PROCEEDINGS, TOUGH Symposium 2012
Lawrence Berkeley National Laboratory, Berkeley, California, September 17-19, 2012

# TOWARDS CLEANER GEOTHERMAL ENERGY UTILIZATION: CAPTURING AND SEQUESTERING $CO_2$ AND $H_2S$ EMISSIONS FROM GEOTHERMAL POWER PLANTS

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## ABSTRACT

Field-scale reactive transport models of $CO_2$ and $H_2S$ mineral sequestration are in development, with a focus on Reykjavík Energy's ongoing gas reinjection tests at Hellisheidi geothermal power plant. A new thermodynamic dataset describing the minerals of interest was developed as a part of this study, since widely used thermodynamic databases did not contain the mineral assemblage needed for the simulations.

Simulations predict efficient precipitation of both $CO_2$ and $H_2S$ into thermodynamically stable minerals, with calcite and pyrrhotite being the favored carbonate and sulfide sequestering minerals, respectively. Despite only being indicative, we conclude from this study that the capture and sequestration of $CO_2$ and $H_2S$ from geothermal power plants is a viable option for reducing their gas emissions, and that basalts may comprise ideal geological $CO_2$ and $H_2S$ storage formations.

## INTRODUCTION

Geothermal energy is a sustainable and clean energy source. Its utilization is, however, generally associated with emissions of geothermal gases, like carbon dioxide ($CO_2$) and hydrogen sulfide ($H_2S$), to the atmosphere. These emissions remain one of the main environmental concerns of geothermal energy utilization.

$CO_2$ is the most abundantly emitted greenhouse gas, and reduction of its anthropogenic emissions is among the great challenges of this century (e.g., Broecker, 2007; Oelkers and Cole, 2008). $CO_2$ emissions from geothermal energy are of volcanic origin and, according to the International Energy Agency, IEA, they are only about 5–6% of the $CO_2$ emissions from a fossil-fuel-burning power plant of a comparable size. It is, nevertheless, important to aim at reducing $CO_2$ emissions from geothermal power plants, so that geothermal energy utilization continues to be considered environmentally benign and friendly.

$H_2S$ is a colorless, flammable gas with the characteristic odor of rotten eggs at low concentrations, while being toxic at high concentrations. $H_2S$ concentration in geothermal fluids is usually in the range of few ppb to several hundred ppm (Arnórsson, 1995a; 1995b). A new regulation regarding atmospheric $H_2S$ concentration was issued by the Government of Iceland in 2010 and will become valid in 2014. The regulation is significantly stricter than the WHO (World Health Organization) Air Quality Guidelines (WHO, 2000), as is shown in Table 1, and puts high demands on the geothermal industry in Iceland to reduce atmospheric $H_2S$ concentrations in the vicinity of geothermal power plants.

***Table 1*** Comparison of $H_2S$ in ambient air standards by WHO and the Icelandic Government.

| ***WHO Air quality guidelines, 2nd Edition :*** | | |
|---|---|---|
| | **Averaging period** | **Value (µg/m$^3$)** |
| | 24 hour | 150 |
| | 30 minute* | 7[a] |
| ***2010 Icelandic Regulation on atmospheric $H_2S$ concentration:*** | | |
| | **Averaging period** | **Value (µg/m$^3$)** |
| | 24 hour** | 50[b] |
| | 1 year | 5 |

[a] Smell nuisance
[b] May be exceeded 5 times annually until July 2014

Reykjavík Energy, the largest geothermal power company in Iceland, is developing innovative methods aimed at capturing the $CO_2$ and $H_2S$ currently emitted from its power plants and sequestering the gases as minerals in nearby basaltic formations (Aradóttir et al., 2011 and Gunnarsson et al., 2011). Pilot scale capture and injection tests have already commenced at the Hellisheidi geothermal power plant in southwest Iceland, as a first step towards that goal.

The current paper provides an overview of ongoing $CO_2$ and $H_2S$ sequestration activities at Reykjavík Energy. Reactive transport models that have been developed of pilot sequestration tests are furthermore discussed, along with results from predictive reactive transport simulations.

