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## The CarbFix Pilot Project – Storing Carbon Dioxide in Basalt

Juerg M. Matter<sup>a1\*</sup>, W.S. Broecker<sup>a</sup>, S. R. Gislason<sup>b</sup>, E. Gunnlaugsson<sup>c</sup>, E.H. Oelkers<sup>d</sup>,  
M. Stute<sup>a</sup>, H. Sigurdardóttir<sup>c</sup>, A. Stefansson<sup>b</sup>, H.A. Alfreðsson<sup>b</sup>, E.S. Aradóttir<sup>c</sup>, G.  
Axelsson<sup>e</sup>, B. Sigfússon<sup>c</sup>, and D. Wolff-Boenisch<sup>b</sup>

<sup>a</sup>Lamont-Doherty Earth Observatory, Columbia University, 61 Route 9W, Palisades, NY 10964 USA

<sup>b</sup>Institute of Earth Sciences, University of Iceland, Askja, Sturlugata 7, 101 Reykjavik, Iceland

<sup>c</sup>Orkuveita Reykjavíkur (Reykjavik Energy), Baejarhalsi 1, 110 Reykjavik, Iceland

<sup>d</sup>LMTG-Université de Toulouse-CNRS-IRD-OMP, 14 Avenues Edouard Belin, 31400 Toulouse, France

<sup>e</sup>ISOR, Iceland Geosurvey, Grensasvegur 9, 108 Reykjavik, Iceland

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### Abstract

*In situ* mineral carbonation is facilitated by aqueous-phase chemical reactions with dissolved CO<sub>2</sub>. Evidence from the laboratory and the field shows that the limiting factors for *in situ* mineral carbonation are the dissolution rate of CO<sub>2</sub> into the aqueous phase and the release rate of divalent cations from basic silicate minerals. Up to now, pilot CO<sub>2</sub> storage projects and commercial operations have focused on the injection and storage of anthropogenic CO<sub>2</sub> as a supercritical phase in depleted oil and gas reservoirs or deep saline aquifers with limited potential for CO<sub>2</sub> mineralization. The CarbFix Pilot Project will test the feasibility of *in situ* mineral carbonation in basaltic rocks as a way to permanently and safely store CO<sub>2</sub>. The test includes the capture of CO<sub>2</sub> flue gas from the Hellisheidi geothermal power plant and the injection of 2200 tons of CO<sub>2</sub> per year, fully dissolved in water, at the CarbFix pilot injection site in SW Iceland. This paper describes the design of the CO<sub>2</sub> injection test and the novel approach for monitoring and verification of CO<sub>2</sub> mineralization in the subsurface by tagging the injected CO<sub>2</sub> with radiocarbon (<sup>14</sup>C), and using SF<sub>5</sub>CF<sub>3</sub> and amidorhodamine G as conservative tracers to monitor the transport of the injected CO<sub>2</sub> charged water.

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### 1. Introduction

The capture and storage of carbon dioxide and other greenhouse gases in deep geologic formations has to be included in every realistic plan to mitigate greenhouse gas emissions [1, 2]. The standard approach to geologic storage is to inject CO<sub>2</sub> as a bulk phase into geologic formations at depth >800 m. At this depth CO<sub>2</sub> is supercritical and buoyant with respect to the host rocks and aqueous fluids at relevant temperatures and pressures. As a result, buoyant CO<sub>2</sub> may migrate back to the shallow subsurface and surface. Leakage of buoyant CO<sub>2</sub> reduces long-term

\* Corresponding author. Tel.: +1-845-365-8543; fax: +1-845-365-8155.

E-mail address: [jmatter@ldeo.columbia.edu](mailto:jmatter@ldeo.columbia.edu).

storage, and could contaminate shallow groundwater resources if dissolved in groundwater [3]. Thus, the effectiveness of geologic CO<sub>2</sub> storage depends on the retention time, reservoir stability, and the risk of leakage [4, 5].

