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A brief history of CarbFix: Challenges and victories of the project's pilot phase

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Abstract

The pilot phase of the CarbFix project ran for over a decade and consisted of the training of students, creating the scientific basis for the fixation of carbon dioxide in the subsurface through the in-situ carbonation of basalts, and the demonstration of this technology by fixing approximately 200 tons of injected CO₂ as carbonate minerals during 2012 and 2013. Over the course of this effort numerous parts of this project have been reported in scientific articles, but a number of challenges including that of separating CO₂ gas from a H₂S-rich effluent gas, the clogging of the original CarbFix injection well and the damage to the project's gas pipe by a third party that eventually shut down the project's pilot phase, have yet to be detailed in the scientific literature. This brief manuscript reviews the CarbFix timeline over the past 12 years, describing in detail some of these challenges. CarbFix demonstrates how interdisciplinary collaboration between the green energy industry, academia, engineers and technicians allows for a fast and efficient development of the idea of battling climate change by permanently petrifying otherwise emitted CO₂ in subsurface basalt formations into an economic industrial scale process useful to the global economy.

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1. Introduction

In February 2005 the Kyoto protocol entered into force committing countries to binding CO₂ emission reductions. During the first commitment period, 37 industrialized countries and the European Community committed to reduce

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greenhouse gas emissions to an average of five percent below their levels in 1990. To address this challenge the Icelandic President approached Einar Gunnlaugsson at Reykjavík Energy, Wally Broecker at Columbia University, Eric Oelkers at CNRS Toulouse (France) and Sigurdur Gíslason at University of Iceland to design a project to aid in limiting the Greenhouse gas emissions in Iceland (Fig. 1).



Fig. 1. Members of the CarbFix scientific steering committee together with the President of Iceland at Hellisheidi Geothermal Power Plant in September 2009. From the left: Dr Einar Gunnlaugsson, Dr Sigurdur Gíslason, President of Iceland Dr Ólafur Ragnar Grímsson, Dr Wally Broecker and Dr Eric Oelkers. *Photo: Sigfús Már Pétursson.*

Carbon storage in Iceland is somewhat challenging due its geology. Iceland is the largest section of the Mid-Atlantic Ridge above sea level, and consists exclusively of basaltic rocks, so that it lacks the sedimentary basins that are commonly thought of as favorable geologic CO₂ storage hosts. Before CarbFix, much of the research team had focused on scientific studies of basaltic rock reactivity in the laboratory and the field [1-4], but not on the challenges associated with capturing CO₂, separating it from other gases typically found in emission streams, acquiring the permissions and funding for a subsurface injection, and sampling deep in the subsurface. Although the larger part of the results from our CarbFix research have been published in various peer review publications, a number of the challenges, decisions, errors, and setbacks made along the way have been thus far unreported. As many of these might also be insightful for others hoping to initiate a carbon capture and storage program, we provide in this manuscript a brief timeline and overview of the CarbFix project since its inception, highlighting some of the challenges that we faced over the past decade.

2. Laying the scientific foundation

Once the CarbFix group was created on the initiative of the Icelandic president, it was incumbent upon us to generate an overall plan and get it approved by our respective institutions. Notably, Reykjavík Energy decided to participate in the project and grant access to its recently commissioned Hellisheidi geothermal power plant, allowing access to a CO₂ source, injection and monitoring wells. This decision was made without any existing regulatory requirements to capture CO₂ and despite the fact that the company was already producing ‘green’ geothermal energy.

After a number of meetings within the CarbFix group starting in 2006, a plan was devised to identify the major tasks, securing funding and characterizing the pilot injection site.

