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Permanent Carbon Dioxide Storage into Basalt: The CarbFix Pilot Project, Iceland

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Abstract

The storage of large volumes of industrial CO₂ emissions in deep geological formations is one of the most promising climate mitigation options. The long-term retention time and environmental safety of the CO₂ storage are defined by the interaction of the injected CO₂ with the reservoir fluids and rocks. Finding a storage solution that is long lasting, thermodynamically stable and environmentally benign would be ideal. Storage of CO₂ as solid magnesium or calcium carbonates in basaltic rocks may provide such a long-term and thermodynamically stable solution. Basaltic rocks, which primarily consist of magnesium and calcium silicate minerals, provide alkaline earth metals necessary to form solid carbonates. In nature, the carbonization of basaltic rocks occurs in several well-documented settings, such as in the deep ocean crust, through hydrothermal alteration and through surface weathering. The goal of the CarbFix pilot project is to optimize industrial methods for permanent storage of CO₂ in basaltic rocks. The objective is to study the in-situ mineralization of CO₂ and its long term fate. The project involves the capture and separation of flue gases at the Hellisheidi Geothermal Power Plant, the transportation and injection of the CO₂ gas fully dissolved in water at elevated pressures at a depth between 400 and 800 m, as well as the monitoring and verification of the storage. A comprehensive reservoir characterization study is on-going prior to the CO₂ injection, including soil CO₂ flux measurements, geophysical survey and tracer injection tests. Results from the tracer tests show significant tracer dispersion within the target formation, suggesting large surface area for chemical reactions. The large available reservoir volume and surface area in combination with relatively rapid CO₂-water-rock reactions in basaltic rocks may allow safe and permanent geologic storage of CO₂ on a large scale.

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Keywords: Geologic CO₂ storage; in-situ mineral carbonation; geochemical monitoring and verification

1. Introduction

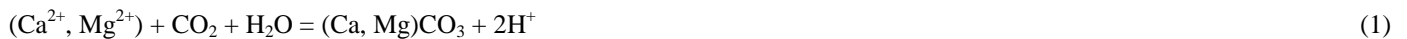
Minimizing climate change impacts from increasing atmospheric CO₂ concentrations calls for dramatic reductions in anthropogenic CO₂ emissions if stabilization is to be accomplished [1-3]. Carbon capture and storage has been proposed as one means to reduce CO₂ emissions from utilizing fossil fuels [4]. The technology involves the capture of CO₂ emissions and the storage in suitable reservoirs. Storage in geologic reservoirs, such as depleted oil and gas reservoirs and deep saline aquifers in sedimentary basins at depths typically >800 m, has been studied extensively over the last decade [4, 5-9]. An alternative storage option is the conversion of CO₂ gas into stable carbonate minerals such as calcite (CaCO₃), dolomite (CaMgCO₃), magnesite (MgCO₃) and siderite (FeCO₃). Mineral carbonation can be carried out *ex-situ* in a processing plant or *in-situ* in silicate-rich geological formations [4, 10]. Basalt or peridotite have been considered as promising reactants for *in-situ* mineralization of CO₂ because of high concentrations of calcium, magnesium silicate minerals in these type of rocks [10-14]. McGrail et al. [11] showed that kilometer-scale interflow zones in the Columbia River Basalt Group lava flows have significant porosity and lateral connectivity for large volume CO₂ injection. In addition, low-permeability interbedded sediments and the impermeable massive basalt between the interflow zones provide a barrier for vertical CO₂ migration. Goldberg et al. [12] demonstrated large storage capacity of sub-oceanic basalt formations. Contrary to the general assumption that fluid-rock reactions are slow, calculations as well as laboratory and small scale field experiments showed relatively rapid CO₂ neutralization and mineralization rates in

basaltic rocks [10,11,13,15,16]. However, *in-situ* mineral carbonation is not without challenges. CO₂ dissolution into groundwater is slow and limits the mineral dissolution and precipitation reactions. Slow reactions require an impermeable cap rock above the injection reservoir to keep the dissolved CO₂ in contact with the reactive rocks and to prevent leakage. Furthermore, mobilization of toxic metals as a result of CO₂-water-rock interaction may lead to their migration within the reservoir. The chemical reactivity of these metals within the reservoir has to be addressed.

