



Long-term Stability Assessment of Geological Sequestration of Carbon Dioxide Based on Mineral Carbonation

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ABSTRACT

Geological sequestration of carbon dioxide (CO₂), being one of the important methods to deal with anthropogenic climate change, is to reduce the large scale emission to the atmosphere. Among all the trapping mechanisms, mineral carbonation, i.e., the reaction between CO₂ and silicate minerals to form stable, solid carbonate minerals, is considered the most secure and long-term solution to effectively isolate CO₂. This paper carries out a thorough and extended examination of the long-term stability of CO₂ sequestered through in-situ mineral carbonation. It probes the key geochemical reactions shaping the change of injected supercritical CO₂ into solid mineral forms, focusing mainly on mafic and ultramafic rock formations abundant with olivine, pyroxene and plagioclase. The assessment framework described here looks at main interdependent factors that affect secure storage for an extended amount of time. Includes a detailed analysis of intrinsic reaction kinetics of silicate dissolution, evolution of host rock's geomechanical property under coupled thermo-hydro-mechanical-chemical (THMC) stress, and the whole-scale integrity of the overlying caprock. Investigate how varying porosity and permeability brought on by competing mineral dissolution and carbonate precipitation alter the reservoir's hydraulic properties and long term containment system. In addition to that the paper talks about the role of advanced numerical modeling of the reactive transport to predict the behavior of the storage reservoir over a span of few decades to millennial scales. And they're important for predicting the rate of mineralization, the extent of the carbonate precipitates, and any geomechanical risks like earthquakes or cap rock fracturing. Lastly we look at state-of-the-art observation and inspection technologies that are needed for observing the subsurface plume and verifying the advancing mineral carbonation and certifying the long-term stable and environmentally safe sequestration project. The integrated assessment shows that although mineral carbonation provides the most solid solution for carbon dioxide storage, it is still based on the strict, site-specific examination of geological, geochemical, and geomechanical parameters from the laboratory scale to the field scale.

KEYWORDS

Carbon Dioxide Sequestration, Mineral Carbonation, Long Term Stability, Geochemical Reactions, Caprock Strength, Reactive Transport Simulation, Geomechanics.

1. INTRODUCTION

The increasing concentration of CO₂ in the atmosphere, which is mainly due to burning fossil fuels for industry and generating electricity, is considered the main cause of global climate change. and this has caused a world wide hunt for scalable, efficient answers to greenhouse gas removal[1]. Carbon capture and storage (CCS), as a crucial path of technology, is for power generation industries and the hard-to-delimitable industries such as cement factories and steel plants to achieve decarbonization. Its principle is to capture CO₂ emissions at point-sources and inject them into deep

formations of rock underground permanently so they don't get back to the atmosphere. The long term security and efficacy of geological sequestration is critical if it is to be accepted by the public, gain regulatory approval and succeed in helping to mitigate climate change[2]. Deep saline aquifers and depleted oil/gas reservoirs - these are typical storage formations - they use various different trapping mechanisms. Structural trapping over an impermeable caprock, residual trapping of CO by immobile liquid droplets in pores, and solubility trapping of CO in the formation brine. It's like the way these systems work is over decades or even centuries, there is still a small chance, no zero risk, of leakage[3]. Furthermore, a structurally defined trap might still be compromised by an undiscovered fault or fractures induced by an injection. Solubility trapping can drop the pH of brine, it may eventually destroy the caprock, a change in reservoir pressure – temperature might cause CO₂ to come out. However, mineral carbonation is what you would call the ultimate and most stable sink for Carbon Dioxide. It basically locks it up into a solid state as Carbonate, a very thermodynamically stable substance. The process imitates the natural breaking of rocks. The injected CO₂ is transformed into non-reactive carbonate rocks. This includes calcites (CaCO₃) and magnesites (MgCO₃) as well as dolomites (CaMg(CO₃)₂), which are stable over long duration of time. The inherent lastingness gives it good protection against CO₂ from underground storage eventually leaking out into the air. So then, if we wish to evaluate the long-term stability of geological sequestration locations where mineral carbonation is the favored primary trap form we must utilize an individual and specific strategy which factors in both geochemistry as well as geomechanics. The goal of this paper is to offer a thorough and thorough scrutiny of the elements that impact the term length stability of Carbon dioxide taken up as the in situ mineral carbonation happens over time, with primary emphasis placed on the geochemical workings and the reservoir's geomechanical adjustments, much sophisticated techniques for prediction being adopted alongside vital technologies aimed at watching the situation and verifying results[4].

