

## Towards Cleaner Geothermal Energy: Subsurface Sequestration of Sour Gas Emissions from Geothermal Power Plants

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### ABSTRACT

Geothermal energy is a sustainable and clean energy source. Utilization of high-enthalpy geothermal systems is, however, associated with emissions of geothermal gases like CO<sub>2</sub> and H<sub>2</sub>S. H<sub>2</sub>S emissions are currently the main environmental problem associated with high enthalpy geothermal energy utilization in Iceland as the Icelandic government has issued a strict regulation on atmospheric H<sub>2</sub>S concentrations. Reykjavik Energy, the largest geothermal power company in Iceland, is developing innovative methods for capturing CO<sub>2</sub> and H<sub>2</sub>S emissions from its power plants and sequestering the gases as carbonate and sulfide minerals in nearby, subsurface basaltic formations. About 350 tons of CO<sub>2</sub> and H<sub>2</sub>S have been injected into two different storage sites near Hellisheidi geothermal power plant in a pilot capture and sequestration phase. The shallower storage formation lies between 400-800 m depth, is 30-80°C warm and consists of porous, relatively fresh basalts. The deeper storage formation is located below 800 m depth, within the high temperature geothermal system, and consists of fractured, hydrothermally altered basalts with aquifer temperatures around 270°C.

Rather than injecting captured gases directly into the storage formations, a technology to dissolve the gases in water prior to injection has been developed. Once dissolved, the gases are no longer buoyant compared to pore fluids, improving considerable security of the injection due to decreased leakage risks. We have demonstrated that the developed method leads immediate to geological solubility trapping of injected gases. A comprehensive monitoring program based on the use of reactive and conservative tracers has furthermore revealed rapid mineral formation of injected CO<sub>2</sub> within the shallower reservoir and mass balance calculations suggest over 80% mineralization within less than one year. H<sub>2</sub>S mineralization is predicted to be even faster but further monitoring will provide more information on the efficiency of H<sub>2</sub>S mineralization in basalts.

Results obtained so far from the pilot injections are in agreement with results from natural analogs, laboratory experiments and reactive transport model simulations, which all indicate CO<sub>2</sub> and H<sub>2</sub>S mineral sequestration in basalt formations to be a viable option in reducing sour gas emissions from geothermal power plants and thereby bringing high enthalpy geothermal energy closer to zero emission energy production. Plans call for up-scaling sour gas capture and sequestration activities at Hellisheidi geothermal power plant in stages. The first up-scale will commence operation in 2014 and involve capture and re-injection of 8,500 tons of CO<sub>2</sub>-H<sub>2</sub>S gas mixture on an annual basis. Additional up-scales and potential further separation of CO<sub>2</sub> and H<sub>2</sub>S will follow in coming years.

### 1. INTRODUCTION

Geothermal energy is a sustainable and clean energy source. Utilization of high-enthalpy geothermal systems is, however, generally associated with emissions of geothermal gases like carbon dioxide (CO<sub>2</sub>), hydrogen sulfide (H<sub>2</sub>S), hydrogen (H<sub>2</sub>), nitrogen (N<sub>2</sub>), methane (CH<sub>4</sub>), and argon (Ar) to the atmosphere. Emission of some of these gases, particularly H<sub>2</sub>S, remains one of the main environmental concerns associated with high-enthalpy geothermal energy utilization in Iceland. A new regulation regarding atmospheric H<sub>2</sub>S concentration was issued by the Government of Iceland in 2010. The regulation is significantly stricter than the World Health Organization (WHO) Air Quality Guidelines (World Health Organization 2000), as is shown in Table 1, and puts high demands on the geothermal industry in Iceland to reduce atmospheric H<sub>2</sub>S concentrations in the vicinity of geothermal power plants.

**Table 1: Comparison of H<sub>2</sub>S air ambient standards by the World Health Organization (World Health Organization, 2000) and the Icelandic Government.**

WHO Air quality guidelines, 2nd edition		2010 Icelandic regulation on atmospheric H <sub>2</sub> S concentration	
Averaging period	Value (µg/m <sup>3</sup> )	Averaging period	Value (µg/m <sup>3</sup> )
24 h	150	24 h	50
30 min	7 <sup>a</sup>	1 year	5 <sup>b</sup>

<sup>a</sup> Smell nuisance

<sup>b</sup> May be exceeded 5 times annually until July 2014; Reykjavik Energy has requested an extension of that time limit

Reykjavík Energy, the largest geothermal power company in Iceland, is developing innovative methods aimed at capturing CO<sub>2</sub> and H<sub>2</sub>S currently emitted from its power plants and sequestering the gases as minerals in nearby, subsurface, basaltic formations<sup>1</sup>. As a first step towards that goal, three pilot scale CO<sub>2</sub> and H<sub>2</sub>S injections were carried out at Hellisheidi geothermal power plant in 2012 within the SulFix and CarbFix research projects (Aradóttir et al. 2014; Aradóttir et al. 2011; Gislason et al. 2010; Gunnarsson et al. 2013; Gunnarsson et al. 2011; Matter et al. 2011). Hellisheidi geothermal power plant is the largest geothermal power plant in Iceland, currently generating 303MW<sub>e</sub> and 133 MW<sub>th</sub>, with the capacity of expanding thermal production up to 400 MW<sub>th</sub>. The plant annually emits some 40,000 and 16,000 tons of volcanic CO<sub>2</sub> and H<sub>2</sub>S, respectively, as a by-product of the ongoing energy production.

Hellisheidi comprises ideal conditions for studying the long-term effect of mineral CO<sub>2</sub> and H<sub>2</sub>S sequestration in basaltic rocks as: (1) Connection to Hellisheidi power plant provides access to a concentrated source of the gases and (2), field data, such as drill cuttings and a calcite-rich cap-rock overlying the high-temperature reservoir, suggest mineral CO<sub>2</sub> and H<sub>2</sub>S sequestration already to play an important role in the evolution of the Hellisheidi geothermal system. CO<sub>2</sub> sequestration is most intensive from 550-800-m depth below surface, while H<sub>2</sub>S sequestration is most intensive below 800-m depth (Wiese et al. 2008; Nielsson and Franzson 2010; Gebrehiwot et al. 2010; Helgadóttir et al. 2010). Injecting and precipitating CO<sub>2</sub> and H<sub>2</sub>S into nearby formations with the objective of imitating and accelerating natural sequestration processes should therefore be considered as an environmentally benign process.

## 2. SUBSURFACE SEQUESTRATION PROCESSES

Sequestration of captured and injected CO<sub>2</sub> and H<sub>2</sub>S at Hellisheidi occurs through two processes: (1) immediate solubility storage and (2) graduate mineral storage.

