

Mechanisms and Application Potential of CO₂ Mineral Sequestration in Basalt

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ABSTRACT

The increasing levels of CO₂ in the atmosphere have prompted the development of CCUS, which faces challenges such as limited mineral trapping, reservoir capacity, and the risk of leakage. To address these challenges, this review proposes in situ mineral carbonation in basalt formations, which are characterized by rapid carbon mineralization and widespread distribution. The review explores the mineral reaction mechanism in basalt reservoirs using laboratory experiments and numerical simulations or modeling. Representative research conducted in the past ten years is emphasized to indicate better carbon mineralization conditions in basalt formations. Two field-scaled pilot projects in Wallula and Iceland are explained to provide examples for future CCUS projects. However, CCUS in

basalt will only become mainstream with further technological development and policy funding.

Keywords: Basalt formation; CO₂ sequestration; In-situ mineralization; CCUS; CO₂-water-rock reaction; Government assistance

1. INTRODUCTION

At 412 ppm in 2021, atmospheric CO₂ levels are the highest in recorded human history (Friedlingstein et al., 2022) due to fossil fuel use causing global warming. To limit global temperature increases, the 2015 Paris Agreement targets 1.5-2°C (Newell et al. 2021), requiring technologies that can reduce atmospheric CO₂. Carbon capture, utilization, and storage (CCUS) is a well-known technology already being considered at numerous climate conferences, as shown in Fig.1. IPCC IP4

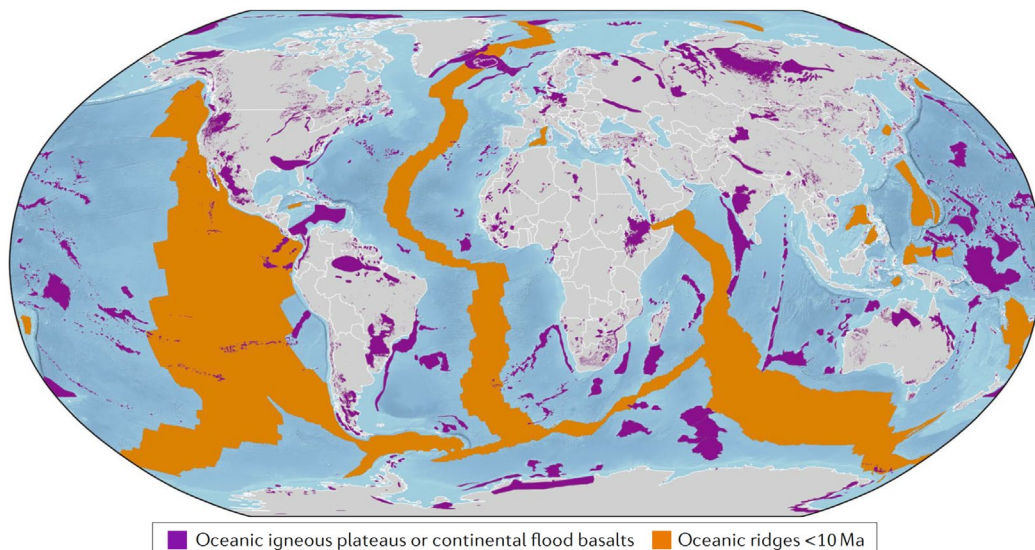


Fig. 2. Locations of feasible geological mafic or ultramafic formations for mineral carbonation (Snaebjornsdottir et al., 2020).

projections set a CCUS objective of 17 bmt by 2050, over 50 times larger than worldwide CCUS totals in 2018. Enhanced oil recovery (EOR) is the foundation of most CCUS operations but faces challenges such as insufficient mineral trapping and constrained reservoir capacity. Alternative CCUS techniques have been suggested, such as in situ mineral carbonation in basalt formations. This technique artificially speeds up the natural process of mineralization by injecting CO₂ into reactive basalt rocks. This review focuses on sequestration in basalt reservoirs, discussing mineral reactions with CO₂, laboratory experiments, numerical simulations, better carbon mineralization conditions in basalt formations, and two field-scaled pilot projects in Iceland and Wallula as precedents for future CCUS projects. However, the widespread use of CCUS and in situ mineral carbonation requires significant financial investments in technology, infrastructure, and effective incentive schemes.

