A Review of Mineral Carbonation from Industrial Waste

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Abstract. For relieving CO₂ emissions and climate change, carbon sequestration and storage has been paid more attention. Mineral carbonation technology is a process that the calcium and/or magnesium-containing minerals react with CO₂ to form stable carbonate precipitation, which in turn fixes CO₂. It originates from natural rock weathering process that calcium and magnesium-containing rocks react with CO₂ dissolved in rain water to form carbonate minerals. The source of mineral carbonation is the solid containing calcium and magnesium compound. The classification of mineral carbonation including direct and indirect mineral carbonation is indicated, and the descriptions of extraction by different extract agent have been given. The pretreatments, operation conditions and reaction kinetics are also discussed. Furthermore, the different processes by carbonizing from industrial waste are reviewed and the energy loss and cost are also described.

1. Introduction
With the acceleration and enlargement of people’s life, the dependence on energy consumption and the influences on world environment become more and more significant. As the dominant fuel, due to high energy density, easy to transport and store, hydrocarbon fossil fuel has been used widely. The combustion of fossil fuel results in a large quantity of flue gas releasing into atmosphere which have a negative impact on the environment, such as global warming and climate change caused by greenhouse gases especially CO₂. Based on CO₂ and other Greenhouse Gas Emissions Data [1], the emission of CO₂ increases from 22.36 billion tonnes in 1990 to 36.18 billion tonnes in 2015, whose rate of increase is about 62%. In addition, the concentration of atmosphere CO₂ also grows from 354 ppm in 1990 to 403 ppm in 2016 [2, 3]. The growth of CO₂ level significantly affects the global temperature which has already been increased about 0.8 °C.

Carbon sequestration and storage (CCS) is a technology to sequestrate and store CO₂ emitted from power plants and other factories that use fossil fuels. It generally includes three steps: (i) separate and capture CO₂ from waste stream; (ii) transport CO₂ to appropriate location to store; (iii) reserve CO₂ by different methods. Because the dominant source of CO₂ is the combustion of fossil fuel, particularly the power plants, the conventional capture consists of pre-combustion capture, post-combustion and oxyfuel combustion. In the pre-combustion process, the fuel is usually pretreated before combustion. When pre-combustion capture is used in Integrated Gasification Combined Cycle (IGCC) power plants, there will be an efficiency loss of 7-8%[4, 5]. Post-combustion technology refers to remove
CO₂ after combustion. It is found that the electricity cost will be increased by 32% and 65% with post-combustion technology in gas and coal-fired plants, respectively [6]. While for oxyfuel combustion technology, air is replaced by oxygen to apply in combustion. The substitution of air with oxygen leads to high energy consumption, accompanied with high SO₂ concentration resulting corrosion [7]. The transportation of CO₂ can be achieved by pipes [8], boats or tanker trucks [9], which is mainly for industrial application or to increase the output of natural gas fields and oil fields. There are many ways to store or sequester CO₂, such as geologic storage, ocean storage, industrial use and mineral carbonation. Geologic storage is injecting CO₂ into a geological storage site, such as depleted oil and gas reservoirs, unmineable coal beds, and saline aquifers, which usually can store large quantities of CO₂ like several tens of millions tonnes [10]. The selection of geological sites is essential and the requirements of geological sites should possess appropriate porosity, thickness, and permeability of the reservoir rock, a cap rock with good sealing capability, and a stable geological environment [11]. It is estimated that CO₂ stored in deep oceans can be kept for several hundred years [12]. A research indicated that the storage of CO₂ injected into deeper than 3 km deep sea sediments can stand against large geomechanical perturbations [13]. CO₂ can also be consumed in industry producing ammonia and urea, food beverages, refrigerants and fire extinguishing gases. Mineral carbonation is a process to make CO₂ react with mineral contained Mg/Ca to form magnesia/calcium carbonate [14].

For different methods of carbon storage, there are various advantages and disadvantages. The advantages for geologic sequestration are feasible on a large scale, substantial storage capacity known and extensive experience, however it needs to be monitored and there is leakage possible. Although the cost of ocean carbon sequestration is low, it has potential harmful effects on aquatic microbes and biota. When CO₂ is used in industry, it is limited by storage capacity. Because mineral carbon sequestration has many benefits, such as permanent storage, environmentally benign, and reuse of fossil fuel carbon, it has been extensively investigated.

