

CO₂ sequestration by mineral carbonation: a review

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Abstract

The mineral sequestration, one of the methods of CO₂ sequestration, is considered advantageous, as it not only facilitates permanent and leakage free storage of CO₂ but also obviates the need for regular monitoring. Mineral sequestration involves the dissolution of minerals and subsequently carbonation of dissolved minerals. In the direct mineral sequestration all the processes occur within a single reactor, whereas in the cases of indirect mineral sequestration, they take place in separate reactors. The main aim of the present study is to investigate the efficacy of these mineral sequestration methods and examine their suitability for industrial application and ensuring environmental friendliness. For this purpose, literature pertaining to these methods was extensively reviewed, and observations made by several researchers were collected, collated and compared based on the various parameters such as reaction pathways, reaction kinetics and cost of the process. The process cost was found to depend on the type of the process, process parameters, input materials and additives. It was noted that the direct mineral sequestration suffers from the sluggish reaction kinetics, thereby becomes economically unviable. The success of direct mineral sequestration process is yet to be achieved despite research carried out for several years. The problem of sluggish reaction kinetics was overcome by using multi-steps indirect carbonation routes, where separate reactors were used for dissolution and precipitation processes. The indirect sequestration method was noted to be most efficient as it offered several advantages such as improved reaction kinetics and recovery of the market value of by-product due to the better quality control of the product. Hence, based on interpretation of an extensive review of literature it can be concluded that the indirect mineral sequestration may be a viable option to carry out the CO₂ sequestration and may be proved as a guiding light to ensure the clean environment for future generation.

Keywords: CO₂ sequestration, carbonation routes, mineral carbonation, mineral sequestration, pre-treatment of feedstock

1. Introduction

CO₂ has been rapidly increasing in the atmosphere in recent years. As a result of this increase in atmospheric CO₂ concentration, there is a rise in the earth's temperature. CO₂ is considered the most influential greenhouse gas, due to its huge volume and rapid growth rate (Brierley and Kingsford, 2009). Reducing anthropogenic CO₂ emissions into the atmosphere helps mitigate global warming and climate change (IPCC, 2005; Shashikant Yadav and Mehra, 2017a). The reduction can be done through geological storage, ocean storage, biological storage and mineral storage (Krekel *et al.*, 2018; Lubrano Lavadera *et al.*, 2018; Muthuramalingam *et al.*, 2018; Sipilä *et al.*, 2008a).

The most prevalent CO₂ sequestration method in practice is the geological storage, where CO₂ is stored in underground geological cavities such as depleted gas and oil reservoirs, coal seams, salt caverns and saline aquifers (Park, 2005). The CO₂ is separated from a flue gas offshore emitting source and is then transported to a storage site and finally injected at high pressure into an underground reservoir (Voormeij and Simandl, 2004). The problem with geological storage though is the risk of leakage, thus requiring constant monitoring throughout the year (Doria, 2005a). Secondly, we have ocean storage, which involves injecting CO₂ into the ocean at great depths of around 1500 m (Voormeij and Simandl, 2004). The gaseous CO₂ reacts with ocean water to form carbonic acid (Gunter *et al.*, 1997). The challenge with this method of storage is the lack of permanency and decreasing pH which can cause environmental hazards which adversely impacts marine life (IPCC, 2005). Another form of ocean storage is storage below the seabed, where CO₂ is pumped into the sea at deeper depths than in usual ocean storage method at about 3000 m (Brewer *et al.*, 2000). At such depths, CO₂ becomes denser than water offering greater permanency of storage and eliminates monitoring requirements (Voormeij and Simandl, 2004). Lastly, mineral sequestration is a method which involves the conversion of CO₂ into stable mineral carbonates (Sanna *et al.*, 2014a). CO₂ is injected into geological formations with alkaline minerals it forms stable carbonates and is stored.

Mineral sequestration can also be performed *ex-situ* that does not rely on large underground reservoirs or oceans (Ababou *et al.*, 2017; Aminu *et al.*, 2017; Kaliyavaradhan and Ling, 2017; Shashikant Yadav and Mehra, 2017a). Another advantage of mineral sequestration is that the reaction of CO₂ with minerals can occur without any external energy input (Shashikant Yadav and Mehra, 2017b).