2. GEOCHEMICAL FOUNDATIONS OF INS - SITU MINERAL CARBONATION

The best long term security of mineral carbonation comes from the series of chemical reactions that take gaseous, buoyant, free, and probably leaky supercritical CO₂ and make it into a very immobile, solid mineral. The first step starts once the CO₂ being injected has dissolved into the surrounding formation brine, thus replenishing the weak H₂CO₃. This acid breaks down to give us bicarbonate(HCO₃⁻) and carbonateCO₃²⁻ ions and it also releases H⁺ - ions which will really cut down the pH of the pore water. This newly acid fluid becomes an aggressive solvent for the host-rock -main constituents of the primary silicate mineral. Geologic formations rich with divalent Cations like huge basaltic areas in the Pacific North West of the US plus the Deccan Traps in India or ultramafic peridotites like in the Omani desert are regarded as very good places to use this method. And these rocks have plenty of chemically eager minerals, like olivine (Mg, Fe)₂SiO₄, pyroxene (Ca, Mg, Fe)₂Si₂O₆, and specific plagioclase feldspar (Na, Ca)AlSi₃O₈. The acidic brine then aggressively solubilizes needed divalent metal cations, such as Ca²⁺, Mg²⁺, Fe²⁺, from the silicate mineral matrix in a dissolution step, which is usually the rate limiting step. For instance, the dissolution of forsterite – the magnesium rich endmember of olivine - can be written as: Mg₂SiO₄ + 4H⁺ → 2Mg²⁺ + H₄SiO₄[5]. And when those cations come out into water, they grab some hydrogen, making that salt water more like a base. And when the quantity of those suspended cations and any available carbonate ions gets to a point of oversaturation, the brand new, steady carbonate minerals start to form from the solution, and the CO₂ is permanently stuck. This precipitation reaction like Mg²⁺+CO₃²⁻→MgCO₃(magnesite) is effectively consuming the dissolved CO₂ and draws more gasified CO₂ into the solution, the above reaction would continue going forward until either the reacting mineral is used up or the supply of CO₂ is consumed. Its permanence is its biggest strength. The mineralised carbonates would be thermodynamically stabilized under normal reservoir conditions. It will be very unlikely for reversal and CO₂ be released without substantial geologic or thermal changes[6]. The over all efficiency as well

as rate of carbonation depends on quite an intricate mix of various factors like the individual mineralogy of the host rock, the active surface area which could participate in reaction, temperature and pressure conditions within the reservoir and the formation fluids' pH as well as water chemistry conditions. Comparison about common silicate mineral with theory of CO₂'s sequestration can be seen on the below table (table 1)[7].

Table 1: Properties of Common Silicate Minerals for CO₂ Carbonation

Mineral	Chemical Formula	Molar Mass (g/mol)	Theoretical CO ₂ Uptake (g CO ₂ /g mineral)	General Reactivity
Forsterite (Olivine)	Mg ₂ SiO ₄	140.69	0.626	High
Fayalite (Olivine)	Fe ₂ SiO ₄	203.78	0.432	Moderate
Wollastonite	CaSiO ₃	116.16	0.379	High
Diopside (Pyroxene)	CaMgSi ₂ O ₆	216.55	0.406	Moderate
Anorthite (Plagioclase)	CaAl ₂ Si ₂ O ₈	278.21	0.158	Low
Serpentine	Mg ₃ Si ₂ O ₅ (OH) ₄	277.11	0.477	Low to Moderate