### 2.1. Solubility storage

In conventional gas injection associated with e.g. CCS projects, geothermal energy utilization and enhanced oil recovery, gases are injected as a separate buoyant phase into porous subsurface geological formations. Ideally, the designated storage formation is located below an impermeable cap-rock formation to keep the injected, buoyant gas phase from escaping back to the surface. Rather than injecting CO<sub>2</sub> and H<sub>2</sub>S directly into geological formations at Hellisheidi, a technology to dissolve the gases into formation fluids and well water prior to or during injection has been developed by the CarbFix team (Sigfússon et al. in review; Gislason and Oelkers 2014). The method involves dissolution of CO<sub>2</sub> and H<sub>2</sub>S under anoxic conditions through the following reactions<sup>2</sup>:



This approach of immediate solubility trapping improves considerably the injection safety with respect to leaks as injected gases are no longer buoyant compared to pore fluids after their dissolution. All gas injections at Hellisheidi have applied the developed injection method of dissolving gases either prior to or during injection.

### 2.2. Mineral storage

When injection water containing dissolved CO<sub>2</sub> and H<sub>2</sub>S comes in contact with the subsurface primary host rock, dissolution reactions occur, leaching cations such as Ca<sup>2+</sup>, Fe<sup>2+</sup> and Mg<sup>2+</sup>, out of the rock formation. As dissolution reactions proceed, concentration of leached cations builds up and at certain concentration; the formation water becomes supersaturated with respect to secondary minerals like carbonates and sulfides, which begin to precipitate resulting in mineralization of injected CO<sub>2</sub> and H<sub>2</sub>S:



#### CO<sub>2</sub> mineral sequestration

Natural basalt alteration under low and elevated CO<sub>2</sub> conditions along with laboratory experiments of CO<sub>2</sub>-H<sub>2</sub>S-water-basalt interactions provide important insight into which alteration minerals can be expected to form as a result of CO<sub>2</sub> and injections at Hellisheidi. At low CO<sub>2</sub> conditions, CO<sub>2</sub>-water-basalt interaction results in precipitation of simple oxides and oxyhydroxides, clay minerals, zeolites, calcite, quartz and chalcedony. At elevated CO<sub>2</sub> conditions, substantial SiO<sub>2</sub> mineral formation inhibits the uptake of Ca, Mg and Fe into zeolites and clay minerals, and thus facilitates these chemicals to form carbonates. Calcite (CaCO<sub>3</sub>), magnesite-siderite solid solutions ((Mg,Fe)CO<sub>3</sub>) and Ca-Mg-Fe carbonate solid solutions have been reported to form under such conditions (Alfredsson et al. 2013; Aradóttir et al. 2012a; Neuhoﬀ et al. 2006; Gysi and Stefánsson 2012).

<sup>1</sup> Research and development related to CO<sub>2</sub> and H<sub>2</sub>S capture at Hellisheidi is described by Gunnarsson et al. (2015) and Júlíusson et al. (2015).

<sup>2</sup> Water used for dissolving gases is depleted of O<sub>2</sub> so H<sub>2</sub>S is not oxidized to H<sub>2</sub>SO<sub>4</sub> during dissolution.

In view of the above, it is expected that CO<sub>2</sub> mineralization at Hellisheidi will predominantly take place through precipitation of calcite and magnesium-siderite solid solution precipitation. It is possible that clay minerals, zeolites, and potentially also hydroxides will compete with carbonates for divalent cations leached from the primary basaltic rock, although it is not yet clear to what extent these processes will occur.

### H<sub>2</sub>S mineral sequestration

The concentration of hydrogen sulfide in aquifer fluids in Hellisheidi geothermal area has been extensively studied, both as a part of the SulFix project and as a part of general geochemical monitoring of the wells in the area (Gunnarsson et al. 2011; Scott et al. 2011; Stefánsson et al. 2011). The calculated natural H<sub>2</sub>S concentration in the high-temperature aquifer fluids at Hellisheidi is in the range of 15–264 ppm, with an average of 130 ppm. The concentration increases with temperature and appears to be controlled by the following mineral buffer assemblages (Stefánsson et al. (2011)):



where *py* is pyrite (FeS<sub>2</sub>), *pyrr* is pyrrhotite (FeS), *pre* is prehnite (Ca<sub>2</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>10</sub>(OH)<sub>2</sub>), *epi* is epidote (Ca<sub>2</sub>Al<sub>2</sub>FeSi<sub>3</sub>O<sub>12</sub>(OH)), and *mt* is magnetite (Fe<sub>3</sub>O<sub>4</sub>). Injection of H<sub>2</sub>S loaded water will significantly increase the reservoir H<sub>2</sub>S concentration, which in turn is expected to result in precipitation of the minerals shown on the left side of reactions 5 and 6 (see Stefánsson et al. 2011. for more detailed discussion).

## 3. STORAGE RESERVOIRS AT HELLISHEIDI

Figure 1 shows an aerial map of the CarbFix and SulFix injection sites at Hellisheidi. SulFix 1 and CarbFix 1 were used for pilot scale gas injections in 2012, while industrial scale injection will commence at the SulFix/CarbFix 2 site in 2014. Sections 3.1 and 3.2 provide an overview of properties and parameters describing the storage reservoirs at the CarbFix 1 and SulFix 1 sites.

### 3.1. CarbFix 1 storage reservoir

The CarbFix 1 storage formation lies between 400–800-m depth, is 30–80°C warm, and consists of relatively fresh basalts. Fresh flood basalts govern the bedrock from surface down to 200-m depth. These basalts host a cold (5–10°C) groundwater system, with the static water table located about 100 m below surface. Below lays a 200-m thick hyaloclastite layer that separates the cold groundwater system from the storage formation below due to significantly lower vertical permeability. The storage formation extends from about 400 to 800 m depth. It is composed of relatively fresh basaltic lavas interbedded with minor hyaloclastite layers/intrusions. The highest permeability is encountered around 500 m depth. The dominant formation waters at this depth are oxygen poor and have pH ranging from 8.4 to 9.4. (Aradóttir et al. 2011; Alfredsson et al. 2013).

