

Sequestering CO₂ by Mineral Carbonation: Stability against Acid Rain Exposure

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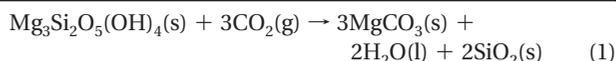
Mineral carbonation is a potentially attractive alternative to storage of compressed CO₂ in underground repositories, known as geosequestration. Processes for the conversion of basic ores, such as magnesium silicates, to carbonates have been proposed by various researchers, with storage of the carbonate as backfill in the original mine representing a solid carbon sink. The stability of such carbon sinks against acid rain and other sources of strong acids is examined here. It is acknowledged that in the presence of strong acid, carbonates will dissolve and release carbon dioxide. A sensitivity analysis covering annual average rainfall and pH that may be encountered in industrialized areas of the United States, China, Europe, and Australia was conducted to determine maximum CO₂ rerelease rates from mineral carbonation carbon sinks. This analysis is based on a worst-case premise that is equivalent to assuming infinitely rapid kinetics of dissolution of the carbonate. The analysis shows that under any likely conditions of pH and rainfall, leakage rates of stored CO₂ are negligible. This is illustrated in a hypothetical case study under Australian conditions. It is thus proposed that sequestration by mineral carbonation can be considered to be permanent on practical human time scales. Other possible sources of acid have also been considered.

Introduction

Carbon capture and storage (CCS) is widely viewed as an essential technology to reduce global emissions of CO₂ from fossil fuel combustion (1). CCS is most applicable to large stationary sources of emissions, such as coal-fired power stations. Currently, storage via geosequestration is receiving most attention. This involves pumping compressed carbon dioxide into underground reservoirs such as expired natural gas and oil fields, saline aquifers, or deep unminable coal seams (1).

Ex situ mineral carbonation is also being investigated as an alternative to conventional geosequestration. Since the pioneering development work by Lackner et al. (2) various mineral carbonation processes have been reported in literature. These processes involve using alkaline earth metal bearing ores to react with carbon dioxide to form solid carbonates, which can be stored in a benign state.

O'Connor et al. (3) demonstrated a process to react magnesium and calcium silicate ores with carbon dioxide to produce solid carbonates. Reaction 1 shows the primary reaction for the case of serpentine.



The process requires reaction of treated silicate ore with supercritical CO₂ in an aqueous medium. The main components of the product stream from the carbonation process would include magnesite (or calcite for a wollastonite feedstock), silica, possibly silicic acid and entrained water, plus any dissolved additives such as sodium chloride and sodium bicarbonate as used in the processes developed by O'Connor et al. (3). It is envisaged that the bulk of the carbonation product would be backfilled into the mining void where the original ore was extracted, forming an artificial global carbon sink.

One particular advantage over other CO₂ storage options that has been highlighted by proponents of mineral carbonation is the relative permanence, at least on practical human time scales, of the sink. Any leakage of sequestered carbon dioxide would raise two potential concerns. The first is a local safety issue. With a greater molecular weight than air, the release of a large quantity of CO₂ has the potential to linger at ground level and asphyxiate humans and animals, such as the incident at Lake Nyos in 1986 (4). However, the release mechanism for carbon dioxide from carbonate would necessarily be a gradual one. The second concern is one of overall efficiency, in that a large-scale rerelease of carbon dioxide to the atmosphere on human time scales would defeat the purpose of CCS. Related to this is the economic aspect, with a monetary value likely to be associated with sequestered CO₂ under carbon emissions trading or tax regimes. Leakage of sequestered CO₂ will thus have both problematic environmental and economic outcomes. Assured permanence would remove these risks and also eliminate the requirement of long-term monitoring, which is seen as a necessary impost on geosequestration. Many factors will influence the ultimate choice of sequestration route. Sufficient capacity, overall process costs, energy penalties, and importantly the safety and permanency of each option will all be necessary considerations.

