, chromium mines, and platinum group element (PGE) mines (Hitch et al 2010). Figure 3 shows the potential large and super-large ultramafic mine sites that are suitable for the integrated mineral carbonation process worldwide (Picot et al 2011). Notably, the annual amount of mining tailings produced globally could potentially offset approximately 1.5% of annual global CO₂ emissions (Power et al 2013b). The majority mineral for sequestration CO₂ in the ultramafic mine waste materials are olivine, serpentine and brucite.

Figure 3. Ultramafic rock-hosted ore deposits lying less than 300km from a CO₂-emission site (Bodénan et al 2014).

3.2 Modified passivation method

The carbonation of ultramafic mine waste initially uses a passive technique at tailing dams from both closed and active mines (Wilson 2005). Instead of human input, passive carbonation exposes tailings to the Earth's atmosphere, hydrosphere, and biosphere, allowing for natural weathering and gradual carbonation. The passive carbonation rate of tailings can be enhanced through simple modification, such as optimizing the surface area of tailings (Wilson 2005) and/or adding carbonic anhydrase to catalyze the hydration of aqueous CO₂ and to promote carbonate precipitation (Power et al 2013a). Furthermore, seeding acid-generating materials with *Acidithiobacillus* spp. enhances magnesium ion leaching from silicate minerals (Power et al 2010); applying a cyanobacteria accelerates the precipitation of platy hydromagnesite (McCutcheon et al 2014); using carbonic anhydrase catalyzes the hydration of aqueous CO₂ (Power et

al 2016); pre-seeding carbonates promotes the carbonation nucleation (Zarandi et al 2016); pumping CO₂ into tailing water increases CO₂ gas content (Harrison 2014); and periodically adding small amounts of water keeps partial pore saturation at optimum levels (Assima et al 2013) which promotes the conversion of CO₂ gas into a carbonate form. Moreover, the by-products of carbonation, such as photoautotroph biomass, can be harvested as biofuel (Power et al 2011). Since the carbonation is under ambient temperature and pressure conditions, the energy consumption of modified passive carbonation is low. In addition, not all tailings are suitable for this method; for example, tailings that are extremely carbonated (Power et al 2010), and massive, serpentine-enriched mine residues (Assima et al 2014). Figure 4 shows two example of modified passivation method applied in mine tailings.

Figure 4. A schematic of two mine tailings facilities depicting (A) an abiotic strategy employing CO₂ injection, and (B) use of bioleaching and microbial carbonate precipitation as strategies for carbon mineralization (Power et al 2013b).

3.3 Ex-situ mineral carbonation

In an active mining operation, it is possible to apply the ex-situ carbonation to fresh tailings. Fresh tailings are more active for carbonation than weathered ones due to minimal authigenic carbonate content (Power et al 2010). These advanced technologies have been adopted to estimate the suitability of mine waste materials as CO₂ feedstock (Table 1). Current investigations into integrated mineral carbonation processes mostly focus on mine waste that contains serpentine and brucite (Bobicki 2014, Harrison 2013). The most common methods for mine waste pre-treatment are thermal activation using calcination (Pasquier et al 2014), microwave heating (Bobicki 2014), steam mediation (Larachi et al 2012), and chemical activation using organic or inorganic acid (Bobicki 2014). These pre-treatment methods dehydrate serpentine and/or disorder its crystal structure, which greatly accelerates 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 (Declercq et al 2013). The authors has studies on mine waste materials consisting of partially-weathered olivine and mechanical activation were used to enhance the reaction rate of the mine waste materials (Li and Hitch 2017a). As partially-weathered olivine is the most abundant form of ultramafic rock in nature (Styles et al 2012). It is of great significance and provide the potential for ex-situ mineral carbonation to become widely employed in

the mining industry. However, the mineralogy of mine waste materials various from mine to mine, for example, olivine with a different degree of serpentinization, or peridotites (i.e. forsterite, and enstatite) with different types of weathering (i.e. serpentinization, carbonation). Further study on the carbonation of these naturally occurring rocks and find out the mechanism of the carbonation would promote the application of integrated mineral carbonation method.