In this review, two kinds of mineral carbonation including in situ and ex situ mineral carbonation are elaborated. The theories of direct and indirect mineral carbonation are listed. Owing to different extraction agent, the indirect mineral carbonation can be accomplished by acid extraction, molten salt extraction, ammonia extraction and bioleaching, which are explained in details.

2. Mineral carbonation
Carbon sequestration by mineral carbonation is a process that CO₂ can be fixed as stable carbonate mineral, which stems from natural rock weathering through the dissolution of CO₂ in rain water reacting with alkaline mineral. In 1990, it is first proposed by Seifritz to sequester CO₂ as an alternative storage strategy [14]. There are two ways to implement mineral carbonation: in situ mineral sequestration and ex situ mineral sequestration [15]. In situ mineral carbonation is to inject CO₂ dissolved in water into unconventional reservoirs such as mafic and ultramafic rocks, which can react with CO₂ to form carbonate mineral such as calcite, magnesite, dolomite and siderite [16-20]. Although in situ mineral carbonation is stable, the requirements of reactivity, porosity and permeability hinder its development. Ex situ involves the aboveground carbonation in a traditional chemical process. It differs from in situ in the reaction taking place aboveground, which makes the operation feasible.

Ex situ mineral carbonation can be accomplished by direct (gas- and aqueous-based) and indirect processes. The basic principles of direct and indirect processes are shown in Figure 1. In direct process, CO₂ react with mineral to form carbonate in one step. However, the alkaline earth metals oxides, such as CaO and MgO, usually are acquired by extraction before carbonation in indirect process.
2.1. Direct mineral carbonation

For direct mineral carbonation, the alkaline minerals react with CO$_2$ can be either in gaseous or aqueous phase. The typical reaction occurs in gas-solid mineral carbonation can be expressed as reaction (1) to (5) [22]. These reaction needs high temperature and pressure, which prevent its application [23, 24]. In addition, both slow reactive rate [25] and lack of storage of calcium-and magnesium-containing oxides or hydroxides in nature limit the improvement of gas-solid mineral carbonation.

$$
\text{CaO} + \text{CO}_2 \rightarrow \text{CaCO}_3 \quad (1)
$$

$$
\text{MgO} + \text{CO}_2 \rightarrow \text{MgCO}_3 \quad (2)
$$

$$
\text{CaSiO}_3 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{SiO}_2 \quad (3)
$$

$$
\text{Mg}_2\text{SiO}_4 + 2\text{CO}_2 \rightarrow 2\text{MgCO}_3 + 2\text{SiO}_2 \quad (4)
$$

$$
\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 3\text{CO}_2 \rightarrow 3\text{MgCO}_3 + 2\text{SiO}_2 + 2\text{H}_2\text{O} \quad (5)
$$

When CO$_2$ an alkaline mineral generate precipitation in an aqueous phase directly, it can be considered as aqueous mineral carbonation. Generally, there are three reactions in the process (reaction 6, 7, 8). At first, CO$_2$ dissolves in the water to form bicarbonate species which react with mineral. Then the liberated proton from reaction 6 decomposes the calcium-and magnesium-containing minerals resulting in metal ion (reaction 7), which will be carbonated as precipitation (reaction 8). In order to enhance the reactivity of mineral carbonation, many methods have been conducted such as increasing temperature and pressure, reducing particle size, and adjusting PH value.

$$
\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{CO}_3 \rightarrow \text{H}^+ + \text{HCO}_3^- \quad (6)
$$

$$
(\text{Ca/Mg})-\text{silicates} + 2\text{H}^+ \rightarrow (\text{Ca/Mg})^{2+} + \text{SiO}_2 + \text{H}_2\text{O} \quad (7)
$$

$$
(\text{Ca/Mg})^{2+} + \text{HCO}_3^- \rightarrow (\text{Ca/Mg})\text{CO}_3 + \text{H}^+ \quad (8)
$$

2.2. Indirect mineral carbonation

Indirect mineral carbonation is different from direct mineral carbonation in more steps. The first step is to extract reactive component from feedstock, such as Ca and Mg, then they will react with CO$_2$ to form precipitation. Because the steps of achieving reactive component and carbonation are separated, the obtained carbonates are more pure, and the impurities can be removed before carbonation.