An alternative storage approach is the mineralization of CO<sub>2</sub> into stable carbonate minerals such as calcite (CaCO<sub>3</sub>), dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>), magnesite (MgCO<sub>3</sub>) and siderite (FeCO<sub>3</sub>). The degree to which mineral carbonation is significant in a storage reservoir and at what rate mineralization occurs depends on rock type, pressure and temperature conditions, and injection methods. Mineral carbonation can be enhanced by injecting CO<sub>2</sub> in silicate rocks rich in divalent cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Fe<sup>2+</sup>) and by injecting CO<sub>2</sub> fully dissolved in water.

In this paper, we will describe the CarbFix Pilot Project in Iceland, which was launched in 2007 and discuss *in situ* mineral carbonation in basaltic rocks [6]. Previous CO<sub>2</sub> pilot projects and existing commercial operations have focused on injecting and storing CO<sub>2</sub> as a separate supercritical phase in depleted oil and gas reservoirs and deep saline aquifers [e.g. 3, 7-10]. The potential for *in situ* mineral carbonation in these reservoirs is limited by the availability of divalent cations from silicate minerals. Hence, the CarbFix project has been created to develop and optimize CCS technology for *in situ* mineral carbonation in basalt.

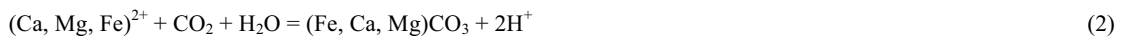
## 2. In situ mineral carbonation

*In situ* mineral carbonation involves the injection of CO<sub>2</sub> into porous rocks in the subsurface, where it can react directly with the host rocks [11]. Rocks containing “basic” silicate minerals with a high proportion of Mg and Ca, such as olivine, serpentine, pyroxene and plagioclase have the greatest mineral carbonation potential because of their high molar proportion of divalent cations [12]. These minerals are primarily found in basalt and mantle peridotite.

After the injection of CO<sub>2</sub> into aquifers, it will dissolve and acidify the formation water and dissociate into bicarbonate and carbonate ions through the following reaction:



Ground waters in basaltic and peridotitic rocks have generally increased concentrations of dissolved divalent cations, such as Ca<sup>2+</sup> and Mg<sup>2+</sup> [e.g. 13-15]. These ions may react with the dissolved CO<sub>2</sub> to precipitate carbonate minerals according to:



Reaction 1 and 2 generate H<sup>+</sup> ions and will not proceed to the right as written unless these ions are also consumed by different reactions. Further water-rock reactions, such as the dissolution of olivine (forsterite) or plagioclase (reactions 3 and 4) consume H<sup>+</sup> ions, resulting in the precipitation of carbonate minerals [e.g. 16-17].



Field and experimental evidence suggest that the rate-limiting step in fixing CO<sub>2</sub> as carbonate minerals is the dissolution of CO<sub>2</sub> into the aqueous phase and the subsequent dissolution of “basic” silicate minerals. In addition to temperature, pressure and salinity, the dissolution of CO<sub>2</sub> into formation waters will depend on buffering pH through fluid-rock reactions. Reactions (3) and (4) buffer pH, thereby shifting Reaction (1) to the right, which results in enhanced CO<sub>2</sub> solubility and mineral carbonation (Reaction 2). Thus, CO<sub>2</sub> storage in pore space can be optimized in host rocks with a high pH buffer capacity, such as basalt and peridotite.