The overall goal of CarbFix was stated by Wally Broecker at a Workshop in Reykjavík on Atmospheric Carbon Capture and Fixation in Basaltic Rocks in January 2006. The goal was manifested at the first CarbFix organization meeting held in October 2006 to create a “demonstration project designed to show the world that CO₂ can be economically removed from the atmosphere and stored in basalt”. As little was known about this possibility at the time, we felt it necessary to both characterize as completely as possible the subsurface system, identify a suite of reactive and non-reactive tracers to unambiguously determine the fate of the injected carbon, and to perform laboratory and modelling studies to predict the basalt-CO₂ interaction prior to the pilot field injection study. The original tasks of the CarbFix projects are summarized in Fig. 2. To address all of these aspects we decided to involve into CarbFix a large number of PhD and MSc students. The involvement of these students held a number of advantages. First, they were relatively inexpensive compared to employing professional scientists, and second through their training in the CarbFix project, it was possible to generate the human capital that would be able to apply the technology developed by CarbFix in the future. A list of the PhD and MSc students and projects completed as a part of CarbFix is presented in Table 1. The generation of a scientifically sound demonstration project required the detailed characterization of the subsurface system including its geology and hydrology. This work started shortly after the beginning of the project; the subsurface geology was defined as part of the PhD thesis of Helgi Alfredsson [25], the hydrology as part of the MSc thesis of Mahnaz Khalilabad [32] and hydrochemical modelling as part of the PhD thesis of Edda SP Aradóttir [10-13].

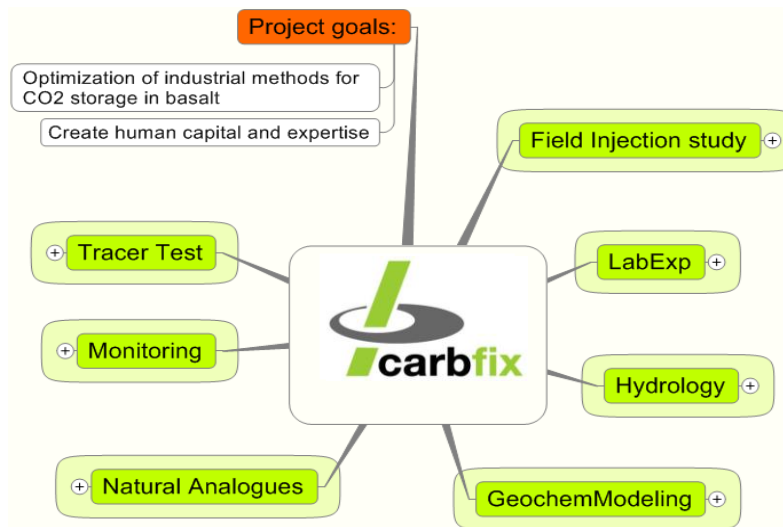


Fig. 2. Objective and work packages of the pilot phase of the CarbFix project.

3. Permits for field study and Public Acceptance

In May 2007 Reykjavík Energy hired Hólmfríður Sigurdardóttir as the CarbFix manager. The manager’s early tasks were to gain acceptance from the authorities and the public, and to obtain permits for the CO₂ injection (Fig. 3). Furthermore, an important task was to foster cooperation between scientists, engineers and mechanics, and to maintain support from top management at Reykjavík Energy as the company was granting the project access to its power plant, field site, and well infrastructures. In the fall 2011 Edda Sif Aradóttir became the CarbFix manager.

Table 1. List of PhD and MSc students/project completed during the pilot phase of CarbFix.

Student	Project title	Years	Published Articles
Therese Flaathen	Water rock interaction during CO ₂ sequestration in basalt	2006-2009	[5], [6]
Alexander Gysi	CO ₂ -water-basalt interactions: Reaction path experiments and numerical modelling.	2007-2010	[7], [8], [9]
Edda Sif Aradóttir	Computational study of chemical changes in Icelandic geothermal areas: Coupling chemical reactions into reservoir models.	2007-2011	[10], [11], [12], [13]
Gabrielle Stockmann	Experimental Study of Basalt Carbonisation	2007-2012	[14], [15], [16], [17]
Iwona Galeczka	Experimental and field studies of basalt-carbon dioxide interaction	2009-2013	[18], [19]
Snorri Guðbrandsson	Experimental weathering rates of aluminium-silicates. Dissolution of crystalline basalt and plagioclase, and precipitation of aluminium rich secondary minerals	2007-2013	[20], [21]
Jonas Olsson	The formation of carbonate minerals and the mobility of heavy metals during water-CO ₂ -mafic rock interactions	2010 - 2014	[22], [23], [24]
Helgi Arnar Alfredsson	Water-rock interaction during mineral carbonation and volcanic ash weathering	2007-2015	[25], [26]
Sandra Snæbjörnsdóttir	Mineral storage of carbon in basaltic rocks	2012-2017	[27], [28], [29], [30]
Deirdre Clark	Mineral storage of carbon in basaltic rocks at elevated temperature. A field and experimental study	2014 - 2019	[31]
Mahnaz Rezvani Khalilabad	Characterization of the Hellisheidi-Threngsli CO ₂ sequestration target aquifer by tracer testing (Masters)	2007-2008	[32]
Diana Fernandez de la Reguera	Monitoring and verification of geologic CO ₂ storage using tracer techniques (Masters)	2008-2010	[33]
Elísabet Vilborg Ragnheiðardóttir	Costs, Profitability and Potential Gains of the CarbFix Program (Masters)	2009-2010	[34]