Up to date, the potential of CO₂ storage in basaltic rocks have been demonstrated on a limited scale. In an attempt to gain further insight into *in-situ* mineral carbonation in basaltic rocks on a larger scale, the CarbFix pilot project was launched in 2007 [17]. In this paper we discuss *in-situ* mineral carbonation in basaltic rocks and provide a technical and scientific overview of the CarbFix project.

2. *In-situ* mineral carbonation

Mineral carbonation requires divalent metallic cations, such as Ca²⁺, Mg²⁺ and Fe²⁺ for the carbonation process. When CO₂ reacts with Ca, Mg, Fe oxides, the corresponding carbonate is formed and heat is released. These oxides are prevalent in silicate minerals. Given the concentration of the oxides and their reactivity, focus on *in-situ* mineral carbonation has been on rocks rich in olivine, serpentine, pyroxene, plagioclase feldspar and basaltic glass. Crystalline basalt is rich in plagioclase feldspars, pyroxene and olivine, and basalt, glassy and crystalline, contains 7-10 wt% Ca, 5-6 wt% Mg, and 7-13 wt% Fe. After injection of CO₂ in deep aquifers in basaltic rocks following exothermic chemical reactions are taking place:



Groundwater in basaltic rock is rich in divalent metallic cations, which may react with CO₂ to precipitate carbonate minerals according to reaction (1). Per mol of carbonate minerals precipitated, 2 moles of H⁺ ions are produced. This reaction will not proceed to the right unless the H⁺ ions are neutralized. Neutralization of H⁺ will take place by reaction with the primary mineral phases in the basaltic host rock, as simplified in reaction (2) and (3).



The precipitation of carbonate minerals is pH dependent as the acidity of the solution (i.e. H⁺ concentration) controls the carbonate ion concentration in solution. The dissolution of basalt minerals and the release of divalent metallic cations cause the pH of the solution to rise until carbonate precipitation begins. Since the precipitation kinetics of calcite is known to be fast (1), the extent of mineral carbonation is determined by the dissolution kinetics of reaction (2) and (3).

There is evidence for the potential of mineral carbonation in basaltic rocks. There are enormous volumes of basaltic rocks present on Earth's surface with a correspondingly large CO₂ storage capacity. The Columbia River Basalt with a total volume >200,000 km³ has an estimated storage capacity in excess of 100 Gt of CO₂ [11], whereas deep-sea basalt aquifers such as the one on the Juan de Fuca plate could potentially store over 700 Gt of CO₂ [12]. The chemical reactivity and the carbonation potential of basalt is demonstrated by surface weathering processes [16-20], as well as by the fast release rate of Ca and Mg ions when reacted with CO₂-rich waters [13]. The flux of Ca and Mg from basaltic rocks is up to two orders of magnitude larger than from other silicate-rich rocks [21]. Concerning the carbonation of basalt, studies of natural analogues on water-basalt-reactions at low P_{CO2} conditions showed calcite, clays, zeolites and quartz/chalcedony as the dominant secondary minerals [22], whereas quartz and Fe, Mg-, and Ca-carbonates are dominate under high P_{CO2} conditions [23].

3. The CarbFix Project, Iceland

The CarbFix project is a multinational research project with the primary goal to imitate the natural storage process of CO₂ observed in basaltic rocks on a field scale (<http://www.carbfix.com>). The project was established by Icelandic, French and American scientists and was officially launched in 2007. The main partners in the project are Reykjavik Energy, as the main sponsor, University of Iceland, The Earth Institute at Columbia University in New York, and Centre National de la Recherche Scientifique Université Paul Sabatier in France. The project consists of a field-scale CO₂ injection into basalt, laboratory experiments on dissolution and precipitation kinetics, geochemical modeling, and study of natural CO₂-rich waters as a natural analogue.