The focus of this paper is the stability of the product of the mineral carbonation process against acid rain. The key considerations and conclusions apply equally to magnesium or calcium carbonates; however, magnesite has been chosen as the main focus of this study since deposits of rocks bearing magnesium silicate ores, such as serpentinite, are widely distributed globally (5–7) and in sufficient quantities to sequester CO₂ arising from industrial emission sources for many decades (8). On the other hand, calcium silicates such as wollastonite are relatively rare, limiting their potential for industrial scale mineral carbonation (3).

Acid rain may be expected to react with carbonates wherever they are encountered, such as in natural deposits of limestone, dolomite, or other carbonate minerals. The issue under study here is analogous to that of natural carbonate deposits; however, it addresses the particular case of artificially constructed carbon sinks for the purpose of long-term CO₂ sequestration. The interaction of acid rain with artificially constructed carbonate deposits is likely to differ from that with natural deposits, due to physical and chemical differences. For example, the artificial mineral carbonation product is likely to be a fine precipitate, which may be expected to react and dissolve more rapidly than a bulk rock mass in natural limestone and dolomite deposits. In both cases however, release of CO₂ is likely to occur.

Theoretical Background

Acid Rain in Regions with Mineral Carbonation Potential. Almost all rain is slightly acidic (averaging pH 5.6) due to the

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equilibrium of carbon dioxide and water present in the atmosphere (10). The pH of acid rain can be substantially lower than this due to anthropogenic emissions of NOx and SOx from coal-fired power plants, chemical and mineral processing plants, and exhaust gases from motor vehicles. Thus the pH of rain around heavily urbanized and industrialized areas is generally lower than in areas that are sparsely populated. However in recent years the trend has been toward an increase in the installation of technology on plants that emit NOx and SOx to limit these emissions.

Large serpentinite occurrences in the U.S.A. have been identified as candidates for the mineral carbonation sequestration process (3, 8). The largest of these occur in the western states where acid rain is less of a concern; however, some prospective deposits have been identified in the eastern states. Clearly, sites in close proximity to coal-fired power generation are of interest and thus may be in areas where acid rain is more acute, where average rain pH has been measured to be as low as 4.3 (11).

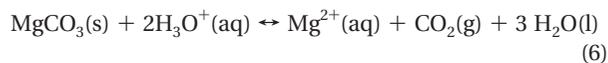
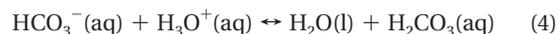
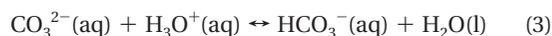
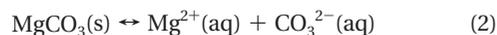
The deployment of CCS is especially relevant to countries with high current and projected growth in coal-fired power generation, notably China. China has an extremely high population density in its southeastern provinces. Larssen et al. (12) conducted a study that focused on the biological effects of acid deposition in this region and highlighted sources of SOx in China. Larssen et al. (12) also presented data on rainfall pH for the most highly industrialized regions of China. In areas worst affected by anthropogenic NOx and SOx emissions, the pH of rainfall was quoted to be as low as 4. The study also suggested that much of the anthropogenic NOx and SOx emissions are neutralized by alkaline dust from both deserts and industrial emissions, and if this were not the case an average rainfall pH as low as 3 may have resulted in some places.

Europe also has deposits of olivine and serpentinite which could be utilized for mineral carbonation. Examples are the mafic and ultramafic rock deposits in Finland reported by Teir et al. (13) and in Turkey reported by Baris et al. (14). Europe has had problems with acid rain since the 1970s (10). A report released by the European Monitoring and Evaluation Program (EMEP) (15) shows that the pH of rainfall varies from about 4.5–6.0 across the European continent.

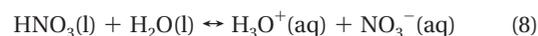
A review of acid deposition in Australia by Murray (16) stated that, due to the demography, geographic isolation, and use of low-sulfur fuels the potential for acid deposition in Australia is low in comparison to Europe and North America. It was found that the worst case occurred in the noncoastal rainfall of the Hunter Valley, where the average rainfall pH was 4.66 (17). However, results from a larger scale study of the same area found the average annual rainfall pH in the Hunter Valley to be 5.3, with pH data between 4.2 and 7.0 (18). Serpentinite deposits within close proximity to the Hunter Valley are considered in the Australian case study presented here.