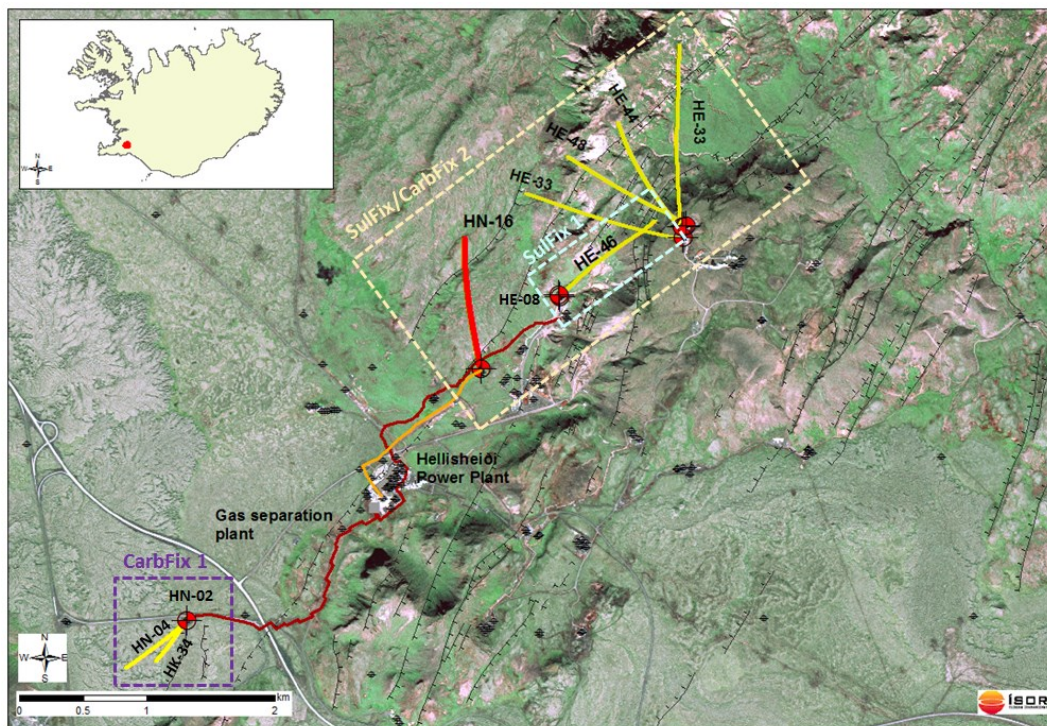
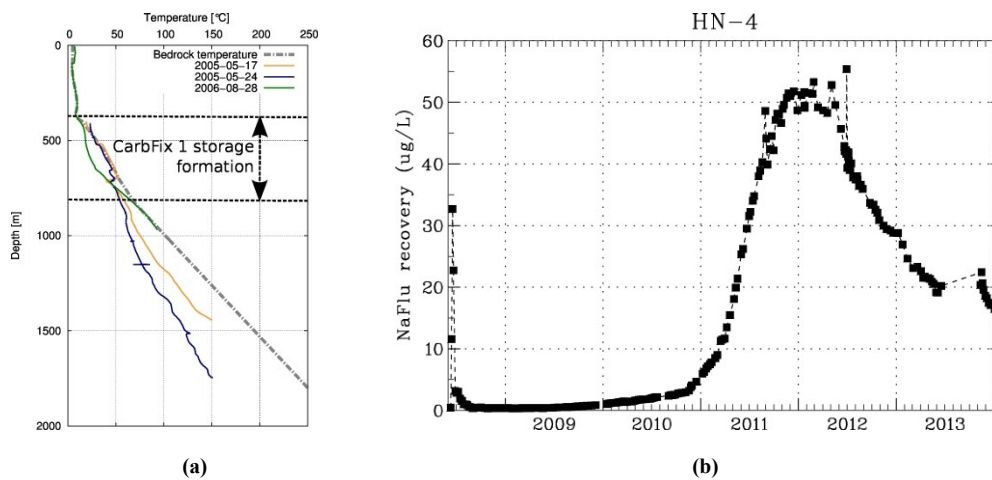


Figure 1: Aerial map of SulFix and CarbFix injection sites in Hellisheidi. Injection sites SulFix 1 and CarbFix 1 were used for pilot scale injections of CO<sub>2</sub> and H<sub>2</sub>S in 2012 (see Table 2 for more detail). Industrial scale injection will be carried out at the SulFix/CarbFix 2 injection site. Injection wells HE-08 and HN-02 (SulFix 1 and CarbFix 1, respectively) are vertically drilled while injection well HN-16 (SulFix/CarbFix 2) is deviated (well deviation shown in bright red). Deviation of main monitoring wells at all injection sites is shown in yellow. Gas pipes from pilot scale gas separation station to SulFix 1 and CarbFix 1 injection sites are shown in dark red, while gas pipe from industrial scale gas separation station to SulFix/CarbFix 2 injection site is shown in orange. Combed lines represent faults.



**Figure 2: (a) Well logs for CarbFix 1 injection well HN-02 and (b) tracer recovery curves from two tracer tests carried out between injection well HN-02 and observation well HN-04. Distance between well feed zones is 70 m and recovery curves indicate slow groundwater flow through a relatively homogeneous, porous matrix.**

Injection well HN-02 at the CarbFix 1 injection site is vertically drilled, 2001 m deep and cased down to 403 m but open to the rock formation below. The main feed zone of the well is located at ca. 500 m depth. Formation temperature at this depth is around 30°C as is shown in Figure 2 (a). Well injectivity of injection well HN-02 prior to gas mixture injection was measured to be 2.0 l/s/bar.

The main monitoring well at the CarbFix 1 injection site is well HN-04, which is 1204 m deep and cased down to 400 m depth. The main feed zone in well HN-04 is located at ca. 420 m depth. Well HN-04 is deviated and its main feed zones are located at ca. 70 m lateral distance away from injection well HN-02.

Two slug type tracer tests have been carried out at the CarbFix 1 injection site since 2007 with the objective of studying regional hydrology in the area and seeing if tracer dispersivity could identify reservoir flow type (fracture or matrix) within the storage formation (see Aradóttir et al. 2011; Aradóttir et al. 2012b). In the first test, 500 g of Na-Fluorescein were injected as a slug into HN-02 on Nov 13<sup>th</sup>, 2007 and a flow field was induced between HN-02 and HN-04 by continuously injecting 6 kg/s into HN-02 and producing 10 kg/s out of HN-04 until May 26<sup>th</sup> 2008. In the second tracer test, 50 kg of Na-Fluorescein were injected as a slug into HN-02 on June 10<sup>th</sup>, 2008. Natural groundwater flow carried the tracer downstream towards well HN-04 in the second tracer test.

Results from the tests indicate the storage formation to consist of a large volume of relatively homogeneous porous media. A low volume and fast breakthrough pathway was found to channel only about 3% of the tracer flow between wells HN-02 and HN-04 (Rezvani Khalilabad et al. 2008), whereas the remaining fluid was found to travel through the relatively homogeneous, porous rock matrix. Figure 2 (b) shows tracer recovery curves in monitoring well HN-04 for tracers injected into well HN-02 in 2007 and 2008.

### 3.2. SulFix 1 storage reservoir

The SulFix 1 injection site is situated below 800 m depth within the Hellisheidi high temperature geothermal system. Static water table in the area is around 200 m below surface. The main rock formation at the injection site is sub-glacially formed hyaloclastite with occasional lava series. Below around 1,400 m below sea level lava series are dominant (Franzson et al. 2010; Helgadóttir et al. 2010). The rock formations at the injection site go through all typical alteration zones of high temperature areas, from fresh rock to epidoteamphibole zone (Helgadóttir et al. 2010). The dominant formation waters at this depth are oxygen depleted and have pH ranging from 6.5-7.5.

Injection well HE-08 at the SulFix 1 injection site is vertically drilled, 2808 m deep and cased down to 932 m but open to the rock formation below. The main feed zones of the well are located between 1300-1345 m depth and at ca. 2210 m depth. Formation temperature at these depths is around 260-270°C, as is shown on Figure 3 (a). Well injectivity of injection well HE-08 prior to gas injection was measured to be 0.72 l/s/bar.

Well HE-46 is the main monitoring well at the SulFix 1 injection site. It is 2744 m deep and cased down to 1032 m depth. The main feed zones in well HE-46 are located at 1100-1170 m depth, 1600 m depth and 2450 m depth. The well is deviated and its main feed zones are located at ca. 450 m lateral distance from the injection well.