The rate of reactions (3) and (4) are relatively fast compared with other silicate minerals such as Na-, and K-feldspars [18-20]. However, these dissolution rates can be further enhanced, as shown by recent studies. Experiments on carbonation of olivine to form magnesite and quartz in NaCl + NaHCO<sub>3</sub><sup>-</sup> rich aqueous solutions at high pCO<sub>2</sub> and high rock/fluid ratios show reaction rates that are up to 1,000 times faster than dissolution of olivine

at the same pH but without high bicarbonate concentration [21]. Flue gases from power plants contain  $\text{SO}_2$ . Co-injection of this gas with  $\text{CO}_2$  can enhance the dissolution rate of Al-silicates, such as feldspars by complexing the  $\text{Al}^{3+}$  [6]. Furthermore, the crystallinity of the rock also affects the dissolution rate. Basaltic glass releases  $\text{Ca}^{2+}$  two times faster than crystalline basalt [20]. Thus, by choosing the optimal silicate rocks, favorable formation waters with high alkalinity, and by engineering the injection fluid composition, dissolution rates and mineral carbonation rates can be enhanced by several orders of magnitude.

### 3. The CarbFix Pilot Project, Iceland

The CarbFix Pilot Project is the world's first fully integrated CCS project with  $\text{CO}_2$  storage in basaltic rocks. The project has been created to optimize *in situ* mineral carbonation in basalt. It involves a combined program consisting of a  $\text{CO}_2$  pilot gas separation plant,  $\text{CO}_2$  injection pilot test, laboratory-based experiments, studying of natural analogs and numerical modeling. The main partners are Reykjavik Energy in Iceland, University of Iceland, Columbia University in New York and the Centre National de la Recherche Scientifique/Université Paul Sabatier in France.

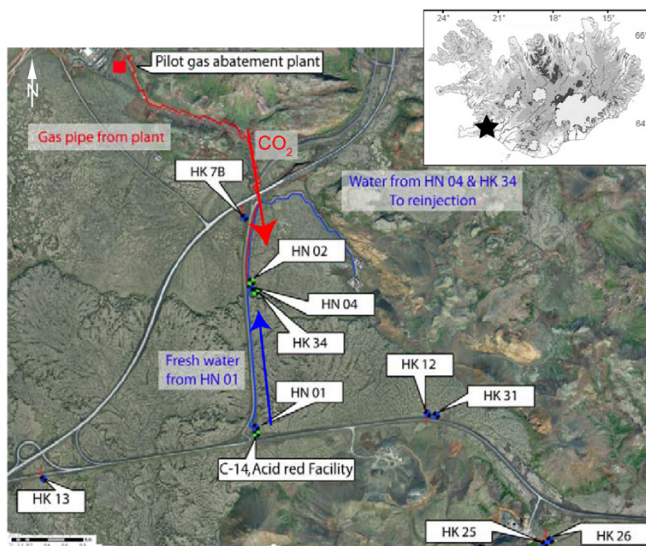


Figure 1. Aerial photograph of the CarbFix injection site at Hellisheiði, SW Iceland.  $\text{CO}_2$  from the gas abatement plant is transported by pipeline to the injection well (HN 02). Fresh groundwater is pumped from well HN 01 and re-injected together with  $\text{CO}_2$  in HN 02 for the purpose of  $\text{CO}_2$  dissolution.  $^{14}\text{C}$  and acid red dye tracers are added to the water stream from HN 01 and  $\text{SF}_5\text{CF}_3$  is added to the  $\text{CO}_2$  gas stream at the pilot gas abatement plant (Source: Reykjavik Energy).

The injection site is situated in SW Iceland, about 3 km south of the Hellisheiði geothermal powerplant (Figure 1). The powerplant currently produces 60,000 tons of  $\text{CO}_2$  per year. The  $\text{CO}_2$  gas is a by-product of the geothermal energy production and is of magmatic origin. The produced geothermal gas mainly consists of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  with minor amounts of  $\text{N}_2$  and  $\text{H}_2$ . The two major flue gases ( $\text{CO}_2$ ,  $\text{H}_2\text{S}$ ) are separated in a pilot gas treatment plant. The separated  $\text{H}_2\text{S}$  is re-injected with the spent geothermal water into the deep geothermal reservoir, whereas the  $\text{CO}_2$  (98%  $\text{CO}_2$ , 2%  $\text{H}_2\text{S}$ ) is transported in a 3 km long pipeline to the  $\text{CO}_2$  injection site. There,  $\text{CO}_2$  is injected fully dissolved in water, resulting in a single fluid phase entering the storage formation. Groundwater for the injection is obtained from well HN 01 that is situated upstream from the injection site (Figure 1).