Figure 1. Basic principles of the direct and indirect carbonation methods [21]
2.2.1. Acid leaching. Many acids can be used to extract the reactive component from feedstock mineral, such as HCl, HNO3 and acetic acid. The typical HCl extraction process is proposed to extract Mg from serpentine, which can be divided into three steps as shown in reaction (9-11). The mineral serpentine dissolves in HCl solution to form MgCl2·2H2O, then it changes to Mg(OH)2 followed by carbonation to MgCO3. Due to its shortcomings of corrosion [26] and high energy cost [27], HCl extraction process has not been attracted interest. Teir et al. [28, 29] used natural serpentinite as feedstock and H2SO4, HCl, and HNO3 as extraction agent to extract magnesium salt at temperatures of 30, 50 and 70 °C. They found that all magnesium was extracted from serpentinite in each of the three acid solutions tested during 1–2 h, associated with extraction of most iron in serpentinite and little silicon (<4%). At the beginning of the dissolution, the chemical reaction is the major limitation. After the product layer of silica formed, the product layer diffusion becomes the control steps of dissolution. Petr et al. [30] dissolved wollastonite (CaSiO3) in the aqueous solution of acetic acid (3 mol/L) at the temperature interval from 25 to 50 °C using mixed batch-type reactor. Acetic and succinic acid [31] are used to leach calcium from wollastonite, and succinic acid performed better extraction efficiency than acetic acid.

\[
\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 6\text{HCl} + \text{H}_2\text{O} \rightarrow 3\text{MgCl}_2 \cdot 6\text{H}_2\text{O} + \text{SiO}_2
\]  
(9) 
\[
2\text{MgCl}_2(\text{OH}) \rightarrow \text{Mg(OH)}_2 + \text{MgCl}_2
\]  
(10) 
\[
\text{Mg(OH)}_2 + \text{CO}_2 \rightarrow \text{MgCO}_3 + \text{H}_2\text{O}
\]  
(11) 

Due to high cost, acid extraction of mineral contained calcium and magnesium does not apply into practical application. For the process of extraction [28], the pH value has to increase to 8.5 allowed carbonation step, which leads to high reagent costs ($600-1600/t \text{CO}_2$ for NaOH and make-up acid). The high cost limit indirect mineral carbon sequestration with acid extraction to compete with other \text{CO}_2 storage techniques.

2.2.2. Molten salt process. In order to reduce the high expense of acid extraction, molten salt extraction process was proposed. In this process, MgCl2 is used as extraction agent to extract magnesium from serpentine, and the reaction is as follows(reaction 12-15). After the molten salt MgCl2·5H2O is added into the reactor, Mg(OH)Cl is formed. Then the precipitation Mg(OH)2 is produced after water added and the generated MgCl2 can be recycled in the process. At the last step, Mg(OH)2 is separated and carbonated.

\[
\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 3\text{MgCl}_2 \cdot 3.5\text{H}_2\text{O} \rightarrow 6\text{Mg(OH)}_2\text{Cl} + 2\text{SiO}_2 + 2.5\text{H}_2\text{O}
\]  
(12) 
\[
2\text{MgCl}_2(\text{OH}) + \text{nH}_2\text{O} \rightarrow \text{MgCl}_2 \cdot \text{nH}_2\text{O} + \text{Mg(OH)}_2
\]  
(13) 
\[
\text{MgCl}_2 \cdot \text{nH}_2\text{O} \rightarrow \text{MgCl}_2 \cdot 3.5\text{H}_2\text{O} + (\text{n} \cdot 3.5)\text{H}_2\text{O}
\]  
(14) 
\[
\text{Mg(OH)}_2 + \text{CO}_2 \rightarrow \text{MgCO}_3 + \text{H}_2\text{O}
\]  
(15)

2.2.3. Ammonia extraction. It is found that ammonia salts can be used to extract metals from silicate rocks. It is proposed by Pundsack [32] in 1967, and the reactions are shown in reaction 16-18. It can be seen that Mg2+ is first leached by the addition of ammonia salts NH4HSO4 followed by carbonation to magnesium carbonate, and NH4HSO4 can be regenerated by thermal decomposition of the (NH4)2SO4. In recent years, Wang et al. [33] developed pH-swing mineral carbonation process by using ammonium salts as extraction agent.