Regulations pertinent to CarbFix at the beginning of the project were; 1) The Planning Act, 2) The Environmental Impact Assessment Act (EIA), 3) Regulations concerning prevention of groundwater contamination, 4) Health and safety regulations, 5) Law on nature conservation, 6) Radiation safety regulations and from 2009, 7) Regulation of certain substances that increase greenhouse effects, and 8) Directive 2009/31/EC on the geological storage of carbon dioxide in basalts.

At least ten permits were needed prior to the CO₂ injection. Some of the permits needed to be renewed every year. In August 2007, the National Planning Agency decided that the planned CarbFix pilot project with adjacent pipelines and infrastructure was not subject to an Environmental Impact Assessment. In October 2007, the environmental authorities granted permission for initiating the CarbFix project by using a dye tracer (sodium fluorescein, C₂₀H₁₀Na₂O₅) and a gas tracer (trifluoromethylsulphur pentafluoride, SF₅CF₃). In November 2007, the Ministry for the Environment issued a positive statement for the use of another gas tracer (sulfur hexafluoride, SF₆). In fall 2007 and spring 2008, a construction permit was granted by the Municipality of Ölfus for drilling of an additional monitoring well, HK-34, and construction of pipelines and injection equipment along with a permit from the Icelandic Road and Coastal Administration. The environmental authorities granted licenses in 2009 for the CO₂ injection and use of the proposed tracers based on a detailed monitoring plan associated with the injection. The Icelandic Radiation Protection Institute granted a license with conditions for the import and use of radioactive carbon (¹⁴C) as a tracer. On May 11th 2009, the Environmental Agency issued a positive statement for the CO₂ injection and the use of the tracers

trifluoromethylsulphur pentafluoride (SF_5CF_3), amidorhodamine G dye and radiocarbon ^{14}C . On July 16th 2009 the Municipality of Ölfus granted an operation license for the CarbFix pilot injection.