2. Mineral sequestration

There are two methods of mineral sequestration, either direct or indirect carbonation (Saran *et al.*, 2017a). Direct carbonation has two phases the gas phase, and the aqueous phase. The gas phase is a single step gas phase where CO₂ reacts with minerals such as rocks *in-situ* and *ex-situ* to give carbonates (Olajire, 2013). Aqueous phase has simple carbonation, and additives enhanced carbonation (De Windt *et al.*, 2011; Doria, 2005b). In additive enhanced carbonation complex agents and acids are reacted with CO₂ to give carbonates (Shashikant Yadav and Mehra, 2017b). In indirect carbonation, the dissolution of reactive mineral ions from the feedstock and the carbonation of dissolved mineral ions take place separately in two different reactors (Bacocchi *et al.*, 2010; Teir *et al.*, 2007a).

Table 1. Feedstock for mineral carbonation (Saran *et al.*, 2017a)

Feedstock	Examples
Natural minerals	Wollastonite, olivine, serpentine, feldspar, basalt, brucite, pyroxene, forsterite, dunite
Industrial wastes	Steel slag, cement waste, fly ash, MSWI ash, paper mill waste, oil shale ash, mine tailing

3. Process routes

3.1. Direct carbonation

Direct carbonation is the most straightforward method to mineral sequestration, and the principal approach is that an appropriate feedstock such as mineral sources or a solid residue (Table 1) rich in calcium (Ca) or magnesium (Mg) is carbonated in a single step process (Huijgen and Comans, 2005). Both the extraction of minerals and carbonation of dissolved minerals occur in the single reactor.

3.2. Direct gas–solid carbonation

Gas–solid mineral carbonation is a more straightforward method for mineral sequestration than direct aqueous carbonation. In this method, mineral oxides interact with gaseous CO₂ at a particular pressure and temperature (Lackner *et al.*, 1997; O'Connor *et al.*, 2005). The dry carbonation method also produces high-temperature steam which can be utilized to produce electricity (Mouedhen *et al.*, 2017; Sanna *et al.*, 2014a). Mining activities can be integrated with carbonation process which may be beneficial to control costs and energy requirements, probably allowing for higher minerals extraction rates and purity. The disadvantage is that direct gas–solid carbonation has slow reaction rates due to thermodynamic limits and hence further studies on direct gas–solid carbonation has mostly been abandoned (Saran *et al.*, 2017b).

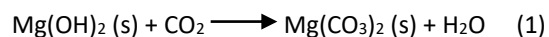
3.3. Direct aqueous carbonation

It is the most effective CO₂ sequestration method up to date since high carbonation degrees have been attained. Still, the process is expensive for large-scale CO₂ sequestration (IPCC, 2005; Sipilä *et al.*, 2008b). Direct aqueous mineral carbonation is usually done *ex-situ*. However, direct aqueous carbonation can also be done *in-situ* which involves rock samples reacting with CO₂. By controlling the input gas composition, porosity loss is minimised, and permeability maximised thereby achieving greater carbonation efficiency (Voormeij and Simandl, 2004).

A study by Koljonen *et al.* (2004) in direct aqueous carbonation of minerals has shown that using a high-pressure superficial CO₂ for carbonation of calcium hydroxide (Ca(OH)₂) at a temperature of 318 K could achieve only a low degree of agglomeration (Koljonen *et al.*, 2004). Direct aqueous carbonation of mineral rocks and solid waste materials also occur through a natural process called weathering where piles of waste ashes are exposed to atmospheric CO₂ (Sipilä *et al.*, 2008b). Generally, industrial by-products, wastes and residues react more promptly than natural minerals (Huijgen *et al.*, 2005; Kaliyavaradhan and Ling, 2017). The characteristics and compositions of the residues are majorly affected by the changes in process conditions such as temperature and pressure (Teir *et al.*, 2006). The carbonation efficiency in direct aqueous carbonation can be enhanced by using additives such as sodium chloride (NaCl) and sodium carbonate (NaHCO₃) (O'Connor *et al.*, 2005).

3.4. Indirect carbonation

Indirect carbonation method is the mineral carbonation process which takes place in two steps. It means the reactive component (Ca or Mg) is first extracted from the minerals as an oxide or hydroxide in the first step and subsequently reacted with CO₂ to form stable carbonates in the second step (Bacocchi *et al.*, 2010; Bertos *et al.*, 2004). The extraction of magnesium oxide (MgO) or magnesium hydroxide Mg(OH)₂ is performed at atmospheric pressure and is subsequently carbonated in the second step at a higher temperature of 500°C and 20 bars pressure (Zevenhoven *et al.*, 2008). Carbonation of Mg(OH)₂ is faster than carbonation of MgO. Therefore, to accelerate carbonation, Mg(OH)₂ can be used with the carbonation reaction given by the equation below.