## CARBFIX AND SULFIX

The Hellisheidi geothermal power plant generates 303 $MW_e$ and 133 $MW_{th}$, and annually emits some 40,000 and 16,000 tons of volcanic $CO_2$ and $H_2S$, respectively, as a by-product of the ongoing energy production. Three experimental pilot projects were commissioned in 2007–2009 as a first step towards minimizing these emissions.

Geothermal gas from the Hellisheiði geothermal field consists primarily of $CO_2$, $H_2S$, and $H_2$, and to a lesser degree of $N_2$, $CH_4$, and Ar. A pilot gas separation station has been built next to the power plant. The station scrubs less soluble gases ($H_2$, $N_2$, $CH_4$ and Ar) from the more soluble $CO_2$ and $H_2S$. Approximately 3% of the total geothermal gas coming from the power station is currently being separated this way. The gas stream coming from the station consists of approximately 75% $CO_2$, 24% $H_2S$ and 1% $H_2$. Two pilot injection projects, CarbFix and SulFix, will use the gas stream for studying the feasibility of sequestering $CO_2$ and $H_2S$ into minerals within the subsurface.

CarbFix (e.g., Aradóttir et al., 2011; Gíslason et al., 2010; and references therein) is a combined industrial/academic research project between Reykjavík Energy, the Institute of Earth Science at the University of Iceland, Earth Institute-Lamont-Doherty Earth Observatory at Columbia University in New York and the Centre National de la Recherche Scientifique/Universite Paul Sabatier in Toulouse, that was developed in order to assess the feasibility of *in situ* $CO_2$ mineral sequestration in basaltic rocks. The project involves a pilot injection of at least 1200 tons of the gas mixture coming from the pilot gas separation station.

The CarbFix storage formation lies between 400-800 m depth, is 30-80°C in temperature, and consists of relatively fresh basalts (Aradóttir et al. 2011, Alfredsson et al., 2008). Rather than injecting $CO_2$ directly into geological formations, CarbFix has developed a technology to dissolve $CO_2$ into formation fluids and well water during injection. Once dissolved, $CO_2$ is no longer buoyant compared to pore fluids, minimizing considerably the risk from leakage. This solubility-trapping approach also promotes carbonation of the host rock and thus facilitates the safe long-term sequestration of $CO_2$ in the subsurface. The dominant aquifer in injection well HN-02 is at 500 m depth, so most of the injected carbonated water should enter the storage reservoir at that depth.

The SulFix project aims as assessing the feasibility of *in situ* $H_2S$ mineral sequestration in basaltic rocks by applying similar methods and technology as CarbFix. The main difference between the projects is that in SulFix, dissolved gases are injected back into the high temperature geothermal reservoir (below 800 m depth), whereas in CarbFix, it is being injected above this level. Furthermore, hot geothermal water (90°C) is used for dissolving gases in SulFix, while relatively fresh, 30°C warm groundwater is used in CarbFix. Aquifer temperature in the SulFix storage formation is around 270°C, and the dominant aquifer in injection well HE-08 is at 1350 m depth.

Different reservoir conditions in SulFix and CarbFix are emphasized in Figure 1, which shows well-logs for SulFix injection well HE-08 and CarbFix injection well HN-02.

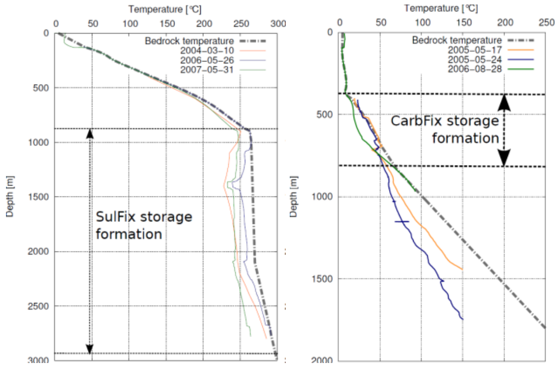


***Figure 1.*** Temperature well logs from SulFix injection well HE-08 (left) and CarbFix injection well HN-02 (right). The SulFix storage formation lies within the Hengill high temperature geothermal system, whereas the CarbFix storage formation lies above it.