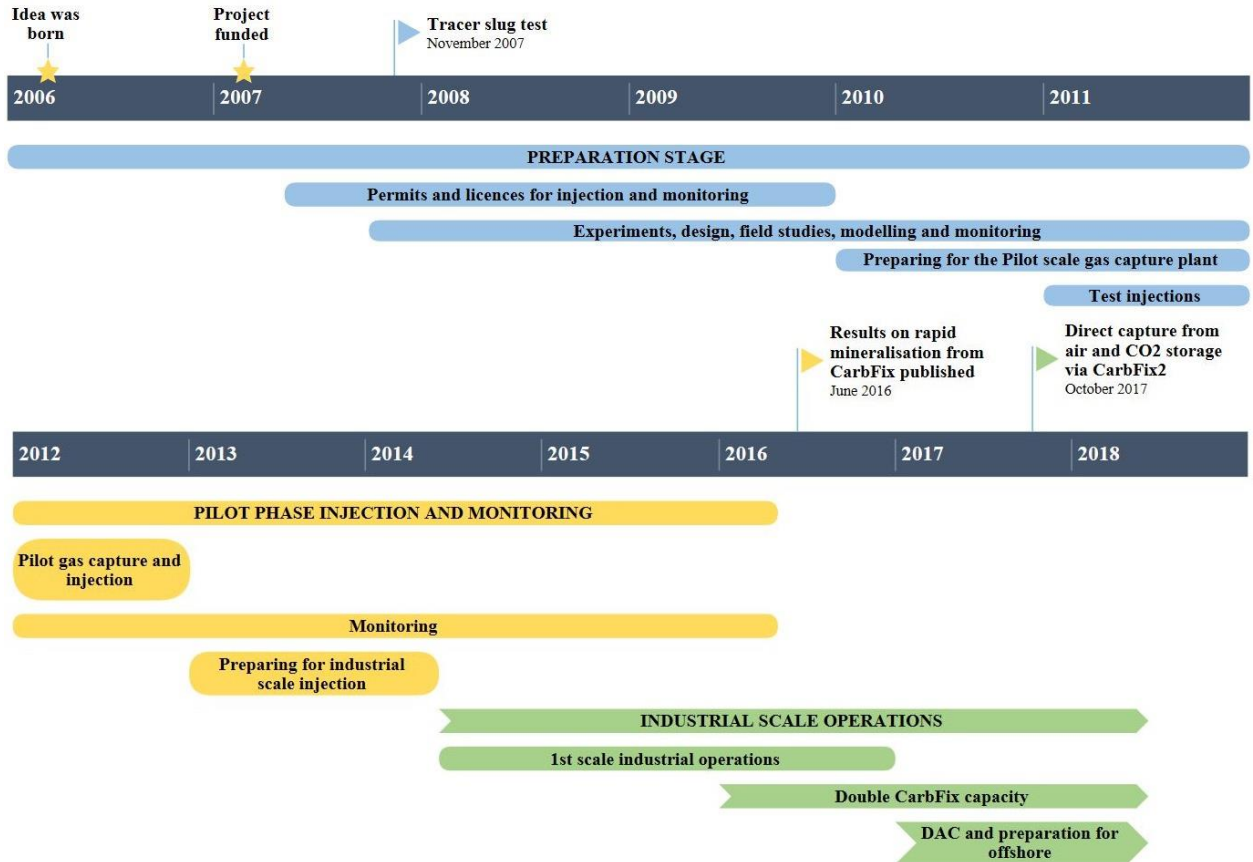


Fig. 3. A brief timeline for CarbFix. In the pilot phase of CarbFix, pure CO_2 and a $\text{CO}_2\text{-H}_2\text{S}$ gas mixture were injected at 350 m depth and 20-50 °C in 2012 (Fig.4). In 2014 a $\text{CO}_2\text{-H}_2\text{S}$ gas mixture was continuously injected below 700 m depth and into a hotter part of the geothermal system (>250°C). The annual capacity of this up-scaled injection was ca. 15,000 tons of $\text{CO}_2\text{-H}_2\text{S}$ gas mixture in 2017. Direct air capture (DAC) started in 2017.

From the beginning, the CarbFix group consulted openly with relevant stakeholders such as the scientific, engineering and environmental sectors as well as non-governmental organizations and the general public. Countless presentations at scientific and non-scientific meetings, school visits, open days at Reykjavík Energy etc. were held. Two books on climate change and the carbon cycle on Earth were published in English and Icelandic [35,36]. Several non-scientific articles were published in the Icelandic and the international press as well as countless local and global TV and radio interviews. Over 60 peer reviewed papers have been published by the CarbFix team. Annual reports have been published on the project website (www.carbfix.com), which includes a description of the project, new developments, list of published scientific papers and presentations, and budget information. All of this helped achieve the public acceptance of the CarbFix project, as well as a strong support of Reykjavík Energy's managers, even during the financial crisis in Iceland that required the company to lay off 25% of its staff in 2010 and bridge a 450 million EUR budget gap during 2011-2016.

4. Capturing the gas

The original CarbFix plan was to capture pure carbon dioxide directly from the gas emissions of the Hellisheidi geothermal power plant. This effort posed particular advantages and disadvantages owing to the composition of the

gas emissions. These gas emissions are approximately 60% CO₂, 20% H₂S, 18% H₂, 2% N₂ by volume and trace amounts of CH₄ and Ar. Although the concentration of CO₂ in this gas mixture is high, facilitating its capture, the presence of the H₂S is problematical as it is highly corrosive and toxic.