For this purpose of  $\text{CO}_2$  dissolution, the CarbFix group developed a new injection system that is installed in the injection well, HN 02.  $\text{CO}_2$  at 25 bar and groundwater from HN 01 are injected together. The  $\text{CO}_2$  gas is carried down to a depth of ~500 m by the co-injected groundwater, where it enters the target storage formation fully dissolved. At this conditions,  $p\text{CO}_2$  is 25 bar and the resulting pH is ~3.7. We expect that the  $\text{CO}_2$  charged water will react with basaltic minerals, resulting in a rising pH and increasing alkalinity. The amount of water required to fully

dissolve the CO<sub>2</sub> depends on the temperature and CO<sub>2</sub> partial pressure. Complete CO<sub>2</sub> dissolution requires 22 tons of H<sub>2</sub>O per ton of CO<sub>2</sub> at the CarbFix test site [22].

#### 4. The CarbFix CO<sub>2</sub> injection and monitoring/verification scheme

In a first experiment, which is scheduled to start in Fall 2010, 0.07 kg/s CO<sub>2</sub> dissolved in 2 kg/s of water at 19°C will be injected into basaltic lavas. This results in 2200 tons of CO<sub>2</sub> per year. If storage proves to be successful, the experiment will be upscaled. Figure 2 shows a cross section of the test site with the locations of the injection and monitoring wells. The target storage formation consists of fresh basaltic lavas with interbeds of hyaloclastites, and is of olivine tholeiite composition [23]. The ground waters between 400 and 800 m depth (target storage formation) are at 18–33°C with a pH range from 8.4–9.4 [23]. Permeability is highest between 400 and 500 m and near the base at 800 m depth but decreases drastically below 800 m [24]. A previous tracer test conducted in 2007 and an ongoing tracer test reveal the existence of a dual porosity system with a relatively uniform network of connected porosity [25]. From the surface down to 200 m are fresh basalt flows that host a shallow aquifer, which is separated from the CO<sub>2</sub> injection reservoir by 200 m of low permeability hyaloclastites. The hyaloclastite acts as a confining layer for the injection zone, separating the shallow from the deeper aquifer.

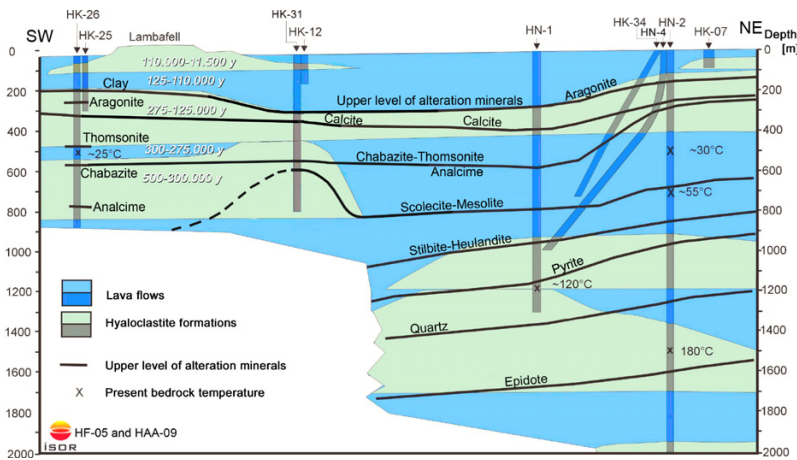


Figure 2. N-S geological cross section at the CarbFix injection site. Injection well (HN 02) and monitoring wells are marked as columns. The target injection reservoir ranges from 400–800 m depth (modified from Alfredsson et al. [23]).