To accelerate the carbonation process acids such as acetic acid are used (Teir *et al.*, 2007a). For instance, acetic acid speeds up the extraction of Ca from feedstock rich in Ca. However, use of additives such as acetic acid is also prone to cause leaching of other elements such as heavy metals during the Ca extraction phase cause impure carbonate precipitation as well as causing environmental hazards (Sanna *et al.*, 2014a; Sipilä *et al.*, 2008b).

Katsuyama *et al.* (2005) investigated the viability of producing CaCO₃ from waste cement using water slurry. The Ca is first extracted from pulverised waste cement at high CO₂ pressure, followed by carbonation and finally

precipitation of CaCO₃ was achieved at lower CO₂ pressures (Katsuyama *et al.*, 2005). Under these conditions, high concentration CaCO₃ (up to 98% purity) was obtained at comparatively high reaction rates. Additionally, if the produced CaCO₃ could be ultra-high purified CaCO₃ (>99% purity) the profits could be significantly increased (Katsuyama *et al.*, 2005). Similar to the process described above, proposed by Katsuyama *et al.* (2005) explained a process for producing CaCO₃ from different solid waste residues in a study (Katsuyama *et al.*, 2005). In the study, two cases of the process were described; one employing steel slag and another employing paper bottom ash. However, both the cases did not provide information on the rate of reaction for the precipitation reaction. The extraction of Ca took place into a stirred reactor filled with water for 15 minutes, which lead to a concentration of 0.46 g per litre for steel slag and 1.1 g of Ca(OH)₂ per litre for paper bottom ash respectively. The hydroxide slurry thus formed was separated from the solids and carbonated by infusing it with 25 ml of CO₂ per min (Katsuyama *et al.*, 2005). However, before making any decision, the viability of this process should be examined by environmental and cost assessment, which is also explained in a study by Gorset *et al.* (2007) (Sipilä *et al.*, 2008b). Gorset described a method of precipitating pure MgCO₃ from olivine. The method included one dissolution step followed by precipitation of MgCO₃ and amorphous silica using two precipitation steps is fast enough to consider it for industrial applications. The required acidity (pH 3-5) is to be attained using pressurised CO₂ (50-150 bar) and high temperature (100-170°C), while the subsequent step comprising MgCO₃ precipitation, occurred in a separate reactor with preferably a higher temperature (140-250°C) and a lower CO₂ pressure (50-80 bar) supporting the precipitation of products. Experiments confirmed a high degree of purity, ranging from 99.44 to 99.28% MgCO₃, of the precipitated carbonate (Sipilä *et al.*, 2008b).

Another variant of the two-step aqueous carbonation process is the pH swing process where the pH of the solution is first dropped to enhance the extraction of divalent mineral ions and then raised to increase the precipitation of carbonates (Ababou *et al.*, 2017; Aminu *et al.*, 2017; Uliasz-Bocheńczyk and Mokrzycki, 2017; Wang *et al.*, 2018, 2017; Shashikant Yadav and Mehra, 2017a). Various acids and bases such as HCl, HCOOH, CH₃COOH, HNO₃, H₂SO₄, KOH, NH₃, NaOH, NH₄Cl, NH₄NO₃, and (NH₄)₂SO₄ have been tested as additives to enhance extraction in the two-step aqueous carbonation process. The acidic solutions were more effective than the basic solutions in Mg and Ca extraction (IPCC, 2005; Olajire, 2013; Sanna *et al.*, 2014a). In the extraction experiments, H₂SO₄ was observed to be the best extraction agent of all the chemicals tested, but none of the acids was capable of extracting Mg selectively (Sipilä *et al.*, 2008c). For this selectivity, the ammonium salts tested achieved better with no Fe or Si presence in the solution after 1 h. However, with the ammonium salts, the quantity of Mg extracted continue to exist low at only 0.3-0.5%. The influence of the particle size did not affect the Mg

extraction rate considerably for a particle size range of 125-500 µm. At a temperature of 70°C and a reaction time of 2 hours, all mineral acids (HCl, H₂SO₄, HNO₃) were able to extract 100% Mg from the serpentine sample. The rate-limiting step was found to be product layer diffusion for Mg extraction (Teir *et al.*, 2007b). Experiments have also shown that substantial extents of Mg and Fe extracted from serpentine leaving behind essentially residue SiO₂ using 5 M H₂SO₄ at temperatures above 80°C (Cheng and Hsu, 2006).