Figure 2 shows an aerial map of the CarbFix and SulFix injection sites in Hellisheidi. The well fields shown in Figure 2 comprise ideal conditions for studying the long-term effect of mineral $CO_2$ and $H_2S$ sequestration in basaltic rocks. Connection to the Hellisheidi power plant provides access to a concentrated source of the gases. Additionally, field data, such as drill cuttings and a calcite cap rock overlying the high-temperature reservoir, suggests mineral $CO_2$ and $H_2S$ sequestration already playing an important role in the evolution of the Hellisheidi geothermal system (Níelsson, 2010, Gebrehiwot, 2010, and Helgadóttir, 2010). $CO_2$ sequestration is most intensive from 550 to 800 m depth below surface, while $H_2S$ sequestration is most intensive below 800 m depth. Injecting and precipitating $CO_2$ and $H_2S$ into nearby formations with the objective of imitating and accelerating natural sequestration processes should therefore be considered an environmentally benign process.

Injection of otherwise emitted $CO_2$ and $H_2S$ commenced in 2012 and Hellisheidi power plant became consequently one of the very few worldwide power plants that operate a full CCS cycle.

## REACTIVE TRANSPORT MODELS

Numerical modeling plays an important role in the CarbFix and SulFix projects, because it provides tools for predicting and optimizing long-term management of injection sites, as well as quantifying the amount of $CO_2$ and $H_2S$ that can be mineralized. The following subsections describe reactive transport models developed for the CarbFix and SulFix experiments.

### CarbFix model

Two and three-dimensional reactive transport models have been developed of the CarbFix pilot injection. Aradóttir et al. (2012a) describe in detail conceptual and numerical models of the CarbFix injection site; a short summary is given below.

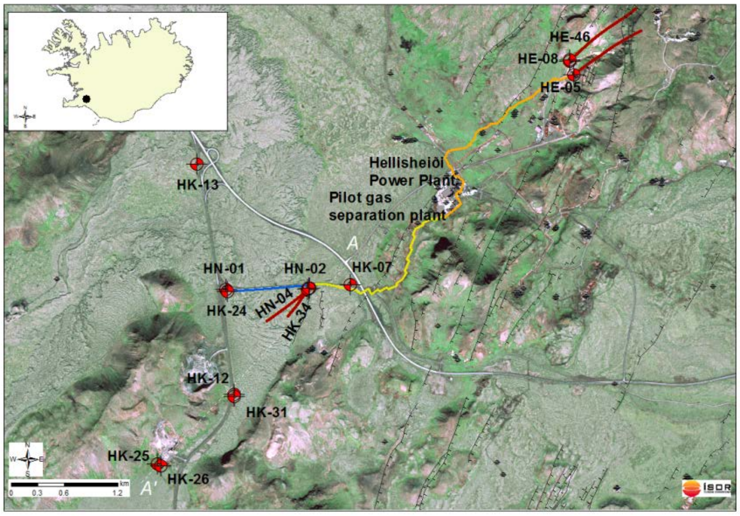


***Figure 2.*** Aerial map of the CarbFix and SulFix injection sites. Gas mixture consisting of 75% $CO_2$, 24% $H_2S$ and 1% $H_2$ will be transported in a pipeline towards the CarbFix (yellow line) and SulFix (orange line) injection sites. HN-02 and HE-08 are CarbFix and Sulfix injection wells, respectively. Other wells are observation wells. Fresh groundwater, used for dissolving gases in CarbFix, will be pumped from well HN-01 and transported to well HN-02 (blue line). Separated geothermal water from HE-05 will be used for dissolving gases in SulFix. Well deviation is depicted by red lines. Combed lines represent faults.

### *Flow setup*

Tracer tests that have been ongoing at the CarbFix injection site since 2007 show that most of the storage formation consists of a relatively homogeneous porous media. A low volume and fast breakthrough path channels only about 3% of the tracer flow between injection well HN-02 and observation well HN-04. Fracture flow thus plays a minor role in the storage formation. In view of this, it was decided to develop the CarbFix numerical models as single porosity models representing the dominant matrix flow in the storage formation.