For the purpose of monitoring the CO<sub>2</sub> storage in the target basalt formation, the CarbFix group designed and tailored a monitoring and verification system specifically for *in situ* mineral carbonation. Since CO<sub>2</sub> is fully dissolved in water, standard geophysical monitoring techniques, such as 2-D/3-D seismic surveys or vertical seismic profiling (VSP) cannot be applied. Our approach is to use geochemical monitoring techniques, which are useful for directly monitoring the movement of CO<sub>2</sub> in the subsurface. Conservative tracers, such as trifluoromethylsulphur pentafluoride (SF<sub>5</sub>CF<sub>3</sub>) and acid red dye (amidorhodamine G) will be mixed into the gas and water stream at the CarbFix site to monitor and characterize the physical transport processes of advection and dispersion of the injected fluid (Figure 3). Fluid samples for SF<sub>5</sub>CF<sub>3</sub> and acid red dye will be collected at reservoir conditions in the injection and monitoring wells using a high-pressure bailer system in the injection well and pre-installed submersible pumps in the monitoring wells. The concentration of SF<sub>5</sub>CF<sub>3</sub> and acid red in the retrieved fluid samples will be defined by gas chromatography and fluorescence spectrometry, respectively.

Furthermore, the injected CO<sub>2</sub> will be tagged with radiocarbon (<sup>14</sup>C) by adding <sup>14</sup>C to the water stream (Figure 3). The rationale behind using <sup>14</sup>C is twofold. First, all natural carbon in deep aquifers is generally free of <sup>14</sup>C because of the long residence time of the groundwater and the relatively short half-life of <sup>14</sup>C (5,730 years). Second, <sup>14</sup>C is a reactive tracer, which means that its ratio to carbon in the groundwater will change as a result of dissolution and

precipitation of carbonate minerals. Thus, if mineral carbonation occurs after the injection of CO<sub>2</sub>, it can be monitored and verified by measuring the isotopic composition of reservoir fluid and rock samples with mass spectrometry.

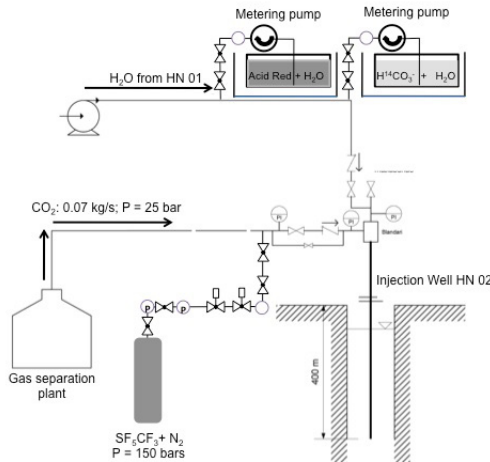


Figure 3. Schematic of the geochemical tracer injection system for monitoring and verifying the transport, reactivity and storage of the injected CO<sub>2</sub>-charged fluid (Source: Reykjavik Energy).