\[
\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 6\text{NH}_4\text{HSO}_4 \rightarrow 3\text{MgSO}_4 + 2\text{SiO}_2 + 3(\text{NH}_4)_2\text{SO}_4 + 5\text{H}_2\text{O}
\]  
(16)
2.2.4. Bioleaching. Applying bacteria to extract Mg/Ca from minerals can be considered as bioleaching. Power et al. extracted metal from ultramafic tailing using combined acid generating substances (AGS) with chemolithotrophic bacteria: Acidithiobacillus (A.) ferrooxidans and Acidithiobacillus (A.) thiooxidans [34]. During this process, there is a cycle with AGS gives food to bacteria, the bacteria produce sulphuric acid, and the sulphuric acid extract Mg/Ca from minerals. At the last step, Mg/Ca ions react with CO$_2$ to form carbonate which fixed CO$_2$.

3. Mineral carbonation from industrial waste

3.1. Steelmaking Slags

During the steelmaking process, not only produces a large amount of steelmaking slags, about 10-15% of crude steel, but also emits CO$_2$ with the quantity of 0.28-1.0 t per ton steel, taking up 6-7% of total CO$_2$ emission [35, 36]. Steelmaking slags are a kind of byproduct from steelmaking process and they are usually classified into the following categories: blast furnace slag (BFS), basic oxygen furnace (BOF), electric arc furnace (EAF), and ladle furnace (LF) slags. The composition of steelmaking slags fluctuates greatly based on different steelmaking process. Although the composition of steelmaking slags is flexible, the steelmaking slags are abundant in calcium oxide and magnesium oxide, which makes steelmaking slags a good candidate for mineral carbon sequestration.

According to the composition of steelmaking slags, many researches are investigated on mineral carbonation of steelmaking slags with various different methods. Chang et al. [37] performed the CO$_2$ sequestration experiments using direct mineral carbonation in an autoclave reactor, and the temperature, reaction time, pressure and initial pH were studied. It is found that higher temperature and higher pressure is beneficial to reaction kinetics. Poletti et al. [38] applied direct mineral carbonation to investigate the effect of parameters. It indicated that the particle size plays an important role in carbonation kinetics and smaller particles are more beneficial for carbonation. The extraction of Mg/Ca from steelmaking slags by acid has been investigated by many researchers. The concentration of acetic acid has a dramatic effect on the extraction efficiency [28,29]. With 10 wt% acetic acid, most of calcium is extracted, while increasing the concentration of acetic acid, there is not growth on the extraction efficiency. Some literatures are focused on extraction by ammonia salts. A process including swinging the pH of the solution, and extracting alkaline-earth metal was proposed to pretreat the mineral by NH$_4$Cl. The product layers deposited on the particles prevent the diffusion of extractant into slag particles, which also limit the carbonation sequestration [39].

3.2. Cement wastes

Cement waste is generated in the process of construction and demolition waste recycling. It is usually applied as binder in the production of concrete. The composition of cement waste consists of calcium, silica, alumina, and iron oxide. Millions of tons of cement kiln dust are also produced during in the cement industry. Both of cement waste and cement kiln dust are rich in CaO which is the main reactive agent with CO$_2$ in mineral carbon sequestration. To increase the reactivity, the cement waste is usually grounded to small particles before carbonation. Due to the properties of cement waste and cement kiln dust, both of them are good candidate for carbon sequestration. In addition, carbonation production can be recycled in cement manufacturing or other industry processes [40, 41].

Various researches has focused on the investigation of mineral carbonation by cement waste and cement kiln dust. Gunning et al. [42] studied mineral carbonation using cement kiln dust with ambient pressure and temperature. The operating conditions including water content and CO$_2$ concentration are
evaluated. The extents of carbonation are greater than 70% of its theoretical capacity, and it can be accomplished without any amendments or modifications to the waste material. Alicja et al. studied the effect of the amount of water on the values of absorbed CO$_2$, and it showed that carbon dioxide absorption decrease with increase of water content [43]. Xuan et al. [44] applied direct mineral carbonation to study CO$_2$ sequestration by cement waste. It revealed that over 75% of the total experimental CO$_2$ uptake is fixed by cement waste. After mineral carbonation with 144 h, dry cement waste and fine recycled concrete aggregates can capture 110 g CO$_2$/kg raw material and 52 g CO$_2$/kg raw material, respectively.