Based on these regional scenarios, a pH of 3 was the lowest pH accounted for in the sensitivity analysis in this paper. This represents a worst-case scenario for acid rain. However, it must be noted that in the context of large scale uptake of CCS, NOx and SOx emissions should decrease since they will be scrubbed from flue gases along with CO₂. Acid rain severity is thus actually likely to decrease as CCS ramps up. Also, the scale of anthropogenic NOx and SOx emissions from typical combustion processes is 2 orders of magnitude lower than that of CO₂ emissions.

Potential for Carbonate Dissolution by Acid Rain. The dissolution of carbonate leading to release of carbon dioxide gas is similar to the natural bicarbonate buffer system found in seawater. Reactions 2–5 describe the dissolution process for magnesite, with overall reaction 6 (19, 20).



Two moles of hydronium ions are required to release a mole of carbon dioxide. For this dissolution process to occur, a source of protons is required. Two obvious sources are acid rain or acidic products from the mineral carbonation process itself. Acid rain is formed by complex reactions between oxides of sulfur and nitrogen in the atmosphere (19, 20). The resulting strong acids dissociate in solution with water, as shown in reactions 7 and 8 (19, 20).



Previous work on the dissolution of magnesite and calcite has shown that strong acids or high concentrations of weak acids are required to dissolve carbonates to a significant degree in short time frames (20–24). The work of these authors focused on batch experiments to determine kinetic rate constants for a series of intermediate reactions described by reactions 2–5.

The equilibrium constant for the overall dissolution of magnesite shown by reaction 6 is 1.12×10^{10} , strongly favoring the products of the reaction. Hence for the system described by reactions 2–6, where the stoichiometric requirements are met, at equilibrium the vast majority of magnesite would be dissolved to form magnesium ions, water, and carbon dioxide gas. The time taken to attain equilibrium will be dictated by reaction kinetics and ambient conditions.

Our analysis focuses exclusively on the stoichiometric requirements for the release of CO₂ from magnesite in the presence of acid. Specifically, we investigate whether acid rain would ever provide a sufficient mass flux of protons to release a significant quantity of sequestered CO₂ from mineral carbonates. This approach obviates the need for any kinetic or experimental studies, if it can be demonstrated that the available mass flux of acidic rainfall represents negligible CO₂ leakage rates for sequestration by mineral carbonation.

Results and Discussion

Mass Flux Sensitivity Analysis of Maximum CO₂ Rerelease.

The mass flux of protons that will fall on a specific surface area of a magnesite deposit can be calculated directly from the average pH and volume of rainfall over a certain period for a particular location. Thus the total flux of protons available for the dissolution of magnesite can be determined, setting an upper bound on the total amount of carbon dioxide that can be rereleased. This represents a hypothetical worst case scenario, effectively assuming the kinetics of reactions 2–6 to be infinitely fast, with all protons reacting immediately to rerelease carbon dioxide. Furthermore, the analysis also assumes that all the rereleased carbon dioxide is in gaseous form, and none of it remains in the form of carbonic acid or bicarbonate ions in the magnesite deposit. Again, this is a worst-case assumption.

Figure 1 shows a sensitivity analysis of the maximum possible carbon dioxide rereleased from stored magnesite per unit surface area as a function of rainfall rate and pH. Thus the maximum leakage rates associated with these proton

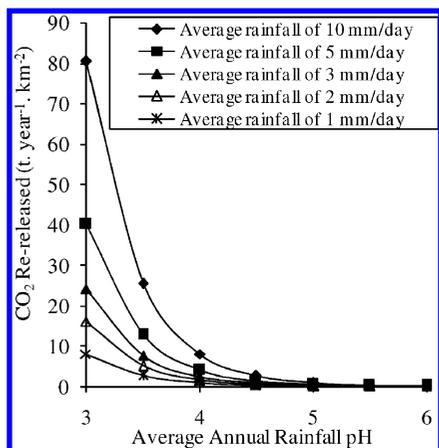


FIGURE 1. Sensitivity analysis for maximum possible CO₂ rereleased per unit surface area from magnesite deposits.

fluxes can be determined for given volumes and surface areas of carbonation product to be backfilled at any particular carbonation mine.