A tracer test was carried out at the SulFix 1 injection site in March 2010 in order to identify flow paths of CO<sub>2</sub> and H<sub>2</sub>S rich water injected into HE-08 to nearby wells (Scott et al. 2011; Gunnarsson et al. 2011). 250 kg of Na-benzoate were injected as a slug into HE-08 and 4 kg/s separated geothermal water continuously injected into the well for 56 days. HE-46 and other nearby production wells were discharging at the time of the test. Elevated tracer levels were found in well HE-46 after only two days, revealing a fast flow path between the two wells. This suggests fluid flow within the SulFix 1 reservoir to be fracture dominant. Figure 3 (b) shows tracer recovery curves in monitoring well HE-46 for tracer injected into well HE-08 in March 2010.



Table 2 summarizes reservoir properties of the CarbFix 1 and SulFix 1 pilot injection sites and provides information about their injection wells and main monitoring wells.

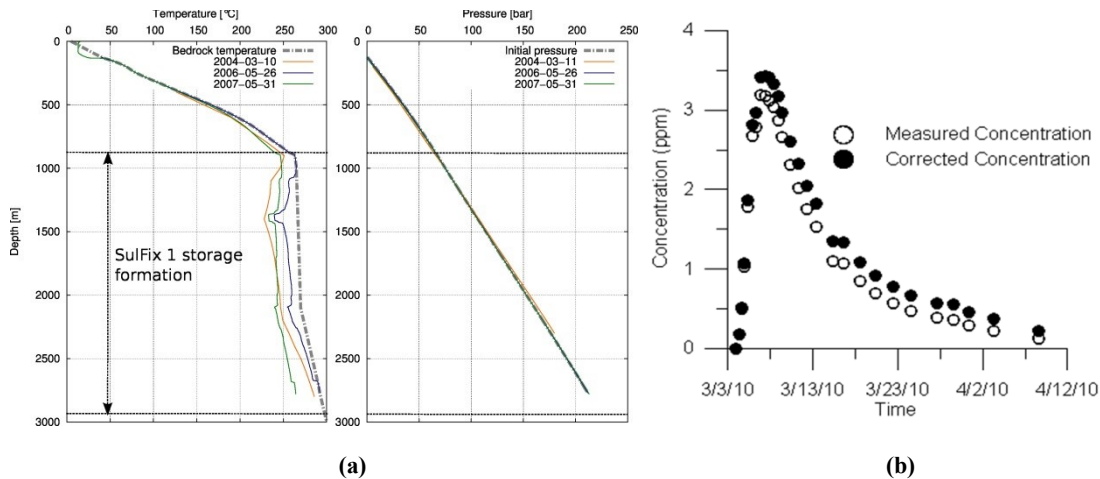


Figure 3: (a) Well logs for SulFix 1 injection well HE-08 and (b) recovery curve from tracer test between HE-08 and observation well HE-46. Distance between feed zones in wells is 450 m and tracer recovery curves indicate fast, fracture dominated flow within the SulFix reservoir.

Table 2: Summary of reservoir properties of the CarbFix 1 and SulFix 1 pilot injection sites and information about injection wells and main monitoring wells.

	CarbFix 1	SulFix 1	
Reservoir properties	Reservoir depth (m)	400-800	> 1000
	Reservoir temperature (°C)	30-60	260-270
	Reservoir pressure (bar)	30-70	> 80
	Flow within reservoir	Matrix dominated	Fracture dominated
Injection well	Well name	HN-02	HE-08
	Well depth (m)	2001	2808
	Depth of casing (m)	403	932
	Depth of main feed zones (m)	500	1300-1345 and 2210
	Injectivity prior to gas injection (l/s/bar)	2.0	0.72
Monitoring well	Well name	HN-04	HE-46
	Well depth (m)	1204	2744
	Depth of casing (m)	400	1032
	Depth of main feedzones (m)	420	1100-1170, 1600 and 2450
	Lateral distance of main feedzone from injection well (m)	70	450

## 4. INJECTIONS

As a first step towards reducing CO<sub>2</sub> and H<sub>2</sub>S emissions from Hellisheidi power plant, three pilot scale CO<sub>2</sub> and H<sub>2</sub>S injections were carried out in 2012 within the SulFix and CarbFix research projects. Overall, about 350 tons of CO<sub>2</sub> and H<sub>2</sub>S were injected into the CarbFix 1 and SulFix 1 storage sites in this pilot capture and sequestration phase.

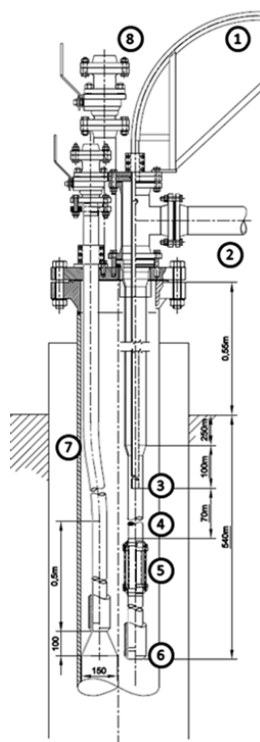
### 4.1. CarbFix 1 pilot injection site

#### C1: Pure CO<sub>2</sub> injection (Jan-Mar 2012)

First injection tests at the CarbFix 1 injection site were carried out in March 2011 with the objective of testing injection equipment. Continuous injection began in January 2012 when 175 tons of commercially bought CO<sub>2</sub> were injected into injection well HN-02 over a 5-week period. CO<sub>2</sub> for the injection was supplied from a 30 m<sup>3</sup> reservoir tank. The pressure in this reservoir tank was maintained at 26-28 bars. Gas flow from the tank was maintained between 55-70 g/s. Co-injected water, used for dissolving CO<sub>2</sub> down well was supplied from an adjacent well. A control valve supplied water to the injection well in a predetermined mass ratio to the CO<sub>2</sub> gas stream (27.7 g water/g CO<sub>2</sub>).

The design of the injection system is shown in Figure 4. Injected CO<sub>2</sub> and H<sub>2</sub>O are injected in separate pipes to a depth of 330 to 360 m. At this depth, CO<sub>2</sub> is released via a sparger into the down-flowing H<sub>2</sub>O, ensuring release of sufficiently small gas bubbles so that they are carried in the water stream to greater depths in the well. The mixture is carried from the sparger via a mixing pipe extending down to 540 m. A static mixer is located at a depth of 420 m to aid CO<sub>2</sub> dissolution. After dissolution, CO<sub>2</sub> concentration in the injection water was around 36,000 ppm and pH of the water was around 3.8.

Fluids injected into the CarbFix 1 storage reservoir during pure CO<sub>2</sub> injection were labeled with tracers to facilitate interpretation of monitoring results. A volatile tracer (SF<sub>6</sub>) was used in order to assess whether degassing is likely to occur within the reservoir. A reactive tracer (<sup>14</sup>C) was furthermore used for determining if and how rapidly injected CO<sub>2</sub> mineralizes within the reservoir (see Matter et al. 2011 for more detail).