Models were aligned in the direction of regional groundwater flow, from NE-SW. Hydrological properties of the storage formation were calibrated against a large set of field data using iTOUGH2. Resulting principal hydrological properties are lateral and vertical intrinsic permeabilities of $300 \times 10^{-12}$ $m^2$ and $1700\times10^{-12}$ $m^2$, respectively, effective matrix porosity of 8.5% and a 25 m/year estimate for regional groundwater flow.

### *Geochemical setup*

Aradóttir et al. (2011) carried out an extensive literature review of water-basalt interaction at low and elevated $CO_2$ conditions, with the objective of predicting which minerals are likely to dissolve and precipitate during *in situ* $CO_2$ mineral sequestration. Widely used thermodynamic databases did not contain the mineral assemblage needed for successfully simulating the alteration processes observed in nature, since important primary and secondary minerals were missing (Aradóttir et al., 2012b). Aradóttir et al. (2012b) thus developed and evaluated a thermodynamic dataset describing 36 mineral reactions of interest for $CO_2$-water-basalt interaction. All minerals in the database were compiled to the CarbFix numerical models, along with kinetic variables describing their dissolution and precipitation. Four primary minerals and/or glasses were used in CarbFix simulations; Plagioclase, Pyroxene, Olivine and basaltic glass. (See Aradóttir et al., 2012a for more detail and references.)

### *Simulations*

Reactive chemistry was coupled to calibrated hydrological models and predictive mass transport and reactive transport simulations carried out for both a 1200-ton pilot $CO_2$ injection and a full-scale 400,000-ton $CO_2$ injection scenario. Pure $CO_2$ was assumed to be injected in the simulations, and the effects of $H_2S$ and $H_2$ in the gas mixture that is actually being injected were thus neglected.

## SulFix model

Development of reactive transport models for the SulFix project has only recently begun; the models are consequently not as complex as the CarbFix models, and results are preliminary.

### *Flow setup*

A tracer test carried out in 2010 indicates a fast flow path between SulFix injection well HE-08 and observation well HE-46, indicating fracture flow to be dominant in the storage formation (Gunnarsson et al., 2011, and Scott et al., 2011). Fractures are, however, connected to a porous matrix that contains the majority of the reactive surface area in the storage formation.

In view of this, it was decided to use a one-dimensional dual porosity model as a first approach to simulating the SulFix injection. Figure 3 shows a simple schematic of the conceptual model along with an illustrative representation of elements and connections.

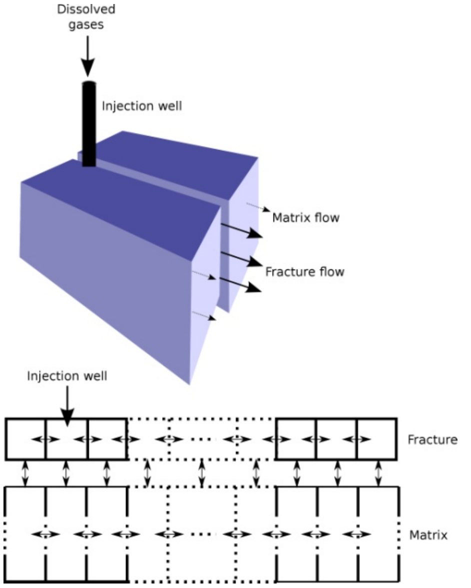


**Figure 3.** Schematic of the conceptual one-dimensional dual porosity model used for SulFix simulations (top) and illustration of elements and connections in the developed model (bottom).

Fracture permeability was calculated to be $733\times10^{-12}$ $m^2$, by assuming laminar flow and fracture diameter of 2 mm. Matrix porosity was assumed to be 2.5%, and the assumed matrix permeability of $0.02\times10^{-12}$ $m^2$ is that of the Hengill reservoir model at a corresponding depth (Gunnarsson et al., 2011).