4. Carbonation using brines

A significant amount of brine is released as a waste product during natural gas and oil extraction. The abundant availability and the presence of high concentration of mineral ions that can form stable carbonates (mainly Mg and Ca) make it suitable for CO₂ sequestration using the carbonation process (Druckemiller and Maroto-Valer, 2005; Maroto-Valer *et al.*, 2005). However, large-scale carbonation of brine is currently restricted by sluggish reaction rates. Carbonation process can be accelerated by increasing the pH of the brine solution, but uncertainties regarding other parameters such as the composition of brine, pressure and temperature need to further research (Nyambura *et al.*, 2011; Soong *et al.*, 2006).

5. Accelerated weathering

CO₂ can be captured and stored by accelerated weathering of limestone. The product of an accelerated weathering of a limestone plant is CaCO₃ solution that could quickly be released into the ocean and diluted with insignificant or even a positive environmental impact (Rau *et al.*, 2007). However, further investigation is necessary before this method applied on a large scale as there are still other concerns to deal with, such as the energy requirement for the transportation of enormous quantities of Ca-rich minerals or industrial wastes to the processing site that is possibly situated in proximity to a CO₂ source as well as a probable disposal site. By merely spreading a Ca or Mg-rich mineral such as olivine on land site having a problem of high acidity would simultaneously improve the soil quality by increasing the pH of the soil as well as capture CO₂ from the ambient atmosphere in approximately 30 years (Sipilä *et al.*, 2008b). This simple method to CO₂ sequestration has been proposed by Schuiling and Krijgsman (2006) who emphasized that this approach should be applied with caution to investigate the environmental impact of spreading vast quantities of mineral rocks on the ground (Schuiling and Krijgsman, 2006).

6. Pre-treatment

Accelerating the reaction rates of mineral carbonation process is a widely researched domain. While several studies have focused on improving the process itself, some studies have investigated the activation of the feedstock by different pre-treatment methods, including heat, mechanical, and chemical pre-treatment (Huijgen and Comans, 2003; IPCC, 2005). An alternative suggested

by Maroto-Valer *et al.* (2005), is a combination of both the chemical and mechanical pre-treatment which, when applied on raw serpentine, increased the surface area from 8 m² per gram to over 330 m² per gram (Maroto-Valer *et al.*, 2005). The major limitation of most pre-treatment method is the high energy demand, and it is found that only conventional grinding is energy efficient and hence economically feasible (Huijgen and Comans, 2005).

7. Comparison of various sequestration routes

Table 2 presents the summary of comparison of different carbonation routes adopted by several researchers. Lackner *et al.* (1995) carried out investigations on the process of direct gas–solid carbonation for the first time. The process involves the direct conversion of silicate or oxide minerals to carbonates with the use of gaseous or supercritical CO₂ (Lackner *et al.*, 1995). The sluggish reaction kinetics and the considerable requirements of energy consumption have limited the usage of the gas–solid method on a large-scale (Bacocchi *et al.*, 2006; Zevenhoven *et al.*, 2008). Hence, the direct gas–solid carbonation pathway for minerals was abandoned, and investigations on other reliable pathways were undertaken. After the year 2005, a considerable amount of advancements have been made with regard to mineral

carbonation and the new approach of multistage gas–solid carbonation has been proposed. However, investigations concerning about its large-scale viability suggested that the deeper understanding and interpretations of experimental and analytical studies are required (Fagerlund *et al.*, 2011; Prigiobbe *et al.*, 2009).

Additionally, in recent years, direct aqueous carbonation method has been extensively studied. However, it is also affected by slow reaction kinetics and high cost in comparison with the other CO₂ sequestration approaches. As determined by O'Connor *et al.*, (2005), the most optimum cost for direct aqueous carbonation process was found to be equal to \$54 per ton of sequestered carbon dioxide, approximately. In this technique, studies were mainly concentrated on the improvement of reaction kinetics by the utilization of additives, or by maximizing the values of temperature and pressure. However, significant developments are yet to arrive. Novel approaches for studying the basic mineral carbonation methods were suggested as there is a scope for advancement, including additive recovery, which has not been investigated thoroughly (Bobicki *et al.*, 2015; Centi and Perathoner, 2014; Jafari *et al.*, 2017; Li *et al.*, 2013; Olajire, 2013; Paul *et al.*, 2017; Xu *et al.*, 2017).