**Figure 4:** Design of the CarbFix gas injection system. Natural water level in injection well HN-02 is 80-90 m below the surface. 1) CO<sub>2</sub> injection pipe, 2) water injection pipe, 3) CO<sub>2</sub> sparger located at a depth of 330-360 m, 4) outer mixing pipe extending to a depth of 540 m, 5) Mixer at 420 m, 6) fluid outlet at 540 m, 7) service pipe for downhole sampling and observations, 8) service entry to well head space. (Figure from Sigfússon et al. in review).

The temporal variation of injected CO<sub>2</sub> and H<sub>2</sub>O during the pure CO<sub>2</sub> injection is shown in Figure 5. Overall 175 tons of CO<sub>2</sub> were injected into the subsurface in dissolved form from January 10<sup>th</sup> to March 15<sup>th</sup> 2012. The typical fluid injection rate was 1.8 kg/s.

#### C2: CO<sub>2</sub>-H<sub>2</sub>S gas mixture injection (Jun-Aug 2012)

Injection of a CO<sub>2</sub>-H<sub>2</sub>S gas mixture captured at a pilot gas separation station located next to Hellisheidi geothermal power plant commenced at the CarbFix 1 pilot injection site in June 2012 and was carried out until August 2012. The injected gas mixture consisted of ca. 75% wt. CO<sub>2</sub> and 25% wt. H<sub>2</sub>S<sup>3</sup>. The same type of injection method as described above was used for dissolving the CO<sub>2</sub>-H<sub>2</sub>S gas mixture down well. After dissolution, CO<sub>2</sub> and H<sub>2</sub>S concentrations in the injection water was around 26,000 and 6,800 ppm, respectively. pH of the water was ca. 3.8.

Injected fluids were labeled with a mixture of conservative, volatile and reactive tracers to facilitate interpretation of monitoring results. The tracers used were Amidorhodamine G (conservative), SF<sub>5</sub>CF<sub>3</sub> (volatile) and <sup>14</sup>C (reactive). Tracers were injected at a predetermined ratio to injected water and gas. Co-interpretation of tracer recovery curves and geochemistry of reservoir fluids in monitoring wells allows for quantification of the sequestration efficiency of the storage reservoir (see Matter et al. 2011 for more detail).

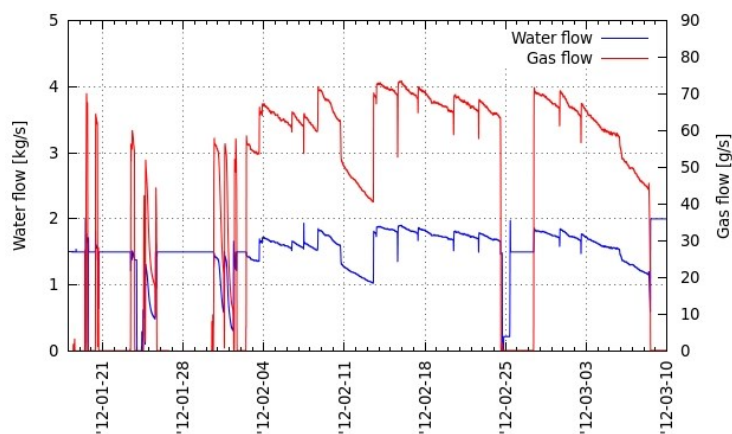
Overall about 60 tons of the gas mixture were injected into well HN-02 between June 12<sup>th</sup> and August 2<sup>nd</sup> 2012. The temporal variation of injected gas mixture and H<sub>2</sub>O during the injection system is shown in Figure 6. The typical fluid injection rate during gas injection was 1-1.5 kg/s.

#### **4.2. SulFix 1 pilot injection site**

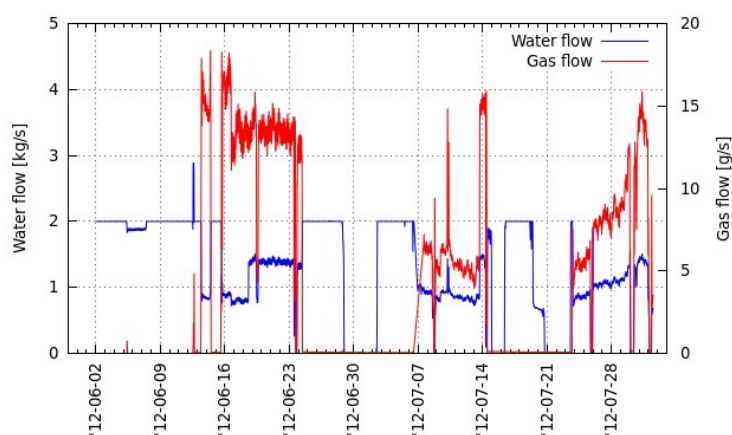
First injection tests at the SulFix 1 pilot injection site were carried out in 2011 with the objective of testing injection equipment. Continuous injection of a gas mixture from the pilot gas separation station at Hellisheidi began in June 2012 and was carried out until December 2012. The gas mixture injected consisted of ca. 65% wt. CO<sub>2</sub> and 35% wt. H<sub>2</sub>S. Unfortunately, operational problems in the pilot gas separation station caused frequent injection stops (see Gunnarsson et al. 2015 for more detail).

Gas injection at the SulFix 1 pilot injection site was carried out applying similar methods and technology as at the CarbFix pilot injection site. In SulFix, however, the gas mixture was dissolved on surface prior to injection, rather than being dissolved down well as in CarbFix. A higher water to gas ratio was furthermore used in SulFix. In the first part of the injection, 95-100°C hot separated water from a nearby high-temperature geothermal well was used for dissolving the gas mixture. 40°C Condensate from Hellisheidi geothermal power plant was used for dissolving the gas mixture in the second part of the pilot injection. Both water types are oxygen depleted.

<sup>3</sup> See Gunnarsson et al. (2015) for more information on gas separation at Hellisheidi.



**Figure 5:** Temporal variation of gas and water flow during injection C1 of 175 tons of pure CO<sub>2</sub> into the CarbFix 1 storage reservoir. Minor operational problems with injection equipment caused fluctuations in the beginning of the injection. Once these were overcome, the injection ran smoothly. Regular flow fluctuations are caused by varying pressure in the tank that provided CO<sub>2</sub> for the injection depending on how full it was. Water injection rate was regulated by gas flow at a predetermined mass ratio.



**Figure 6:** Temporal variation of gas and water flow during injection C2 of 60 tons of 75% wt. CO<sub>2</sub> and 25% wt. H<sub>2</sub>S gas mixture into the CarbFix 1 storage reservoir. Operational problems at the pilot gas separation station at Hellisheidi power plant caused fluctuations in gas flow to the injection site as well as frequent operational stops.