In spring 2010 the construction of a pilot gas capture plant was finalised at the Hellisheidi geothermal power plant (Figs. 3 and 4). The pilot station was designed to separate CO₂ from the geothermal gas coming from the condensers of the power plant by sequential extraction. First, a water scrubber was to wash CO₂ and H₂S from less soluble gas species (H₂, N₂, Ar and CH₄). A deaerator was then to remove CO₂ and H₂S from the scrubbing water. The resulting CO₂ and H₂S was to be separated from each other in a distillation column. The H₂S was to be re-injected with spent geothermal water to a deep geothermal reservoir and the CO₂ was to be available for the CarbFix project.



Fig. 4. The pilot gas capture plant, with the scrubber tower (right), deaerator (lowest column in the middle) and the distillation column, the tallest column on the left (Photo: Ingvi Gunnarsson).

In early March 2010 it was discovered that the gas from the condensers in the Hellisheidi power plant contained air and hence oxygen. After thorough examination, necessary amendments were designed and implemented. This incident did however cause delays in the planned CarbFix schedule, as it was not possible to start the pilot gas separation plant until after power plant machinery modifications and repairs in late June 2010.

In July 2010 the pilot gas capture plant started operation with a planned three week testing phase prior to delivery of gas to CarbFix for injection. Experiments with the distillation column commenced and pure CO₂ and H₂S gases were produced. However, after a short period of time, the device that condensed H₂S broke down. In August, September and October there were further delays as more components and equipment broke down almost daily. Therefore, the time plan of delivery of CO₂ for CarbFix and injection of the gas needed constant revision. In late October, samples consisting of 80% clean CO₂ for the CarbFix project had been produced and analysed. However, further breakdowns of gas capture/separation plant continued. It was determined that the origin of the breakdowns was the corrosion of the distillation column by H₂S interaction with its steel components, potentially accelerated by the presence of some water vapour in the distiller. After the destruction of two distillation columns, at a cost of in excess of EUR 250,000, this separation approach was abandoned and it was decided in 2011 to 1) begin the CarbFix subsurface injections using purchased pure CO₂ and 2) switch to injection of a CO₂-H₂S gas mixture following the

pure CO₂ injection. Although the possibility of importing pure CO₂ for the first phase of the injection was considered, this option was discarded, as it would not be readily accepted neither by the CarbFix group nor by the environmental authorities in Iceland. As such, the CarbFix project purchased all available pure CO₂ in Iceland for the first phase of the injection. Excess pure CO₂ was only available in Iceland in January and February, when the demand for carbonated beverages was low. Consequently, 175 tons of beverage grade CO₂ was purchased to start the pilot carbon mineralization study over the winter of 2012 as shown in Fig. 3.

5. Injection system design and testing

The design of an injection system for CarbFix was an integral part of the project from the beginning. The plan was to inject CO₂ charged water into the basalts for two major reasons. First, as CO₂-charged water is denser than fresh water this would limit the risk of the injected gas returning to the surface, and second, as CO₂-charged water is acidic, it would promote the dissolution of basalt, which liberates divalent cations (e.g., Ca²⁺, Mg²⁺ and Fe²⁺) into the solution promoting the eventual precipitation of stable carbonate minerals. Some of these reactions are summarized in Fig. 5. In contrast to basalts, permeable sedimentary basins used for CO₂ injections (Fig. 5a) are mostly composed of quartz (SiO₂), lacking the divalent metal cations needed for mineralization of carbon. The dissolution of CO₂ into water is favored at elevated pressure as prescribed by Henry's law, and the solubility of CO₂ is approximately proportional to its partial pressure. To maximize the efficiency of the dissolution process, it was decided to co-inject CO₂ with sufficient local groundwater so that the gas would be completely dissolved at the depth of its release into the target subsurface basalt aquifer. The greater depth, the higher the pressure and thus the less the amount of water required for the complete dissolution of the injected CO₂. The CO₂ was injected in a separate pipe to a depth of 350 m, where it was released into the down-flowing groundwater as small bubbles. This CO₂ dissolution method was first tested by Diana Fernandez de la Reguera, Martin Stute and Juerg Matter [33] in what we referred to as "the Manhattan Project" since it was performed in the stairwell of one of the tallest building at Barnard College, New York.