3. GEOMECHANICS AND STRUCTURE OF STORAGE COMPLEX

Even though it is under this geological environment with the geochemistry providing a very good permanent storage process for mineral carbonation, the stability and containment security of the whole sequestration also depends on the geomechanical and structural stability of the storage facility. This block is not only confined to our target storage reservoir, but it can include the cap rock above as well as those faults and fractures who might cut through the structure. coupled mineral dissolution - precipitation changes the host rock's own physical and mechanical structure. The breakdown of primary silicate minerals might cause a substantial rise in nearby porosity and permeability, and this could possibly foster high-flow paths (or "wormholes") or impact the pressure distribution throughout the reservoir. Chemical erosion also weakens the rock's structure, decreases the rock's compressive strength and makes it easily compacted by the weight of the rock above it[8]. On the other hand, the precipitation of secondary carbonate minerals, which generally have a higher molar volume than the reactant silicate, tend to fill pores and fractures. This process can greatly reduce permeability and increase the sealing capacity of the formation, which is a desirable condition known as "self - healing." It is the competition between these two processes that results in the final geomechanical state of the reservoir. The biggest concern about any geological storage is making sure the cap rock is safe. The cap rock is like a very low permeability layer (like shale) that is the main hydraulic barrier to the buoyant, mobile CO₂ that wants to go up. But even though mineral carbonation cuts down on the mobile CO₂ bit by bit as time goes by, during the early part right after injection happens, the cap-rock's sealing power is quite important. It is possible that the acidic brine created when CO₂ dissolves could interact with the cap rock, altering its hardness, how easily it shatters, and its movement characteristics as a whole, see Table 2[9]. Injection of huge quantities fo fluids at far-higher pressures

than the in-situ hydrostatic results in the change in hydro static in the form of the effective field. The increased pore stresses may cause the normal stress acting against them preexisting faults is lower and therefore may cause such fault to be brought nearer to shear failure producing microseisms. A full analysis of stability calls for a marriage between geochemical reaction modelling and intricate geomechanical studies, so as to forecast how the rock fabric will alter and check that such alterations don't jeopardize containment of CO₂.

Table 2: Comparison of Caprock Sealing Mechanisms and Potential Failure Modes

Caprock Lithology	Primary Sealing Mechanism	Dominant Minerals	Potential Failure Modes	Interaction with CO ₂
Shale / Mudstone	Low Permeability, High Capillary Entry Pressure	Clay minerals (e.g., illite, smectite), Quartz, Calcite	Desiccation cracking, Hydraulic fracturing, Fault reactivation, Geochemical dissolution	Acidic brine can dissolve carbonate cements but may also induce swelling in some clays, reducing permeability.
Evaporite (Halite)	Extremely Low Permeability, Ductile/Self-Healing	Halite (NaCl)	Dissolution by undersaturated brines, Ductile flow under high differential stress	Largely unreactive with dry CO ₂ , but dissolution can occur if water is introduced. Very effective seal.
Anhydrite	Low Permeability	Anhydrite (CaSO ₄)	Brittle fracturing, Dissolution	Relatively stable, but can undergo slow conversion to gypsum with hydration, causing volume changes.
Carbonate (Limestone)	Low Permeability (if not fractured)	Calcite (CaCO ₃), Dolomite (CaMg(CO ₃) ₂)	Dissolution creating karst or wormholes, Fracturing	Highly reactive with carbonic acid, leading to rapid dissolution and potential loss of seal integrity.

4. LONG-TERM RESERVOIR PERFORMANCE PREDICTION

Given the millenniums span for when the mineral carbonation reactions will run their course, it is out of question for one person to watch all the sequestration happen in their lifetime. Therefore, whether it has long-term stability and how well it performs can only be assessed through numerical modeling and simulation. Reactive transport models (RTMS) is the key part of this attempt to predict, they are kind of labs in our minds, where we mix all the tricky rules about moving fluids, how substances