Injection at the SulFix 1 pilot injection site was carried out in such a way that CO<sub>2</sub>-H<sub>2</sub>S gas mixture was released into the water stream at ca. 400 m distance from injection well HE-08 at 8 bar pressure. Three static mixers were located along the length of the pipe that transported the CO<sub>2</sub>-H<sub>2</sub>S-H<sub>2</sub>O mixture to the injection well to aid gas dissolution. Gas and water phases flow separately within the pipe closest to the gas inlet but as the gases dissolve along the length of the pipe, the two phases merge into one, resulting in a single phase fluid being injected into injection well HE-08. After dissolution, CO<sub>2</sub> concentration in injection water is around 350-550 ppm CO<sub>2</sub> and 200-300 ppm H<sub>2</sub>S and pH of the water is ca. 5.

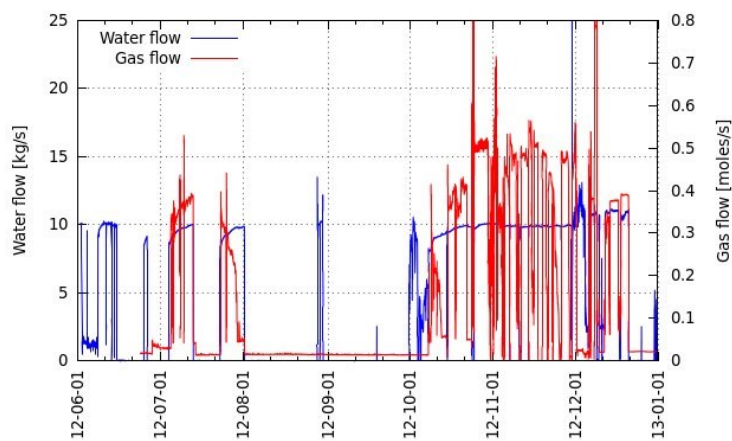
A conservative tracer (Potassium iodide, KI) was co-injected into well HE-08 at a predetermined tracer to gas ratio in order to facilitate interpretation of monitoring results in observation well HE-46.

Overall about 100 tons of the CO<sub>2</sub>-H<sub>2</sub>S gas mixture was injected into well HE-08 between late June and end of December 2012. The temporal variation of injected gas mixture and H<sub>2</sub>O during the injection system is shown in Figure 6. The typical fluid injection rate was 10 kg/s. Separated water was used for dissolving the gases until 11 December. After that, condensate was used for dissolving the gases.

## 5. RESULTS

### 5.1. Solubility trapping

The three pilot scale injections of CO<sub>2</sub> and H<sub>2</sub>S at the CarbFix 1 and SulFix 1 injection sites at Hellisheidi confirmed that the developed method of dissolving the gases during (CarbFix) or prior to (SulFix) injection results in immediate subsurface solubility trapping of the gases. The method thus eradicates leakage risk due to buoyancy which is a common problem when injecting gaseous or supercritical phases into the subsurface as these are less dense than surrounding formation waters, providing a driving force for injected gas to escape back to the surface via fractures or abandoned wells.



**Figure 7: Temporal variation of gas and water flow during injection S1 of 100 tons of 65% wt. CO<sub>2</sub> and 35% wt. H<sub>2</sub>S gas mixture into the SulFix 1 storage reservoir. Operational problems at the pilot gas separation station at Hellisheidi power plant caused fluctuations in gas flow to the injection site as well as frequent operational stops. 95-100°C separated water from a nearby geothermal well was used for dissolving the gas mixture until 10 December. After that, 40°C condensate from Hellisheidi power plant was used for dissolving the gas mixture.**

Verification of the complete dissolution of CO<sub>2</sub> during its injection at the CarbFix 1 injection site was performed by digital downhole camera and by high-pressure well water sampling using a custom made bailer (Alfredsson et al. 2011). Digital images were taken using a downhole camera at depths from 90 to 550 m. These images, such as presented in Figure 8 show the well to be void of gas bubbles consistent with the complete dissolution of CO<sub>2</sub> 1.5 m above the fluid outlet at 540 m as the fluid flows towards the main feed zone at 500 m (Sigfússon et al. in review). Similarly, Figure 9, verifies dissolution of CO<sub>2</sub> and H<sub>2</sub>S prior to injection at the SulFix 1 injection site. The figure shows dissolution of the CO<sub>2</sub>-H<sub>2</sub>S gas phase along the length of the 400 m long pipe that transports the water and gas to the injection well.

## 5.2. Hydrological effects of injections

Figure 10 shows groundwater level fluctuations in injection well HN-02 at the CarbFix 1 injection site during and after pilot scale gas injections in 2012. Changes in groundwater level during the 175 ton pure CO<sub>2</sub> injection at the CarbFix 1 injection site (injection C1 in table Table 3) follow the same trend as changes in injection rate; i.e. when injection rate increases, groundwater level rises but lowers when injection rate is decreased. After gas injection was stopped, the groundwater level remained the same as prior to gas injection (taking seasonal variations into account). The transmissivity of the injection well was therefore not affected by injecting pure CO<sub>2</sub> dissolved in groundwater into the 30-80°C warm, 400-800 m deep storage reservoir at the CarbFix 1 pilot injection site.

Changes in groundwater level in injection well HN-02 during the 60 ton CO<sub>2</sub>-H<sub>2</sub>S gas mixture injection at the CarbFix 1 injection site (injection C2 in table Table 3) show similar behavior as in the pure CO<sub>2</sub> injection, i.e. the water level responds to changes in injection rate. However, after the gas injection was stopped the water level rose quickly by 40 m, or from around 180 m.a.s.l. to around 220 m.a.s.l. In view of this unconventional behavior of groundwater level within well HN-02, the well injectivity was measured. The measurement indicated a significant drop in the well injectivity as it had decreased from  $2.0 \pm 0.1$  l/s/bar to  $0.2 \pm 0.05$  l/s/bar. We were able to increase the injectivity of the well to  $1.0 \pm 0.1$  l/s/bar by carrying out airlift pumping of the well for several hours. Analysis done on fluids discharged from the well and its near vicinity during the airlift pumping indicated sulfide precipitation to have led to clogging of the injection well after injection of acidic water containing dissolved gases was stopped.



**Figure 8: Image taken at a depth of 538.5 m in injection well HN-02 at the CarbFix 1 injection site. The camera enters the injection well through the service pipe (7 in Figure 4). At the time of the photo, CO<sub>2</sub> and H<sub>2</sub>O inlet flows were 27.1 and 750 g/s, respectively. No CO<sub>2</sub> bubbles are visible consistent with the complete dissolution of CO<sub>2</sub> during its injection. (Figure from Sigfússon et al. in review).**