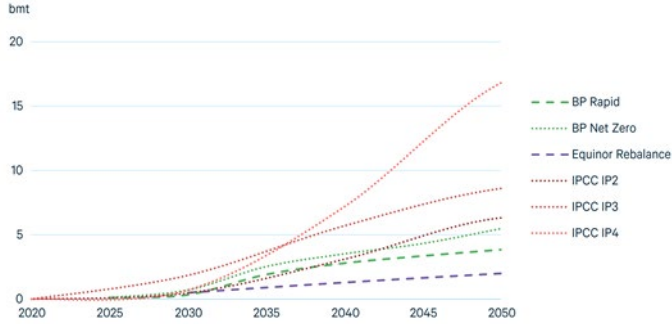


Fig. 1. Required CO₂ removal via CCUS in different scenarios (Newell et al. 2021).

2. CO₂ BASALT REACTION MECHANISMS

2.1 Materials for mineral carbonation

The Columbia River basalt Group, the Decan Traps, Emeishan basalts, and so on, were among the probable basalt formations mentioned in the earlier study as sources of carbon mineralization (Mcgrail et al., 2003). Fig.2 depicts the distribution of the main basaltic formations throughout the world. In this overview, the fundamental chemical knowledge that produced the reaction environment is first briefly discussed. The reaction would always include two primary reactants:

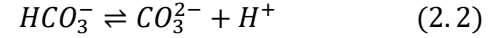
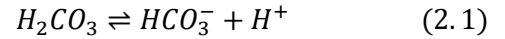
table 1: Bulk mineralogy of the WA-basalt (Al-Yaseri et al. 2021)

Mineral Phase	Weight%
Labradorite	58.6
Anorthite	21.3
Augite	18.9
Nontronite	<1
Ilmenite	<1

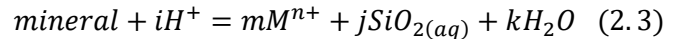
injected CO₂ and subsurface basalt. Basalt formation often involved Wollastonite (CaSiO₃), Forsterite (Mg₂SiO₃), Serpentine or Chrysotile (Mg₃SiO₅(OH)₄), Anorthite (CaAl₂Si₂O₈), Basaltic glass (Na_{0.08}K_{0.008}Fr(II)_{0.17}Mg_{0.28}Ca_{0.26}Al_{0.36}Fe(III)_{0.02}SiTi_{0.02}O_{3.45}) and so on (Oelkers et al., 2008). Table 1 provided a summary of the basalt composition. Numerous injection strategies were demonstrated in earlier studies, including direct pure CO₂ injection, sc-CO₂ injection, CO₂ injection in water solution, and CO₂ injection with H₂S (Matter et al., 2016). Pressure, pH, the composition of acid ions in the formation, and the phase state in the pores are all impacted by the injection method. However, infusing H₂S or another acid solution was mostly done to reduce the pH in the pores rather than to add more acid ions (Schaefer et al., 2010).

2.2 Mineral reaction process

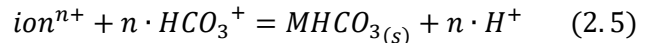
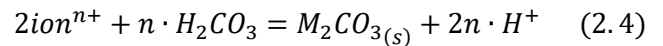
The first scenario involved basalt and a carbonic acid solution precipitating. To release hydrogen ions, the carbon dioxide first dissolves in the water. Additional mixed-in acids may also release hydrogen ions. As a result, carbonic acid can exist as H₂CO₃, HCO₃⁻, or CO₃²⁻ (Khatiwada et al., 2012). The following was the two-step mechanism for dissolving carbonic acid:



Then an acidic solution was used to dissolve the soluble minerals. Even though there were numerous varieties of minerals, the fundamental chemical formula was as follows (Giammar et al., 2005):



Where i, j, k, and n are positive integer variables; M refers to all possible metal ions; and m times n is equivalent to i, j, where j is allowed to be zero. Precipitate eventually developed, indicating that the reaction had successfully mineralized the carbon dioxide and that the process had come to a conclusion:



In the second scenario, water was dissolved in the sc-CO₂ solvent, and CO₂ gas or wet sc-CO₂ was directly in touch with the basalt rock (Schaefer et al., 2011). The sc-CO₂ molecules would react with metallic atoms or materials.

3. MODELING AND EXPERIMENTAL METHODS

3.1 Modeling methods

reactant composition came from prior research, a dataset created using a solver software or in a pilot project, and some experimental studies of the fluid and

Table. 2. Modeling and Experimental studies in past ten years. ~: means changeable or abundant.