From Figure 1 it can be seen that, for the pH and rainfall ranges considered, the maximum possible rerelease rates would be in the range 0–80 t CO₂ per year per km² of surface area exposed to acid rain. It must be noted that 80 t CO₂ per year per km² is the theoretical maximum at rainfall rates in excess of 3600 mm pa at a pH of 3, which is a highly unlikely scenario. Most feasible scenarios, where pH is 4 or more, would be subject to maximum possible losses of less than 10 t CO₂ per year per km². To put this in context, a mineral carbonation sink of depth 500 m backfilled at a representative bulk density of 2000 kg·m⁻³ would hold in the order of 300 Mt CO₂ per km² as carbonate product. Maximum possible leakage rates would thus be in the order of 0.03–0.3% of the sequestered CO₂ after a 10000-year period of rainfall of 3600 mm pa sustained at a pH of 3–4. More realistic maximum leakage rates are illustrated in the following region-specific case study.

Australian Case Study. Generating approximately four times the amount of carbon dioxide of the Australian car fleet at around 170 Mt pa (25), the Australian coal-fired power industry is a prime target for CCS. The potential for a mineral carbonation process in Australia has been outlined and discussed by Brent and Petrie (26). Two regions where large outcropping serpentinite deposits occur close to existing coal fields and power generation were identified; namely the Great Serpentinite Belt (GSB) in NSW and the area around Gladstone in Queensland. Neither of these regions is in close proximity to currently identified prospective geosequestration reservoirs. The example of NSW is examined here in a case study of CO₂ leakage rates for a mineral carbonation carbon sink.

The total current CO₂ emissions from all NSW power stations are around 60 Mt pa, with the four largest facilities in the Hunter region making up over 70% of these emissions (27). The largest single power generation facility in NSW produces just over 14 Mt of CO₂ pa (28). The case study examined here will consider sequestration of the CO₂ from such a facility at a mineral carbonation mine located in the GSB. Assuming additional CO₂ is generated by the mineral carbonation process itself (26), around 45 Mt pa of serpentinite would be required for sequestration of all the CO₂ from the power generation and the carbonation process.

Open pit dimensions to mine the serpentinite and store the carbonated product can be estimated as follows. Over a 50-year period, the total CO₂ sequestered would amount to 700 Mt, requiring around 2.2 Gt of serpentinite for 100% CO₂ sequestration. The mined volume, and hence mining void, is thus around 800 Mm³ for serpentinite with an in situ density

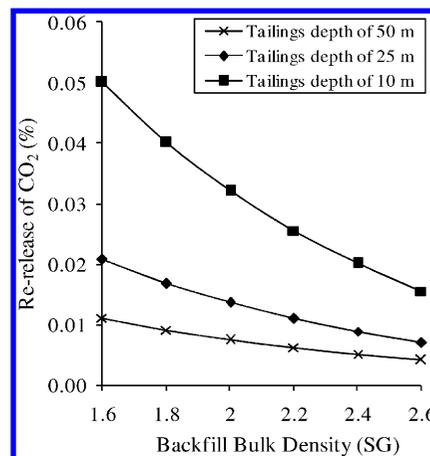


FIGURE 2. Maximum possible carbon dioxide rerelease over 10000 years as a function of backfill bulk density and excess product depth for the Australian case study.

of 2600 kg·m⁻³, which is typical for this region (29). The mass of silica and magnesite product from the mineral carbonation process will amount to about 2.8 Gt, including around 10% moisture. The density of the mineral carbonation product will be a function of the mineral composition, water content and voidage. With full conversion of magnesium content to the carbonate and voidage in the range 10–35%, the density will be in the range 1600–2600 kg·m⁻³, constituting a volume of 1080–1750 Mm³.