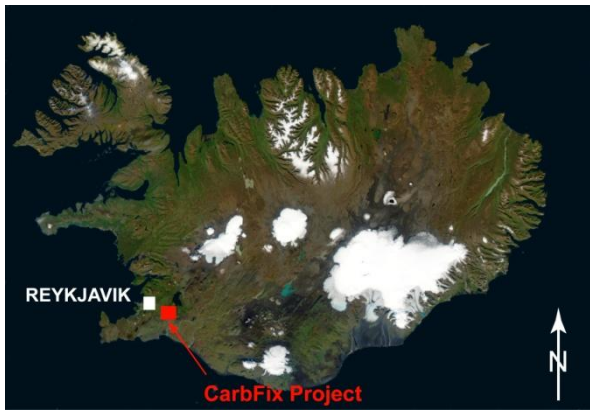


Figure 1: Satellite image of Iceland indicating the location of the CarbFix Site.

The field site is situated in SW Iceland, close to the Hellisheidi geothermal power plant (Figure 1). The geothermal power plant produces up to 30,000 tons of CO₂ per year, although this number will increase as the plant expands. The source of the CO₂ is the geothermal gas which is a by product of the geothermal steam production. The geothermal steam contains around 1% of geothermal gases, which is composed of CO₂ (83%), H₂S (16%) and CH₄, N₂, and H₂ (together 1%). The different geothermal gases will be separated in a pilot gas processing plant, which is under construction. The gases coming from the condenser will be compressed and cooled. The CO₂ and H₂S will be separated and the H₂S will be re-injected with the geothermal water into the deep geothermal reservoir. The CO₂ with 2% of H₂S will be available for the injection into the basalt aquifer. In addition, the H₂ component in the geothermal gas will be separated from the gas mixture by membranes and will be available for the market.

The CO₂ injection site comprises of one injection well with a total depth of 2000 m and several vertical and inclined observation wells with total depths between 100 and 1400 m (Figure 2).

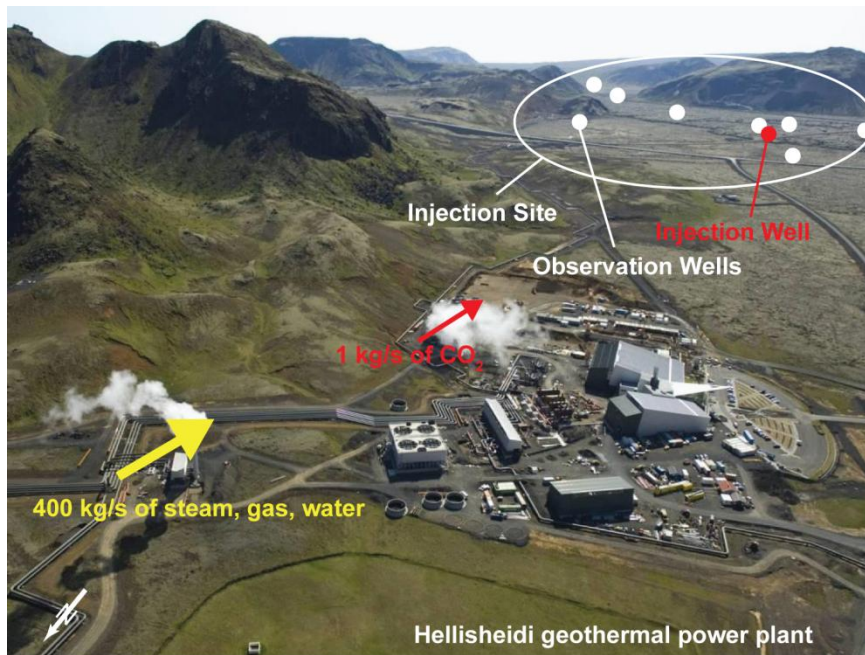


Figure 2: Air photograph from the injection site. The geothermal power plant as the source of the CO₂ is approximately 3 km to the north of the injection site. The injection site includes one injection well and several observation wells.