### *Geochemical setup*

The thermodynamic dataset developed for CarbFix by Aradóttir et al. (2012b) was used as a basis for a thermodynamic database for SulFix simulations. Several minerals needed to be added to the dataset, since the alteration mineralogy associated with the injection is different from that in CarbFix, due to additional $H_2S$-water-basalt interaction and a considerably higher reservoir temperature.

Minerals added to the database were epidote, goethite, grossular, magnetite, prehnite, pyrite, pyrrhotite, sulfur and wollastonite. $Fe(OH)_4^-$ was furthermore added as a primary species. Data for $Fe(OH)_4^-$ was taken from Diakonov et al. (1999) and SUPCRT92 used for calculating logK values at discrete temperatures. Thermodynamic data for goethite and magnetite was taken from Holland and Powell (1998), and SUPCRT was used for calculating logK values for corresponding dissolution reactions. Equilibrium constants for other minerals were taken from Stefánsson et al. (2009).

Kinetic variables describing dissolution of epidote, goethite, grossular, magnetite, prehnite and wollastonite were taken from Palandri and Kharaka (2004). Mineral precipitation was represented using the same kinetic expression as that for dissolution. Kinetic variables describing dissolution and/or precipitation of sulfur, pyrite, and pyrrhotite at reducing conditions were not found in the literature. These minerals had thus to be assumed to precipitate and dissolve under equilibrium.

Basaltic glass (Oelkers and Gíslason, 2001 and Gíslason and Oelkers, 2003) was the only primary mineral used in SulFix simulations.

### *Simulations*

Predictive simulations were carried out of a 6-month continuous pilot injection. Gas mixture containing 75% $CO_2$, 24% $H_2S$, and 1% $H_2$ was dissolved in 10 kg/s HE-05 water and continuously injected into well HE-08. Simulation of water-rock interaction was then continued for up to 10 years.

## RESULTS

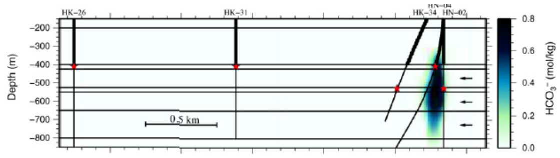
### CarbFix model

Reactive transport simulations of the 1200 ton pilot injection predict 85% $CO_2$ mineral capture within 5 years and 100% capture within 10 years. Simulations of the full-scale injection scenario predict 80% $CO_2$ mineral capture after 100 years. $CO_2$ sequestration rate is predicted to range between 1200 and 22,000 tons/year in both scenarios, a value in good agreement with the natural sequestration rate of the Hellisheidi geothermal system, which has been estimated to be 4100–23,500 tons/year. Relatively fresh basaltic formations are thus predicted to be promising geological $CO_2$ storage formations.

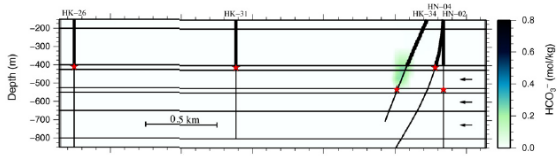
Figure 4 shows a cross-sectional view of modeled reservoir $HCO_3^-$ concentration in the pilot injection scenario, as predicted by three-dimensional reactive transport simulations. The figure indicates how injected carbonated water travels downstream as a plume with regional groundwater flow. Carbonate precipitation leads to significant dilution of the plume within five years and its disappearance within 10 years.

Calcite is predominantly the most abundant carbonate to form in reactive transport simulations, which is in agreement with natural basalt alteration in Iceland. Magnesite-siderite solid solution also precipitates, but in significantly lower quantities. Figure 5 shows a cross-sectional reservoir view of calcite precipitation after five years simulation time in the pilot injection scenario. $SiO_{2(am)}$, quartz and antigorite (Gunnarsson et al., 2005) precipitation is associated with calcite precipitation in the simulations, which is in agreement with natural alteration in the vicinity of mineral springs in Iceland.