It is clear that the original serpentinite mine volume will not be sufficient to store the total volume of product from the mineral carbonation process. There may be a practical or legal limit on the height to which further product may be stacked above the original mine surface level, perhaps conservatively of the order of 50 m. Unless a substantial proportion of the carbonation product can be utilized in other applications, such as construction material, some additional disposal will be required outside of the original mine footprint, preferably in adjacent low-lying regions. This is analogous to tailings dams that are commonplace in the mining industry.

The average annual pH of rain in the GSB area is estimated to be 4.5–5 (16). The average annual rainfall for the past 15 years based on formal data at the Woodsreef area, a past serpentinite mining area centrally located in the GSB, is 710 mm pa, or just under 2 mm per day (30). From Figure 1, it can be seen that under these conditions, the annual total mass flux of protons would allow a maximum release rate of 0.5 t CO₂ per km² pa.

Figure 2 shows the rerelease of carbon dioxide as a function of backfill density and changing tailing depths outside of the original mining void, for a 10000-year time frame as a percentage of the total sequestered in a 50-year operational life of the original power station and mine.

As the backfill bulk density and excess product depth decrease the maximum rerelease of carbon dioxide increases. However the magnitude of rerelease is still considered to be a negligible fraction of the stored CO₂, even for the lowest likely densities and allowable excess carbonate product depths.

Large Catchment Areas and Watercourses. It must be noted that in the context of total proton flux from acid rain, the exposed surface area could include land that forms part of a catchment for any watercourses that run through the carbonation site. Mines should thus preferably not be sited on any existing or potential watercourses with large catchment areas subject to intense acid rain. Even with this consideration, the potential catchment area would need to exceed the directly exposed carbonate stockpile area by several orders of magnitude before significant release rates

of CO₂ would be possible on a 10000-year time scale. In the case of the GSB, the serpentinite is located at elevations of several hundred meters above surrounding low lying land so any watercourses are likely to be limited in size and catchment area. Elevated outcrops of serpentinite are likely to occur in other regions.

Process Additives. A sodium chloride/sodium bicarbonate system was used in the aqueous mineral carbonation process proposed by O'Connor et al. (3). If the bicarbonate ion from this system was to leave the process with the bulk tailings, the bicarbonate ion would only require one mole of hydronium ions to rerelease one mole of carbon dioxide gas, compared to the two moles required for dissolving carbonate. This would start the dissolution process at reaction 4 instead of reaction 3 and effectively double the amount of CO₂ released per mole of acid. However, taken over a period of 10000 years, the ultimate limiting factor remains the total proton flux provided by acid rain. Even assuming that all the acid reacts with bicarbonate instead of carbonate, thus doubling CO₂ release, for all scenarios examined in this case study the rerelease of CO₂ over a 10000 year period would still be below 0.1% of the total sequestered.

Silicic Acid. Aqueous mineral carbonation occurs in two steps; dissolution of metal ions into solution followed by carbonation of the metal ions in solution. During the dissolution step, silicates in the ore may be released into solution and have theoretically been proposed to occur in the process as both silica (SiO₂) and ortho-silicic acid (H₄SiO₄) (3). Despite its theoretical occurrence, it is noted that no silicic acid was actually reported in the analyses of the solid or liquid product streams by O'Connor et al. (3). Ortho-silicic acid is a very weak diprotic acid with a pK_{a1} and pK_{a2} of 9.8 and 13.2, respectively. This means that the silicic acid will only start to noticeably dissociate to form protons at pH greater than 9.8, as shown by reactions 9 and 10.



Hence if ortho-silicic acid was formed, extremely alkaline conditions would be required to dissociate the acid to form a source of protons. From consideration of reactions 2–6 the release of carbon dioxide gas would not occur under alkaline conditions.

Other Sources of Acid. It is possible that other sources of acid may lead to rerelease of CO₂ from artificial carbonate sinks. For example, acid may be transported into the sink from external sources, such as acid mine drainage in sulphidic ore bodies. However, given the location of the proposed sinks within large basic ultramafic rock formations, such acid sources are unlikely to be present in the immediate vicinity. Consideration should however be given to factors such as watercourses or other potentially large sources of acid before locating mineral carbonate sinks. Again, similar calculations around maximum possible acid fluxes could be performed to set upper bounds on the possible rerelease of CO₂.

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