move around, heat and cold stuff, and special chemical reactions happening in the ground together, trying to guess what will happen to CO₂ deep underground for really long time. These models can simulate the advectations and dissolution of injected CO₂ as well as the displacements of brines and the transport of dissolved chemicals within the network of complex chemical species fluid and host-rock interaction reactions. To create a good and representative model, we require plenty of site characterization data [10]. Includes details of the geological model such as the reservoir's structure and stratigraphy, initial mineralogical composition and spatial distribution, petrophysical attributes including porosity, permeability, and the native gradient of thermal and pressure conditions. The accuracy of any long-term forecast depends on the kinetic rate laws for mineral dissolution and deposition. These rates come from lab tests done with ground up mineral powders, and need to be tuned and scaled up to match much slower reactions that happen over surface area on real stuff in place. The great quantity and complexity of the input parameters are shown in table 3. They run these time-consuming computer simulations for a lot of model time – thousands of years - to predict how much of the mobile carbon dioxide actually gets locked up in the form of carbonate minerals, figure out the long-term storage capacity, and make maps showing the changing size of the carbonated rock over the years. More so, they're also very important for doing a precise calculation of risk. They can spot long-term problems like the caprock slowly becoming weak because of chemical stuff, how carbon dioxide might make use of tiny leaks no one knew about, like spaces between rock cracks, how the place where it stays gets squeezed and let go for a long time, and if it might make the Earth shake. This advanced modeling work gives operators and regulators a sense of what the storage site might look like decades after injection stops, so they can be confident with good science that the CO₂ will stay safely and forever.

Table 3: Example Input Parameters for a Geochemical Reactive Transport Model

Parameter Category	Specific Parameter	Example Value/Description	Significance for Stability Assessment
Reservoir Properties	Initial Porosity	0.15	Governs fluid storage volume and flow characteristics.
	Permeability	100mD (millidarcy)	Controls the rate of CO ₂ plume migration and fluid mixing.
	Reservoir Temperature	85°C	Strongly influences reaction kinetics and fluid properties.
	Reservoir Pressure	15MPa	Determines CO ₂ phase (supercritical) and affects mineral solubility.
Mineralogy	Initial Mineral Volume Fractions	Forsterite: 30%, Diopside: 20%, Anorthite: 15%, Quartz: 35%	Defines the total potential for mineral carbonation.
	Reactive Surface Area	10cm ² /g	A key kinetic parameter controlling the speed of dissolution.

Parameter Category	Specific Parameter	Example Value/Description	Significance for Stability Assessment
Fluid Properties	Injection Fluid	Pure Supercritical CO ₂	The substance being sequestered.
	Formation Brine Salinity	35,000 ppm Total Dissolved Solids	Affects mineral solubility and CO ₂ dissolution.
Injection Strategy	Injection Rate	0.5 million tonnes/year	Influences pressure buildup and the extent of the CO ₂ plume.
	Injection Duration	25 years	Defines the total mass of CO ₂ introduced into the system.

5. CO₂ SEQUESTRATION MONITORING AND VERIFICATION

While predictive modeling gives us an important idea about what might happen in the long term, a really strong and having extra parts Monitoring, Measurement, and Verification (MMV) program is very important for making sure that a place where they trap gases in rocks is safe and works right when we watch it in real time and after putting the gases there. The main purpose of an all-encompassing MMV endeavor is to precisely gauge the underground movement of the injected CO₂ plume so as to spot any possible containment loss well in advance, and with a particular focus on mineral carbonation, to obtain data that can later be utilized to ascertain that the CO₂ has been truly transformed into a secure mineral form. To do this we will have to take a multi-faceted approach and use quite a wide variety of technologies, both in space and over time. We need Geophysical techniques to image deep sub-surface remotely. And 4D-time-lapse seismos-the ones that repeatedly take the same high res 3D seismos in about the same place at different times-are extra helpful: It's because the presence of compressible supercritical CO₂ displacing brine in the rocks porosity causes a large change in the seismic velocity and impedance of the reservoir. They can map where the CO₂ plume is because. The Rock would become stiffer, denser as the mineralization continues, this should be seen on the seismic aswell in the long term. The microgravity surveys and satellite-based surface deformation monitoring(InSAR) can find the small changes caused by the deep mass shift (The replacement of fluid CO₂ to dense carbonate minerals and the change of geomechanical due to the pressure) Downhole Methods which is defined as methods performed in a certain monitoring well. It provides a high-resolution dataset from the storage reservoir. Includes doing regular sampling of the fluids plus geochemical analysis to keep tabs on how the brine's chemistry changes, an obvious raise in Mg²⁺ or Ca²⁺ shows silicates dissolving, that would go with pH and alk going up too because carbonate is coming out of solution as direct and obvious as the mineral gets trapped up there. An overview of key monitors with application is shown in table 4. By incorporating data from all these various streams of monitoring in an integrated framework of some sort, operators could confirm and limit the predictions of their own reactive transport models with hard data, show secure containment to regulators and to the public as well, and build up a convincing, fact-filled argument for the lasting permanence of the sequestered carbon dioxide into the far future.