Radiocarbon can be used for verifying and accounting the amount of CO<sub>2</sub> sequestered by changing the isotopic composition of the injected CO<sub>2</sub> from <sup>12</sup>CO<sub>2</sub> to <sup>14</sup>CO<sub>2</sub> [26]. Carbon has two stable, nonradioactive isotopes, <sup>12</sup>C and <sup>13</sup>C, and the radioactive isotope <sup>14</sup>C. The ratio of <sup>13</sup>C/<sup>12</sup>C is 0.01, whereas the ratio of <sup>14</sup>C/<sup>12</sup>C in the atmosphere is 1.3×10<sup>-12</sup>. During the mineral carbonation process, the solid phase will be enriched in the heavier isotopes (<sup>13</sup>C and <sup>14</sup>C) and the remaining dissolved CO<sub>2</sub> will become isotopically lighter. Carbon-14 is only minimally affected by fractionation during phase transitions relative to its precision of measurements [27]. Surface carbon contains ~ 1 part per trillion (ppt) of <sup>14</sup>C. Tagging fossil CO<sub>2</sub> with enough <sup>14</sup>C to make it atmospheric level requires only small amounts of <sup>14</sup>C. A gigaton of fossil CO<sub>2</sub> requires 320 g of pure <sup>14</sup>C. For example, an industrial sequestration site accepting 1000 kg of CO<sub>2</sub> per second would require a daily dose of 0.29 g of <sup>14</sup>CO<sub>2</sub> with an activity of 123 mCi. CarbFix has a license to use up to 20 mCi (7.4×10<sup>8</sup> Bq) of <sup>14</sup>C over 12 months. A stock solution with dissolved H<sup>14</sup>CO<sub>3</sub><sup>-</sup> will be dynamically added to the groundwater pumped from well HN-01 (Figure 3). The resulting <sup>14</sup>C activity in the injected CO<sub>2</sub>-charged water will be 320 PicoCi/L (12 Bq/L). In the subsurface, the injected <sup>14</sup>CO<sub>2</sub> will be diluted by mixing with ambient low <sup>14</sup>C activity groundwater, and the <sup>14</sup>C to carbon ratio will steadily drop toward the pre-injection conditions. It is important to note that at this level of activity the added <sup>14</sup>C does not pose any significant health hazard.

Using this multi-tracer approach with non-reactive and reactive tracers will allow us to separate physical and geochemical processes by examining the change of the ratio between these different tracers. Mixing fractions (i.e. dilution factors) will be defined by the conservative tracers (SF<sub>5</sub>CF<sub>3</sub>, acid red), whereas carbonate dissolution or precipitation reactions will be characterized by the measured <sup>14</sup>C/C and <sup>13</sup>C/C ratios in the groundwater and precipitated carbonates. It is expected that these ratios will change as a result of CO<sub>2</sub>-fluid-rock reactions. If mineral carbonation occurs after the injection of CO<sub>2</sub>, it can be monitored and verified by <sup>14</sup>C because carbonate minerals will contain <sup>14</sup>C from the injected anthropogenic CO<sub>2</sub>. Since the natural dissolved carbon in the storage reservoir is low in <sup>14</sup>C, the <sup>14</sup>C tagged injected CO<sub>2</sub> from the geothermal power plant will be easily identified in fluid and solid samples from the aquifer. For the purpose of verifying mineral carbonation, CarbFix will retrieve small diameter cores from the storage reservoir by wireline core drilling during the post-injection characterization phase.

## Summary

The long-term safety and permanence of geological CO<sub>2</sub> storage will be determined by *in situ* fluid-rock reactions. Injection of CO<sub>2</sub> will perturb the subsurface environment resulting in changes in physical, chemical and biogeochemical parameters. *In-situ* mineral carbonation in basalt offers the potential for permanent CO<sub>2</sub> storage in form of carbonate minerals. However, up to now, the potential of CO<sub>2</sub> storage in basalt has not been demonstrated on a pilot scale. In addition, little is known about geochemical reactions caused by CO<sub>2</sub> injection and *in situ* mineral carbonation rates in basaltic storage reservoirs. Hence, the CarbFix pilot project was launched in 2007 with the goal to test the feasibility of *in situ* mineral carbonation in basaltic rocks at the Hellisheidi site, SW Iceland. The project calls for a 2200 tons per year of CO<sub>2</sub> injection. Since mineral carbonation is facilitated by the dissolution of CO<sub>2</sub> gas into the aqueous phase, CarbFix will inject the CO<sub>2</sub> fully dissolved in water at a rate of 0.07 kg/s. None of the standard approaches for monitoring and verification of geologic CO<sub>2</sub> storage provide a surveying tool for *in situ* mineral carbonation. Therefore, CarbFix tests and implements the tagging of the injected CO<sub>2</sub> with <sup>14</sup>C as a direct method for monitoring geochemical reactions (CO<sub>2</sub> mineralization) and accounting of the stored CO<sub>2</sub>.

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