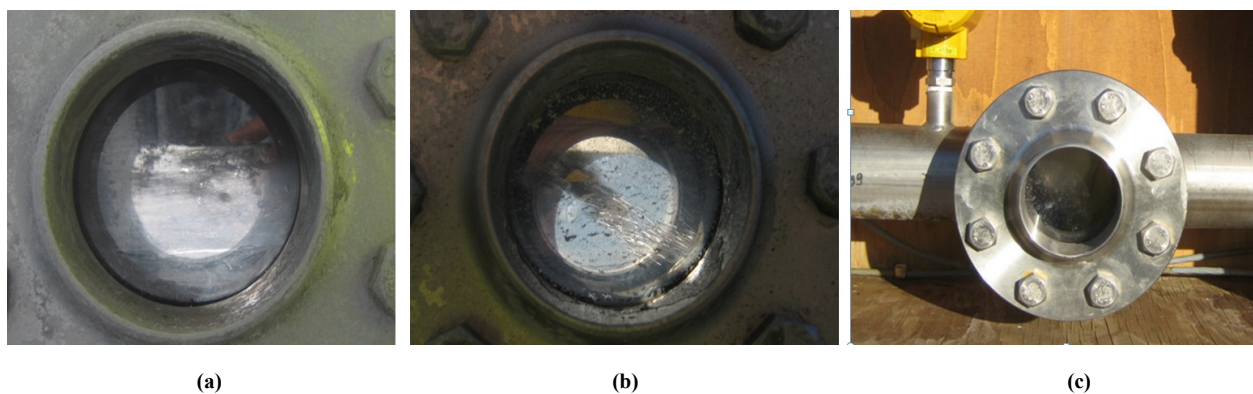


Figure 9: Three observation windows along the length of the 400 m long pipe that transports  $\text{CO}_2\text{-H}_2\text{S-H}_2\text{O}$  mixture towards injection well HE-08 at the SulFix 1 injection site allow us to see how gas gradually dissolves as it flows towards the injection well. Window (a) is located right after the mixing point of water and gas at 400 m distance from the injection well. There, flow of water and gas is still separated with gas flowing on top of the water phase. The second observation window (b) is located a few meters down from the first window, corresponding to flow of ca. 5 seconds. It can clearly be seen that the gas has started dissolving and gas bubbles are travelling within the water phase. The third observation window (c) is located at the wellhead of HE-08 and there, all gas has dissolved resulting in a single fluid phase being injected into the injection well. (Photos: Ingvi Gunnarsson).

SulFix 1 injection well HE-08 was not equipped with downhole data logger so time series of groundwater level fluctuations during and after injection of 100 tons of  $\text{CO}_2\text{-H}_2\text{S}$  gas mixture into the SulFix 1 injection site (injection S1 in table Table 3) are not available. The injectivity of the well was, however, measured prior to and after the gas injection. The measured injectivity prior to injection of dissolved gases into HE-08 was measured to be  $0.75 \pm 0.05$  l/s/bar but  $1.1 \pm 0.1$  l/s/bar after gas injection had stopped. The rise the injectivity constant is significant and indicates near-well clogging not to be an issue when injecting dissolved  $\text{CO}_2\text{-H}_2\text{S}$  gas mixtures back into the high temperature geothermal system.

### 5.3. Chemical effects of injections and mineral trapping

Chemical monitoring of observation wells at the SulFix 1 and CarbFix 1 injection sites is still ongoing as well as co-interpretation of collected data on e.g. tracer recovery curves and geochemistry of storage formation waters. First results from the CarbFix 1 injection site do, however, indicate that more than 80% of  $\text{CO}_2$  injected into the CarbFix 1 injection site has carbonated within a year from its injection (Gíslason and Oelkers, 2014; Gíslason et al. 2013; Matter et al. 2013). A further validation of the rapid mineral trapping efficiency of the CarbFix 1 storage reservoir is continued operational problems of a pump located down hole in observation well HN-04, which are caused by considerable amounts of calcite scaling as a result of  $\text{CO}_2$  injection into the nearby HN-02 injection well, as demonstrated in Figure 11. Indications are that  $\text{H}_2\text{S}$  precipitation is even faster than  $\text{CO}_2$  precipitation, as iron sulfides were clearly detected in water discharged from injection well HN-02 during airlift pumping of the well. This conclusion remains to be confirmed, though.

Results are in agreement with predictive reactive transport modeling of pilot scale injections at the CarbFix 1 and SulFix 1 injection sites (Aradóttir et al. 2014). Simulations predict rapid and efficient mineralization of both  $\text{CO}_2$  and  $\text{H}_2\text{S}$  into thermodynamically stable minerals, with calcite ( $\text{CaCO}_3$ ), magnesite ( $\text{MgCO}_3$ ) and pyrrhotite ( $\text{FeS}$ ) begin the favored carbonate and sulfide minerals to form. Injected gases are predicted to have been mineralized within 1-5 years which is in good agreement with monitoring results obtained so far at the injection sites. At intermediate depths and low temperatures ( $25\text{-}90^\circ\text{C}$ ), calcite is simulated to be the main  $\text{CO}_2$  sequestering carbonate to form, while magnesite is the only carbonate predicted to form at high temperatures ( $>250^\circ\text{C}$ ).

Table 3 summarizes parameters describing the three pilot injections carried out at CarbFix 1 and SulFix 1 injection sites and results obtained so far from the injections.

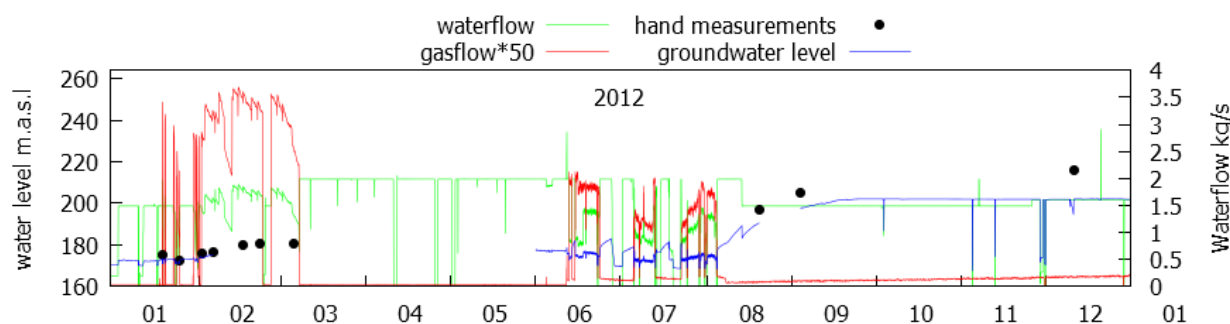


Figure 10: Groundwater level in injection well HN-02 at the CarbFix 1 injection site during and after pilot scale gas injections in 2012. Water level remains about the same after injections of 175 tons of pure  $\text{CO}_2$  (Jan-Mar), indicating that the transmissivity of the well was not affected by the. Water level remains constant during injection of  $\text{CO}_2\text{-H}_2\text{S}$  gas mixture (Jun-Aug) when changes in injection rate are into account. However, after the injection was stopped, water level in HN-02 rose by ca. 40 m (pressure higher than range of data logger explains discrepancies between hand measurements and continuous measurements from Aug to Dec). Transmissivity of the well dropped significantly in concurrence with the rise in water level due to precipitation within the well and its near vicinity.



Figure 11: Calcite precipitation observed on monitoring equipment situated down hole in monitoring well HN-04 at the CarbFix 1 injection site. Left figure shows how the pipe going from the downhole pump and up to the surface is covered in greenish colored precipitation which was measured to be calcite ( $\text{CaCO}_3$ ). The greenish color of the precipitate is believed to stem from remnants from Na-Fluorescein tracer test carried out at the injection site (see Figure 2b). The right figure shows calcite precipitation inside the downhole pump. (Photos: Sandra Ó Snaebjörnsdóttir and Ingvi Gunnarsson).