The downhole injection system (Fig. 5b) was successfully designed, installed and tested at the CarbFix site by Bergur Sigfusson and co-workers [37]. First tests of the injection system were carried out in March 2011, while problems with separating CO₂ and H₂S were still in full swing at the pilot gas capture station. The CarbFix team convened at Hellisheidi for these first injection attempts, which were intended to calibrate the overall system so that the flow of CO₂ would control the flow of water and tracers to be co-injected with the CO₂. Different tracers were used to label the injected CO₂ so that its journey and fate in the subsurface could be monitored [30,31,38]. When injection tests began, the sparger designed and used to create small CO₂ bubbles allowing for full dissolution of CO₂ within the injection well clogged and nothing could be injected. The same result occurred after the only spare sparger on-site was installed. The interdisciplinary composition of the CarbFix team allowed for an overnight re-design and construction of a modified sparger. This new sparger worked successfully the following day, allowing for calibration and use of the overall injection system.

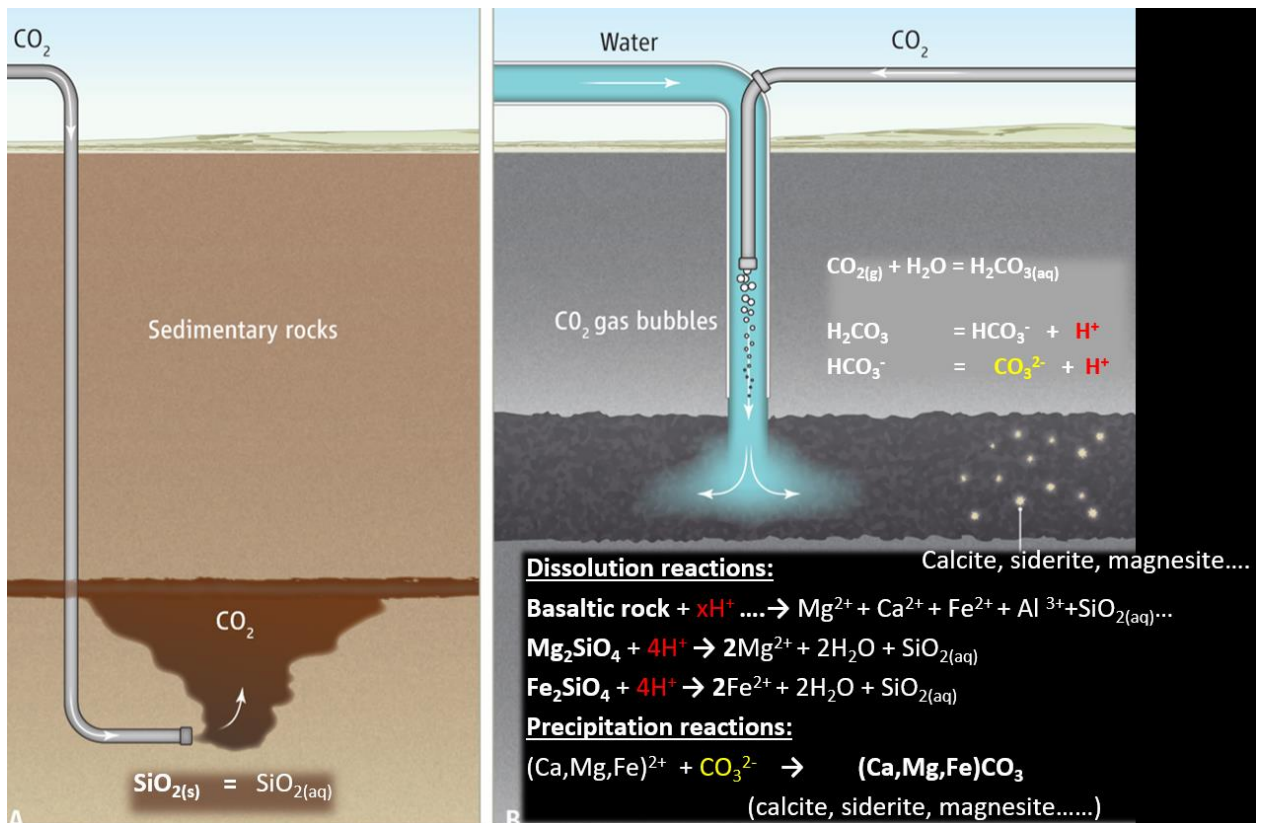


Fig. 5. a) Conventional injection of supercritical CO₂ into sedimentary basin with impermeable cap rock. b). Injection of CO₂-charged water into basaltic rocks at the CarbFix pilot injection site at Hellisheiði, Iceland. Superimposed on a) and b) are the potential dissolution and precipitation reactions occurring after injection, assuming that porous sedimentary basins are dominated by quartz sandstone (modified from [39]).