The target injection formation at a depth of 400 to 800 m consists of basaltic lava flows and hyaloclastite [24]. Figure 3 shows a N-S cross section of the injection site, including the existing borehole infrastructure. The temperature at injection depth interval is between 30°C and 50°C. Some of the observation wells cut into the target injection reservoir, whereas others are shallow wells in the shallow aquifer (Figure 3). Additional observation wells are situated to the North of the injection site. This setup of observation wells will allow the monitoring of the injection reservoir as well as the shallow aquifer along the hydraulic gradient. CO₂ will be injected at a partial pressure of around 25 bars and at a rate of 0.5 to 0.7 kg/s for a time period of 6 months. Since mineral carbonation requires dissolution of CO₂ into an aqueous solution, groundwater will be injected simultaneously with CO₂ at a rate of approximately 20-25 l/s. This will require a large volume of water. Dissolution of one ton of CO₂ at 25 bars partial pressure and 25°C requires approximately 27 tons of water [10]. An alternative would be to inject dry CO₂. In such an injection scenario, mineral carbonation is expected to be slower since the CO₂ has to dissolve into the groundwater to be chemically reactive. Because water availability is not an issue in Iceland, CO₂ and water are simultaneously injected in the CarbFix project with the goal to speed up *in-situ* CO₂ mineralization.

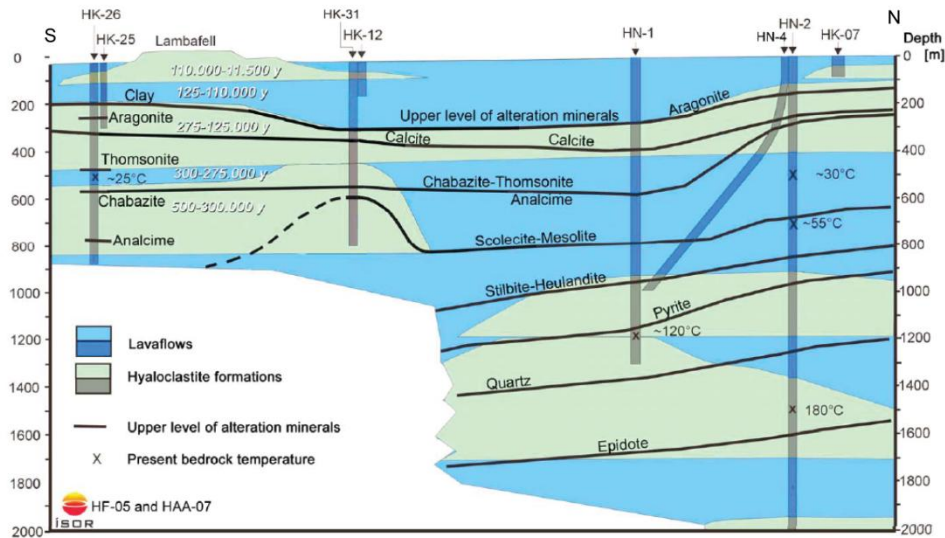


Figure 3: N-S geological cross section of the injection site, including injection well (HN-2) and monitoring wells. The injection interval at a depth between 400-800 m consists of lava flows and hyaloclastite (basaltic glass) formations (Alfredsson et al. [24]).