3.3. Mining Waste

During mineral process, there are a large amount mining waste and mineral processing wastes generated. By analysis of the waste, it can be found that the composition of mining waste and mineral processing waste includes metal such as magnesium and calcium, which are dominant metal to carbonize CO$_2$. Among many kinds of mining waste, the asbestos tailings, red mud and nickel tailings are good candidate for mineral carbonation, because all of them comprise ultramafic rock, such as the asbestos tailings including dunite, serpentine, and gabbronorite, nickel tailings including serpentine and red mud including CaO. Carbon sequestration using them as source not only relieves CO$_2$ emission, but also decreases the risk of stacking waste contaminating the environment. This becomes an effective way to carbonation carbon by external CO$_2$ stream.

For different mining waste and mineral processing waste, there are various different methods to carbonation CO$_2$ due to their properties. Asbestos tailings are often classified as hazardous, as inhaled asbestos fibers aggravate lung tissues, which cause them to scar. Carbonation using asbestos tailings becomes benefits for our environment. The asbestos tailing from Clinton Creek has been investigated by bioleaching metal using Acidithiobacillus spp [34]. The leaching efficiency has been enhanced by combining acid-generating substance with bioleaching, and the magnesium concentration has also increased by at least order of magnitude. It is also found that high temperature and pressure is helpful for carbonation of asbestos tailing. Nickel tailings are usually abundant in ultramafic which can be extracted to carbonize. The extraction of nickel tailing by different extractant like acids, bases and ammonium salt has been studied. A high carbonate conversion of 94% has been accomplished and quite pure hydromagnesite has been generated. However, the cost of the process is extremely high and this limits its development [28, 29]. Red mud is a byproduct from the Bayer process to produce alumina from bauxite, and its compositions usually are Fe$_2$O$_3$(30-60%), Al$_2$O$_3$(10-20%), SiO$_2$(3-50%), Na$_2$O(2-10%), CaO(2-8%) and TiO$_2$(trace-10%) [45]. Some carbonations of red mud with direct mineral carbonation have been performed at low pressures and ambient temperature. Yadav et al. [46] showed that the main mineral phases responsible for carbonation are chalcanthite and cancrinite, and the medium density (1.8 g/cm$^3$) of red mud material was found more effective to capture CO$_2$.

3.4. Waste Ash

Waste ashes come from industrial combustion processes, and they consist of CaO which is ideal for mineral carbonation. Generally, the waste ashes are presented as small particle size distribution which does not need to grind. Direct gas-solid and aqueous processes are employed to carbonize CO$_2$. The carbon sequestration capacity from waste ash is relatively low, but the operation conditions have been explored by many researchers. The carbonation can be achieved at ambient temperature and low CO$_2$ pressure using municipal solid waste incinerator ash [47]. By using coal fly ash as source, a carbonate conversion of 82% can be obtained with 30°C and 10 bar CO$_2$ pressure [48]. Compared with municipal solid waste incinerator ash and coal fly ash, oil shale ash exhibited better CO$_2$ sequestration capacity. Uibu et al. [49] conducted the experiment of carbonation oil shale ash at ambient temperature and pressure in a continuous flow reactor, and they found that the captured CO$_2$ from flue gases are 1-1.2 million ton at the 2007 production level of the SC Narva Power Plants.
4. Conclusion
Mineral carbonation is an effective method to fix CO₂ as stable carbonate, and mineral carbonation from industrial waste has been reviewed in this paper. Direct and indirect mineral carbonation are described and different extraction processes are elaborated. The advantages of steelmaking slags lie in high CO₂ capacity, but its drawback is that they have to be grinded into small particles. Due to a large amount of CaO contained in cement waste, it is easy to operate. Mineral carbonation from mining waste and mineral processing wastes, including asbestos tailings, nickel tailings and red mud, not only remedy the harm to our environment, but also capture and store CO₂. The carbon sequestration capacity from waste ash is relatively low without pretreatment which results in low cost. Mineral carbonation from industrial waste is a promising way to reduce CO₂, and the development of mineral carbonation should be focused on reducing cost and increasing CO₂ capacity in future.

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