Material	pH	Temperature, °C	Pressure, MPa	Product	Perspectives	Reference
Model simulation		250		Pyrite, pyrrhotite, prehnite and epidote	H ₂ S could be sequestration	(Stefansson et al., 2011)
Model simulation	~	[25, 90]	3		Reaction was depended on pH, time, cations	(Gysi & Stefansson, 2011)
Basalts, peridotite with seawater	3.6	25	0.4		Dunitic peridotites Si release was similar to basalt	(Wolff-Boenisch et al., 2011)
Model simulation		[60, 100]	<10	Magnesite, siderite and ankerite		(Pham et al., 2012)
Model simulation		40	<10	Siderite, Fe, Mg carbonates		(Pham et al., 2012)
SW Iceland basalt	~	40	[0.1, 1.28]	Zeolites, clays	CO ₂ concentration changed reaction path	(Gysi & Stefansson, 2012a)
SW Iceland basalt	~	[75, 250]	[1, 2.5]	Ca–Mg–Fe carbonates	Reaction depended on Ca, Mg, Fe and Al-Si mineral kinetics	(Gysi & Stefansson, 2012b)
Basalt seawater		250, 350	50	Calcite		(Shibuya et al., 2013)
Olivine, NaHCO ₃ , NaCl		<70	20	Ferric oxide	NaHCO ₃ was effective	(Gadikota et al., 2014)
Basalt and SO ₂		90	[8.8, 11.4]		Carbon mineral appeared before S exhausted	(Schaefer et al., 2014)
Flood/serpentinized basalt	~	100, 150	10	Siderite	Siderite occurred in 1 day	(Xiong et al., 2017)
SW Iceland basalt	~7	90	>6	Calcite, magnesite	Seawater was a better carbonate fluid	(Wolff-Boenisch & Galeczka, 2018)
Nevada, USA basalt		[0, 200]	15	Calcite, magnesite, siderite, dolomite and dawsonite	Precipitation occurred	(Sturmer et al., 2019)
Stapafell basaltic glass	<6	50	8	Siderite	PCO ₂ , pH effect products	(Clark et al., 2019)
Deccan basalt	~	[100, 200]	[0.5, 12]	~	Time should be concerned	(Kumar & Shrivastava, 2019)
altered gabbro, ophiolite	~5	40	<0.1	~	Ophiolitic rocks and offshore mafic reservoirs were important	(Marieni et al., 2020)
Nevada basalt, SO ₂		[100, 200]		Magnesite, siderite, dolomite, dawsonite	Adding NO ₂ could help CO ₂ mineralization with SO ₂	(Sturmer et al., 2020)
Madagascar mine powder		[90, 185]	<16.5	~	Ca/Mg-bearing silicates were more reactive	(Gadikota et al., 2020)
GMMAU epidote	[2,11]	25, 100, 200		Calcite	Epidote might help dissolution	(Marieni et al., 2021)
Model	<6	[25, 260]	<0.9 PCO ₂	Pyrite, aluminosilicates, anhydrite	80% H ₂ S mineralized	(Marieni, Voigt, Clark, et al., 2021)
North Atlantic seawater and basaltic glass	~	130	[0.25, 1.6]	Magnesite	Magnesite formed at about 1MPa PCO ₂	(Voigt et al., 2021)

The essential formulas employed in CCUS modeling are the same ones that describe the three-phased flow of oil, gas, and water through porous media in oil fields. The fundamental formulas can also be connected with other physical phenomena, such as those that forecast geo-mechanical effects and geochemical reactions (Ajayi et al., 2019). To explain the multiphase flows and reactions in certain porous media, several complete models have been created. The PHREEQC computer code was frequently used in basalt CCUS modeling research throughout the first decade of the twenty-first century to perform thermodynamic calculations (Marieni, et al., 2021). PHREEQC3, iTOUGH2, STOMP-COMP, EQ3/6, the GAS module of GDA, STOMP-CO₂, TOUGHREACT, and TOUGH3 were added to the basalt CCUS modeling application in the second decade of the twenty-first century. The majority of the information regarding the

solid compositions of sampling materials also served as the foundation for the model simulation.

3.2 Experimental methods

Experimental research is typically conducted using a batch reactor, reaction cell, or flow-through. The duration of laboratory studies is frequently a limiting factor; they typically last a few days or no more than a few years. Optical microscopy, infrared (IR) spectroscopy, electron microprobe scanning electron microscope (SEM), powder X-ray diffractometer (XRD), and mass spectrometry (TRA-MS) are frequently used to analyze the composition of core samples (Schaefer et al., 2011; Gysi & Stefansson, 2012a; Schaefer et al., 2014). The elemental composition of the core samples was analyzed using thermogravimetric analysis (TGA) and total inorganic carbon (TIC) and Total Carbon Analysis (TCA)

and the results were compared (Gadikota et al., 2014). When conducting experiments, it can be challenging to account for the geographical variability of the formation. As a result, the experiments are often spatially constrained and struggle to accurately reflect the actual mine conditions. Experiments are still necessary to comprehend the underlying mechanisms and determinants of mineral carbonation, though.