Table 3: Summary of parameters describing pilot injections carried out at the CarbFix 1 and SulFix 1 injection sites and results obtained so far.

	C1: pure $\text{CO}_2$ injection at CarbFix 1 injection site	C2: $\text{CO}_2$ - $\text{H}_2\text{S}$ gas mixture injection at CarbFix 1 injection site	S1: $\text{CO}_2$ - $\text{H}_2\text{S}$ gas mixture injection at SulFix 1 injection site	
Injection description and parameters	Gas composition	Pure $\text{CO}_2$	75% wt. $\text{CO}_2$ and 25% wt. $\text{H}_2\text{S}$	65% wt. $\text{CO}_2$ and 35% wt. $\text{H}_2\text{S}$
	Amounts injected	175 tons	60 tons	100 tons
	Source of gas	Commercially bought	Pilot gas separation station	Pilot gas separation station
	Time of injection	Jan - Mar 2012	June - Aug 2012	June - Dec 2012
	Continuity of injection	Continuous	Frequent stops due to operational problems in gas separation station	Frequent stops due to operational problems in gas separation station
	Typical water flow during injection (kg/s)	1.8	1.0 - 1.5	10
	Water used for dissolving gases	Oxygen poor groundwater	Oxygen poor groundwater	Separated water or condensate (both oxygen depleted)
	Gas concentration in injection water (ppm)	36,000	26,000 ppm $\text{CO}_2$ and 6,800 ppm $\text{H}_2\text{S}$	350-550 ppm $\text{CO}_2$ and 200-300 ppm $\text{H}_2\text{S}$
	pH of injection water	3.8	3.8	5
	Tracers co-injected	$\text{SF}_6$ (volatile) $^{14}\text{C}$ (reactive)	Amidorhodamine G (conservative) $\text{SF}_6$ (volatile) $^{14}\text{C}$ (reactive)	Potassium iodide (conservative)
Results of injection	Change in groundwater level	No unusual changes observed	40 m rise after injection stopped	Data not available
	Change in injectivity coefficient of injection well	Data not available	Dropped from 2.0 to 0.2 l/s/bar	Increased from 0.75 to 1.1 l/s/bar
	Efficiency of solubility trapping	Instant solubility trapping confirmed	Instant solubility trapping confirmed	Instant solubility trapping confirmed
	Efficiency of mineral trapping	80% mineral trapping within 1 year	Indications of rapid mineral trapping	Indications of rapid mineral trapping

## 6. NEXT STEPS

Pilot  $\text{CO}_2$  and  $\text{H}_2\text{S}$  capture and injection phase finished at Hellisheidi at the end of year 2012. As results of the pilot phase indicated developed methods and technology to provide a viable option in reducing sour gas emissions from Hellisheidi geothermal power plant, it was decided to scale the solutions up to industrial scale gas capture and sequestration in stages. The first upscale will involve designing and building a gas separation station that operates in a similar manner to the pilot gas separation already built at Hellisheidi (see Gunnarsson et al. 2015). The station will use condensate from Hellisheidi power plant for separating  $\text{CO}_2$  and  $\text{H}_2\text{S}$  from less soluble gases, resulting in a  $\text{CO}_2$ - $\text{H}_2\text{S}$ -captured gas mixture. Plans call for starting injection of 400 g/s of the gas mixture dissolved in 60 kg/s condensate into one of the injection wells at the SulFix/CarbFix 2 injection site in Hellisheidi in 2014 (see location of injection site on Figure 1). Following injection of the  $\text{CO}_2$ - $\text{H}_2\text{S}$  gas mixture, a second stage will potentially be added to the new gas separation station, which will further separate  $\text{CO}_2$  and  $\text{H}_2\text{S}$ . The resulting  $\text{H}_2\text{S}$ -rich gas stream will be continued to be dissolved in water and injected back into the geothermal reservoir at Hellisheidi, while plans call for potential utilization of the captured and purified  $\text{CO}_2$ .<sup>4</sup>

## 7. CONCLUSIONS

Reykjavík Energy and collaborators have over the last 7 years put great effort into developing innovative methods and technologies aimed at reducing sour gas emissions from Hellisheidi power plant by capturing and sequestering  $\text{CO}_2$  and  $\text{H}_2\text{S}$  in nearby basalt hosted storage reservoirs. Results obtained in this study show that the approach developed of dissolving the gases during (CarbFix) or prior to (SulFix) injection results in immediate subsurface solubility trapping of the gases. The method thus eradicates leakage risk due to buoyancy which is a common problem when injecting gaseous or supercritical phases into the subsurface as these are less dense than surrounding formation waters, providing a driving force for injected gas to escape back to the surface via fractures or abandoned wells. In addition to reducing leakage risks, indications are that the method also promotes carbon and sulfide precipitation within the host rock and thus facilitating the safe long-term sequestration of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  in the subsurface.

<sup>4</sup> For more information on up-scaled capture and sequestration activities at Hellisheidi, see Gunnarsson et al. (2015) and Júlíusson et al. (2015).

As a first step towards reducing CO<sub>2</sub> and H<sub>2</sub>S emissions from Hellisheidi power plant, three pilot scale CO<sub>2</sub> and H<sub>2</sub>S injections were carried out in 2012 within the SulFix and CarbFix research projects. Overall, about 350 tons of CO<sub>2</sub> and H<sub>2</sub>S were injected into the CarbFix 1 and SulFix 1 injection sites in this pilot capture and sequestration phase. The CarbFix 1 storage formation lies between 400–800-m depth, is 30–80°C warm, and consists of relatively fresh basalts. The dominant formation waters at this depth are oxygen poor and have pH ranging from 8.4 to 9.4. Groundwater flow within the reservoir is matrix dominant with slow flow velocities. The SulFix 1 storage formation is situated below 800 m depth within the Hellisheidi high temperature geothermal system. Formation temperature at these depths is around 260–270°C, formation waters are oxygen depleted and have pH ranging from 6.5–7.5. Groundwater flow within the reservoir is fracture dominant with high flow velocity.

Instant solubility trapping was confirmed at both injection sites and first monitoring results indicate rapid mineral trapping, or over 80% within a year from injection. It is thus concluded from this study that basalts comprise ideal geological CO<sub>2</sub> and H<sub>2</sub>S storage formations.

Shallower and colder storage reservoirs with oxygen poor formations waters and slow, matrix dominant groundwater flow, similar to the CarbFix 1 storage reservoir, were found to be optimal for pure CO<sub>2</sub> injection. Co-injection of H<sub>2</sub>S into such reservoirs might result in clogging of the near vicinity of injection wells as was observed at the CarbFix 1 injection site after injection of CO<sub>2</sub>-H<sub>2</sub>S gas mixture was ceased. H<sub>2</sub>S should rather be disposed of in storage reservoirs similar to the one used in SulFix 1, i.e. fast flowing, fracture dominant reservoirs with oxygen depleted formation waters.

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