6. Injection of pure CO₂ and gas mixture, well clogging and the death of the pilot injection

Following its calibration in 2011, the CarbFix injection system successfully dissolved and injected all of the 175 tons of purchased pure CO₂ during January through March 2012 (Fig. 3). Owing to its success, it was decided to continue the injection at the site using the CO₂-H₂S mixture obtained from the pilot gas separation/capture plant prior to the distillation step to separate these two gases, as our attempt to complete this last separation step was unsuccessful (see above). After a pause of approximately one month, to redirect inlet gas lines, the injection of this mixed gas into the original CarbFix well was initiated. Operations of the pilot gas separation station were however not stable and a simultaneously ongoing injection into a deeper system nearby further increased this issue. This resulted in discontinuous injection operations at the CarbFix site, which in turn resulted in decreasing permeability of the injection well. Discontinuous gas injections resulted in pH fluctuations and ensuing supersaturation of iron-sulphite and iron-hydroxide minerals within the injection well. After 3 weeks of further discontinuous mixed gas charged water injection, the permeability of the well dropped so much that the injection needed to be stopped. In total, only 73 additional tons of the mixed gas had been injected.

Several attempts were made to restore the well, first by the addition of dilute acid, and subsequently by the airlifting of the well, a process to remove particles and water from the well and its surroundings by pulling material back out of the well. The airlifting was able to significantly recover the injectivity of the well. Biochemical analysis of the fluids recovered from the airlift by Benedicte Menez and co-workers, suggested the bio-clogging of the subsurface aquifer by iron oxidizing bacteria [40,41].

After a successful airlifting of the CarbFix injection well, the injection of the CO₂-H₂S gas mixture was to commence again in mid-January 2013. However, the CarbFix pilot project took yet another hit. When gas was being

pressurized in the pipe connecting the pilot gas capture plant to the injection well prior to re-starting injection, the gas pipe exploded. It turned out that contractors working on road construction close to the injection site had severely damaged the gas pipe with their heavy machinery during the fall of 2012. This happened in spite of the workers being repeatedly informed of pipeline locations by Reykjavík Energy employees. The damaged part of the gas pipe could not be seen through the visual inspection carried out prior to opening the gas supply as it had been covered with sand and gravel. Several attempts were made to repair the gas pipe, but damage from the explosion turned out to be too severe. It was not considered justifiable to spend a large sum of money to re-construct the line during times that were very difficult financially for Reykjavík Energy. The damage of the CarbFix gas pipe due to nearby road construction therefore killed the continuation of the CarbFix pilot injection.

7. Results from monitoring campaign

Despite needing to shut down the pilot injection phase of CarbFix prematurely, monitoring continued for the next years (Fig. 3). Mass balance calculations, based on the recovery of non-reactive tracers co-injected into the subsurface together with the acid-gases, confirmed that more than 95% of the CO₂ injected into the subsurface was mineralised within a year, and essentially all of the injected H₂S was mineralised within four months of its injection [29,30,38]. Furthermore, carbonates that precipitated on the pump and within pipes in the monitoring well contained the injected radioactive carbon tracer. This proved that carbon dioxide can be sequestered quickly and permanently in basaltic bedrock and thus reduce emissions of this greenhouse gas.

8. Funding and overall cost of the pilot phase of CarbFix

The overall cost of the pilot phase of CarbFix was approximately 12 million EUR. This number includes laboratory studies, pre-injection field hydrology studies, numerical modelling, studies of natural analogs, design and construction of injection and tracer equipment, operation of pilot injections, monitoring and coring. Cost of the pilot gas capture plant and pre-existing well infrastructure, both donated by Reykjavík Energy to the project, are excluded. Funding from national and international research programs in Europe and USA covered about 2/3rd of the cost of the pilot phase of CarbFix or ca. 8 million EUR. Direct financial contribution from Reykjavík Energy covered the remaining ca. 4 million EUR.