4. MODELING AND EXPERIMENTAL STUDIES

Based on the above experimental and modeling methods, researchers have carried out a lot of work, some representative work is shown in Table.2.

5. PILOT PROJECTS

5.1 Wallula project

After a seismic study and "drill-and-test" characterization drilling in 2013, 977 metric tons of pure supercritical CO₂ were injected into the Columbia River flood basalts (CRBG) approach to Wallula, Washington, USA (McGrail et al., 2017). In two deep basalt brecciated zones, the supercritical CO₂ was injected, and it was confined in three interflow zones or flowtops that were divided up by high-density flow inner zones beneath primary and secondary caprock confining zones. Then it mostly moved into the very permeable upper flow top zone. With the help of a temperature and pressure sensor on a wire rope and an isotope tracer approach, the movement of CO₂ and the reactions between brine, CO₂, and subterranean basalt were carefully observed (McGrail et al., 2014). Radical geochemical processes were seen during post-injection monitoring in the brine-CO₂-basalt system. Ankerite, which had isotopic fingerprints identical to the injected CO₂, was found in the carbonate nodules from the side-wall cores after injection. This indicates that after 24 months of injection, the injected CO₂ was converted to a stable carbonate mineral (McGrail et al., 2017).

5.2 Carbfix project

Reykjavik Energy was given funding in 2012 to carry out the CarbFix project by Iceland, the Centre National de la Recherche Scientifique Université Paul Sabatier, and The Earth Institute at Columbia University. This project involved conducting a pilot basalt CCUS project close to the Hellishei geothermal power plant in southwest Iceland (Matter et al., 2009). CO₂ (98%) and H₂S (2%) mixture, as well as 230 tons of pure CO₂ from the power plant, were injected into basaltic rocks at a depth of about 500 meters during this experiment. To

dissolve each ton of gas at the pressures and temperatures between 400 and 800 meters below the surface, which constituted the goal depth, around 25 tons of local groundwater were required (Matter et al., 2009). To precisely add ¹⁴C along with the injected CO₂, isotopic tracers and a chemical monitoring technique were used to follow the path of the injected CO₂. Since the CO₂ was dissolved in the water and thus chemically activated, almost 95% of the injected CO₂ was mineralized in just 2 years (Matter et al., 2016). Since it is situated in a deeper and warmer formation than the initial site, the CarbFix2 location allowed for the continuous injection of more CO₂ and H₂S in 2014. It then scaled up twice, separately in 2016 and 2017. In a CO₂-H₂S-dominated power plant exhaust gas stream, almost 97% of the H₂S and 56% of the CO₂ were immediately dissolved into clean water at a scrubbing tower. The CO₂ and H₂S-charged solution was then injected into the target reservoir at 800 m and 250 °C (Aradottir et al., 2015; Gunnarsson et al., 2018; Sigfússon et al., 2018).

6. CONCLUSIONS

CO₂ emissions from factories or Direct Air Capture (DAC) can be solved with the use of carbonate mineralization technology. Basalt is regarded as the perfect rock for its extensive distribution and advantageous characteristics like its high concentration of soluble metallic minerals. However, difficulties including choosing an appropriate basalt reservoir, adjusting precipitation or dissolution to fit reservoir conditions, altering the physical or geochemical characteristics of rock during injection, and the challenging calculation of storage capacity must be carefully taken into account. For instance, an acid diffusion leak could dissolve poisonous metal and subsequently contaminate subsurface water. Rapid mineralization of the basalt reservoir had become insurance against leaks compared to carbonate and sandstone reservoirs, but further field experiments need to be carried out. To be a practical treatment for CCUS, in situ mineral carbonation must overcome the cost issues. The cost presents the most difficulty among those considerations. The financial incentive for CCUS does not currently outweigh the cost of emissions. To fully utilize the potential of mineral carbonation, it will be necessary to close knowledge gaps in the basic processes and streamline the procedure to cut expenses. To determine the most likely paths for reactions in natural systems, the rates at which different rock types react, the impacts of fluid composition, and any potential uses for chemicals or catalysts, more research is necessary. More field

experiments and basalt CCUS-specific computer programs could help the process move faster.

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DECLARATION OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. All authors read and approved the final manuscript.

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