Prior to the CO₂ injection, which will begin early in 2009, a pre-CO₂ injection field characterization study is ongoing. The goal is to understand the hydrogeology and geology of the target reservoir and the overlying formations. Various methods have been applied since 2007; including soil CO₂ flux measurements, systematic monitoring of groundwater chemistry and borehole geophysical measurements (resistivity, natural gamma, flowmeter, acoustic televiewer). In addition, two tracer tests have been

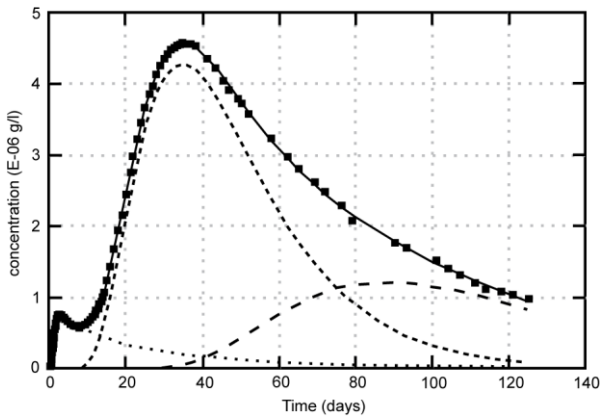


Figure 4: Observed and simulated Na-fluorescein recovery in one observation well. The simulated recovery curves assume three distinct flow paths (modified from Khalilabad et al. [25])

conducted within the injection reservoir. A preliminary tracer test, using Na-fluorescent dye as a conservative tracer, was carried out from November 2007. In addition, a large scale tracer test with sulfur hexafluoride (SF₆) and Na-fluorescent dye is ongoing since June 2008. The preliminary tracer test was conducted as a forced gradient test between two nearby wells, HN-2 and HN-4. A flow field between the two wells was induced by continuous injection of water at a rate of 5 kg/s in well HN-2 and pumping of water at a rate of 10 kg/s from well HN-4 (Figure 3). The distance between the two wells is 60 m at 400 m depth and 360 m at 600 m depth (note that well HN-4 is inclined). According to Khalilabad et al. [25], the analysis and interpretation of the preliminary tracer test revealed that preferential flow paths within the target reservoir exist but that most of the basaltic bedrock consists of relatively homogeneous porous media (Figure 4). Thus, a uniform network of interconnected pore space may provide high tortuosity and large reactive surface area for CO₂-water-rock interaction.

For the large scale tracer test, 50 kg of Na-fluorescent dye and ~14 g or SF₆, each dissolved in 1 m³ of groundwater, were injected as a slug into the injection reservoir to study solute transport under natural groundwater flow conditions. Unlike in the preliminary tracer test, no flow field was induced by pumping. The results of this test will allow the quantification of transport parameters and measurement of subsurface properties, which are important input parameters for the numerical simulations of the CO₂ injection. In addition, potential detection of these tracers in the shallow aquifer will provide information about the hydraulic connection between the injection reservoir and overlying formations.

4. Summary and Perspectives

Understanding the long-term fate of CO₂ injected into geological reservoirs for the purpose of carbon storage is crucial for the success of this carbon mitigation option. Injection of large volumes of CO₂ will greatly perturb the subsurface environment leading to changes in physical, geochemical and biogeochemical parameters. These changes will be the driving force for unknown processes that may enhance or impede permanent storage of CO₂. In-situ mineral carbonation in basaltic rocks offers the potential for permanent and safe storage of CO₂. It, however, requires a better geological and hydrogeological characterization of basalt formations in terms of fluid flow and solute transport, a more accurate quantification of reaction kinetics of mineral and

glass dissolution and mineral carbonation reactions at relevant P/T conditions, as well as the development and testing of monitoring techniques suitable for reactive rock systems. The verification that the injected CO₂ has been fixed in carbonate minerals on a relevant time scale, and that leakage is a minimal risk will be of great importance for the success of in-situ mineral carbonation as an alternative storage option. The CarbFix project addresses these challenges with a multifaceted approach, including a field-scale pilot injection of CO₂ charged waters into a permeable basalt formation, laboratory based experiments, the study of natural CO₂-rich waters as a natural analogue, and reactive transport modeling studies.

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