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

Majority minerals	Pre-treat	Carbonation method	References
Anorthosite	NA	DC gas-solid DC aqueous	(Ghacham et al 2015)
Serpentine	TA, CA	DC aqueous	(Bobicki 2014)
Serpentine	TA	DC aqueous	(Pasquier et al 2014)
Serpentine	TA, MS	DC gas-solid	(Veetil et al 2014)
Enstatite, plagioclase feldspar	CA	IDC aqueous	(Meyer et al 2014)
Olivine, serpentine	TA, CA, MA	DC aqueous, IDC aqueous	(Bodénan et al 2014)
Chrysotile, lizardite,	NA	DC gas-solid	(Sarvaramini et al 2014)
Chrysotile	TA	DC gas-solid	(Larachi et al 2010)
Serpentine	CA	IDC aqueous	(Teir et al 2009)
Olivine, serpentine	NA	DC aqueous	(Jacobs 2014)
Olivine, serpentine	MA	DC aqueous, DC gas-solid	(Li and Hitch 2016a, 2016b)

Note: HA 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.

4. Economical Assessments

The economic analysis on the integrated mineral carbonation process in mining industry was first studied by (Hitch and Dipple 2012). Figure 5 shows the model shows the tradeoff of carbonation cost versus carbonation efficiency. In a case study of Turnagain Nickel Deposit located at northern BC, the theoretical carbonation potential was estimated to be 81.9 Mt CO₂ over a 29-year mine life (Hitch et al 2010). The integrating mineral carbonation process yielded a NPV of \$131.5 million at a discount rate of 8%, with an operating cost of 82.5 \$/t CO₂ (Hitch and Dipple 2012). With the mechanical activation as pre-treatment method in the integrated mineral carbonation process the carbonation efficiency reach 60%, the mine can potentially sequester 14.62 Mt/y CO₂ using mine waste rock and tailings during

the life of mine, and the operating cost ranged from 104 to 107 \$/t CO₂ (Li and Hitch 2016c). Pasquier et al (2016) has investigate an integrated mineral carbonation process using southern Québec mining wastes as CO₂ feedstock with heat treatment, adsorb CO₂ directly from flue gas, and recover the by-products (pure carbonate). The mine could sequester 0.39 Mt/y. The global process cost is estimated at 144 \$/tCO₂, and the generated revenues is 644 \$/tCO₂. The payback period is 1.4 years. From an economic point of view, the process is highly beneficial (Pasquier et al 2016). However, these estimations were based on hydro power plant, which on longer not applicable in other areas using coal or natural gas as power supply, which is the most applying energy in the world.

Figure 5. Conceptual model of mineral carbonation cost versus efficiency for various methods of mineral carbonation. Corroborating data point (a) is nature alteration, (b) is heap leaching, (c) is autoclave reaction (Hitch and Dipple 2012).

Although the cost of mineral carbonation is reduced via integrating into mining industry, its cost is still too high to apply. The revenue affiliated with the carbonated product is the key to overcome the obstacle. The pure carbonates produced through indirect mineral carbonation method is 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. Continuous study on the potential application of carbonate products would add value to integrated mineral carbonation process and made it economically feasible.

5. Conclusions

Integrated mineral carbonation in mining industry has been studied widely in recent years. They show great potential to go towards commercial application. The modified passivation method in tailings pond is cheap. However, the reaction rate is slow and not all tailings suitable for this method. The ex-situ carbonation of mine waste mostly focuses on the serpentine minerals. The carbonation of partially weathered olivine needs to be investigated further to find out the more proper carbonation routines. The economic analysis of integrated carbonation shows great potential of application, especially when the carbonated product could be

used properly. The cost model for adopting integrated mineral carbonation process in the area using coal or natural gas generated power is required to build, to estimate if the technology could apply globally.

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