Table 4: Overview of Long-Term Monitoring Techniques for Geological CO₂ Sequestration

Monitoring Technique	Measured Parameter	Application for Mineral Carbonation	Resolution & Scale	Timeframe
4D Seismic Surveys	Changes in acoustic impedance	Tracks the CO ₂ plume; changes in rock properties can indirectly indicate mineralization.	Reservoir scale (tens of meters)	Intermittent (e.g., every 2-5 years)
Gravity Monitoring	Changes in local gravity field	Detects mass changes; replacement of fluid CO ₂ with denser carbonate minerals causes a measurable gravity increase.	Large scale (hundreds of meters)	Continuous or intermittent
InSAR (Interferometric Synthetic Aperture Radar)	Ground surface deformation (uplift/subsidence)	Monitors geomechanical response to injection pressure and volume changes from mineralization.	Large area, centimeter to millimeter precision	Frequent (days to weeks)
Downhole Fluid Sampling & Geochemical Analysis	pH, alkalinity, dissolved ion concentrations	Provides direct evidence of mineral dissolution (increased Ca ²⁺ , Mg ²⁺) and precipitation (changes in pH).	Local scale (at the wellbore)	Intermittent (quarterly to annually)
Cross-well Electrical Resistivity Tomography (ERT)	Changes in bulk electrical resistivity	Tracks saline brine displacement by less conductive CO ₂ ; mineralization also alters resistivity.	Inter-well scale (meters)	Can be semi-continuous

Monitoring Technique	Measured Parameter	Application for Mineral Carbonation	Resolution & Scale	Timeframe
Well Logging (e.g., Neutron, Density)	Porosity, rock density	Detects changes in pore space and bulk density resulting from carbonate mineral precipitation.	Wellbore scale (centimeters)	During well workovers or with permanent sensors

6. CONCLUSION

Geological sequestration of carbon dioxide by in-situ mineral carbonation is the most rock-solid and science-backed solution for the permanent isolation of industrial CO₂ emissions from the planet's atmosphere over the long term. By chemically changing up and down movement of CO₂ gas to solid, geological carbon carbonate minerals beneath, it makes sure long-time leakage of CO₂ is handled, so no need to always watch over the place using just physical ways to trap CO₂. The long term stability of mineral carbonation storage system is dependent on a large number of interdependent and interconnected factors of geochemistry, multiphase fluids, thermal and geomechanical. Any mineral carbonation project's success and strict security depends on a whole, multi-scale evaluation frame. It must have a good, site-specific knowledge on host formation's mineralogy and fabric so as to supply adequate reactive silicate and favourable reactive surface area. Also needs complete caprock system characterization to secure that is completely contain through through pre-mineralizations stage. Reservoir dynamic changes caused by countering processes of silicate dissolution and carbonate precipitation have to be carefully modelled and looked over in order to get good results such as permeability decline, but also to handle possible problems like rock mechanics weakness or seismicity triggered by injection. Predicting RT is something that needs to be there, it's how we tell them our science, our science says things about the future 60K 1,000's of years down the road. However, we can say that these models are as good as the data on which they are built and must continue to be monitored through this rigorous, multifaceted process of monitoring and validation. This MMV program could stay close watch on the CO₂ that was carried off and supply real, right there evidence as the ongoing changing to stone continues. While some major obstacles surrounding slow reaction kinetics and difficulties associated with translating lab results to subsurface heterogeneity continue to be active and key areas of research, fundamental mineral carbonation principles affirm that it is the gold standard for carbon sequestration. Investment in more pilot-scale field project will be needed if we are to continue narrowing the gap between laboratory theory and large-scale deployment so that we can solidify mineral carbonation as a centerpiece of global climate mitigation efforts.

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