Table 2. Comparison of different carbonation routes

Route	Description	Advantages	Disadvantages	References
Gas–solid	Solid feed directly reacts with CO ₂	Straightforward Production of steam and electricity Utilization possibility of waste steam	Sluggish reaction Thermodynamic limitation non-viable	(Lackner <i>et al.</i> , 1997; O'Connor <i>et al.</i> , 2005)
Aqueous	Carbonation reaction under aqueous medium. Additive chemicals are also used to enhance carbonation rate	High capacity	Energy-intensive Needs additives No additive recovery Expensive	(Shashikant Yadav and Mehra, 2017a, 2017b)
HCl extraction	HCl employed to extract reactive components	Easy recycling of HCl	Energy-intensive Costly	(Haug <i>et al.</i> , 2010; Zhao <i>et al.</i> , 2010)
HNO ₃ extraction	HNO ₃ used to extract reactive components	Energy efficient Low cost	Non-recovery of chemicals	(Doucet, 2010; Teir <i>et al.</i> , 2009)
Molten Salt	Molten salt used as the extraction agent	Energy efficient than HCl	Highly corrosive Unwanted by-products	(Newall <i>et al.</i> , 2000; Olajire, 2013)
Ammonia extraction	Ammonium salts employed to extract reactive components	Pure products Fast reaction Recyclable	Expensive Limited literature	(Fagerlund <i>et al.</i> , 2012; Sanna <i>et al.</i> , 2014b)

In order to improve the reaction kinetics, many multi-steps indirect carbonation routes have been investigated, where extraction of minerals and then carbonation of extracted minerals were performed in separate reactors. Hence, the necessity of keeping two opposite reactions (i.e. dissolution and precipitation) within the same reactor would be done away with. This makes the aqueous carbonation by the indirect method as the most appropriate approach. For most of these pathways,

considerable quantities of strong bases and acids are essential, thereby resulting in the identification of substantial demand for low-cost acid/base alternatives and acid/base recovery (Doucet, 2010; Dri *et al.*, 2013; Eloneva *et al.*, 2012). Nevertheless, the costs involved in recovery continue to be overpriced, and the extent of carbonation is less during the utilization of low-cost or weak acid/base components. Further, other elements, including heavy metals are subjected to leaching in the

course of the extraction process, thereby resulting in the precipitation of impure carbonate and various other environmental hazards to the surroundings. In order to reduce the cost of CO₂ sequestration, it is important to have such a carbonated product in the post-processing, which possesses the significant market value. For instance, studies have shown that the cost of production of calcium carbonate of ultra-high purity is around \$323 per cubic meter in comparison with the commercial cost of \$10000 per cubic meter (Katsuyama *et al.*, 2005). Also, carbonation of waste substances, including cement waste and steel slags could lead to a reduction in the mining of fresh rocks for CO₂ sequestration processes. Moreover, the utilization of waste materials in the form of CO₂ sequestration feedstock reduces the quantity of wastes being dumped as landfill. It helps in decreasing the costs incurred due to storage of waste material and the running costs involved in mineral carbonation (Shashikant Yadav and Mehra, 2017a, 2017b).

However, there is no ideal pathway of mineral carbonation, which warrants further research for the establishment of novel approaches or improvisation in the course of the existing pathways. For the identification of a reliable route, it is essential to review the upstream and downstream processes as well. For instance, employing flue gas instead of pure carbon dioxide and circumventing the capture costs. However, the transportation costs of the material to the source of carbon dioxide should also be taken into account.

8. Conclusion

Direct aqueous carbonation currently considered the most effective mineral sequestration method. However, it is costly in comparison to other CO₂ sequestration methods. In the light of the advancements made in indirect aqueous carbonation process, the increased investment costs of a more complex reactor system could be offset by eliminating the requirement for balancing two opposite, dissolution and precipitation reactions, in a single reactor making indirect aqueous carbonation the new attractive route. However, multi-stage mineral carbonation need more research before any critical decisions can be made regarding its large-scale feasibility. The overall understanding of Mg and Ca carbonation reactions has developed significantly. However, little advancement has been achieved for the recycling and recovery of additives, which is a significant hurdle in large-scale industrial implementation of mineral sequestration. For mineral carbonation to become a large-scale CO₂ sequestration alternative, further studies are required for efficient use and/or environmentally safe disposal of the massive amounts of carbonates produced.

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