CarbFix is a clear example of how active involvement of a strong industrial player ensures the survival of R&D and innovation projects that typically extend over a longer times than individual projects are funded by national or international research programs. The pilot phase of the project ran over 10 years from 2006-2016 (Fig. 3). Although research projects cover the largest part of the total cost, the project would not have survived intermittent times of funding without the continued support of top-level management at Reykjavík Energy. However, the opposite is also true, i.e. CarbFix would not have maintained its support from top-level management at Reykjavík Energy during the extremely challenging times following the economic crisis in 2008 had it not been for a substantial grant from the EU through the FP7 framework program (CarbFix, coordinated action 283148).

9. Conclusions and future prospects

Although original timelines for CarbFix were delayed and changed repeatedly due to the number of challenges described above, CarbFix was upscaled to industrial scale in 2014.

Following the damage beyond repair of the pilot gas transport pipeline in 2013, CarbFix was at a bit of an impasse. Although it was no longer feasible to continue injecting into the pilot CarbFix site there was 1) a need to capture and store H₂S in response to an Icelandic government regulation limiting the emission of this gas, and 2) the impossibility to separate H₂S from CO₂ industrially using distillation. To address these issues, Reykjavík Energy and other members of the CarbFix consortium laid the foundation of industrial scale gas capture and injection at Hellisheidi. As of the summer of 2014, a H₂S-CO₂ mixture has been captured directly by condensate in a water scrubbing tower that was built next to Hellisheidi geothermal power plant (Fig.3). The resulting acid gas charged water is directly injected into a geothermal system having a temperature of more than 250 °C in the target storage basaltic reservoir. It was anticipated that at these high temperatures the risk of aquifer bio-clogging, as experienced during discontinuous injection at the

CarbFix pilot injection site, would be minimized. Results of a subsurface fluid chemical sampling and geochemical calculations show that the majority of the injected H₂S and CO₂ are mineralized as sulphide and carbonate minerals, respectively, within 6 to 12 months of injection. Moreover, results show that the permeability of the new injection well remained steady as the dissolution of host basalt near the injection well dominates the flow properties over the precipitation of secondary minerals further away from the well. These industrial scale CarbFix operations are currently capturing 34% of CO₂ emissions and 68% of H₂S emissions from Hellisheidi power plant [42]. The vision of Reykjavík Energy calls for zero emission of the power plant as soon as possible. Reykjavík Energy has estimated the monetary value of savings from being able to use the CarbFix method as a way to reduce H₂S emissions from Hellisheidi power plant compared to conventional industrial sulphur removal methods was over 100 million EUR through March 2018 [43].

Further application of the CarbFix approach may be best applied for storing CO₂ under the seafloor, which has abundant porous basalts adjacent to an almost inexhaustible supply of seawater [44–46]. Estimates suggest that over 100,000 Gt CO₂ could be stored in the mid-ocean ridges worldwide [27]. Continued efforts, as part of the currently EU funded CarbFix2 project, will be to develop the technology to store CO₂ in submarine basalts by injecting CO₂-charged seawater to the subsurface. The CarbFix2 project (Fig. 3) further involves implementing capture of CO₂ from ambient air (DAC, Direct Air Capture) as a source for subsequent permanent mineral storage within basalts.

The CarbFix project is an example of collaboration between an Icelandic company and universities on both sides of the Atlantic. This was a prerequisite for the development of a promising idea to mitigate climate change into an efficient, automatic process implemented at industrial scale in less than 10 years. Cost of the overall CCS chain being carried out at Hellisheidi power plant has been demonstrated to be two to four times lower than conventional CCS methods [47]. This, along with the fact that captured and injected CO₂ is being permanently mineralized and removed from the atmosphere, could make CarbFix important to the global economy. The degree to which this technology is embraced, however, will depend on the passing of national legislations providing enough incentive or obligations for such solution to be applied at a large or even global scale.

Acknowledgements

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