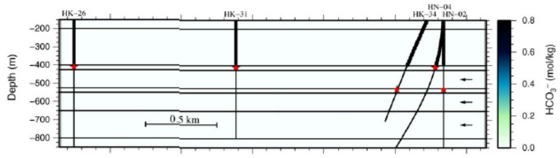
Ca leached from primary minerals is modeled to preferentially form calcite rather than zeolites, while clay minerals, simple oxides, and oxyhydroxides are predicted to compete with magnesite–siderite solid solution for leached Mg and Fe.



(a) 1 year



(b) 5 years



(c) 10 years

***Figure 4:*** Cross sectional view of modeled reservoir $HCO_3^-$ concentration in the pilot injection scenario after 1, 5 and 10 years, as predicted by three-dimensional reactive transport simulations. Cross sections are drawn along A-A' on figure 2. (Figure from Aradóttir et al, 2012a).

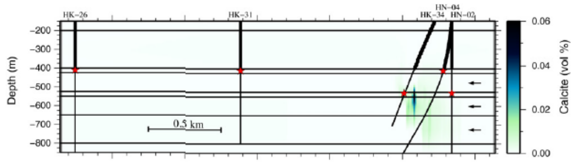


***Figure 5:*** Cross sectional view of modeled calcite precipitation in the pilot injection scenario after 5 years simulation time.The cross section is drawn along A-A' on figure 2. (Figure from Aradóttir et al, 2012a).

## SulFix model

Predictive simulations of the pilot SulFix injection show injected $H_2S$ to be sequestered into pyrrhotite. Pyrrhotite is a part of the mineral buffer assemblages believed to control $H_2S$ concentrations in geothermal systems, so, to that extent, simulated results are in agreement with observations from nature (Stefánsson, 2011).

Magnesite-siderite solid solution precipitation provides the primary $CO_2$ sink in the SulFix reservoir.

Other minerals that form in simulations are quartz, simple oxides and oxyhydroxides, magnetite, wollastonite, and clays.

Clay minerals, magnetite, and siderite appear to be competing with pyrrhotite for Fe leached from the primary basaltic glass. Precipitation of secondary minerals occurs both within the fracture and the matrix continuum.

## DISCUSSION

It is interesting to compare simulated $CO_2$ sinks in CarbFix and SulFix simulations. Calcite is predominantly the most abundant carbonate to form in the CarbFix reservoir, while magnesite-siderite solid solution precipitation provides the primary $CO_2$ sink in the SulFix reservoir. This is in agreement with well drill cuttings from Hellisheidi, which indicate calcite stability decreasing around 270°C (e.g., Helgadóttir et al., 2010). CarbFix and SulFix simulations were carried out at 25–90°C and 270°C, respectively. Thermodynamic data used for carbonates in the simulations is thus able to capture natural behavior of carbonate minerals.

Various indications point towards it not being feasible to inject large quantities of $CO_2$ along with $H_2S$ into deep and hot formations, if the objective is to sequester $H_2S$. On the one hand, more water is needed for dissolving the gas mixture than purer $H_2S$, as $CO_2$ is less soluble than $H_2S$. On the other, calcite is no longer the primary $CO_2$ sink under such reservoir conditions, but rather magnesite-siderite, a solid solution that competes with pyrrhotite and pyrite for Fe. The competition reduces the efficiency of mineral $H_2S$ sequestration. The above arguments support the separation of $CO_2$ and $H_2S$ prior to injection.

Concentration of $H_2S$ in aquifer fluids in Hellisheidi appears to be controlled by mineral buffer assemblages. Pyrite, prehnite, pyrrhotite and epidote or pyrite, pyrrhotite and magnetite have been identified as potential governing mineral buffers (Gunnarsson et al., 2011 and Stefánsson et al., 2011). Stefánsson et al. (2011) concluded that $H_2S$ concentrations in Hellisheidi equilibrate to the prehnite-bearing mineral assemblage, because Icelandic geothermal areas are usually low in magnetite, indicating that it is unstable in Icelandic geothermal systems. This is not in agreement with SulFix reactive transport simulations, in which magnetite and pyrrhotite buffered injected $H_2S$ but not prehnite, epidote, and pyrite. This suggests that the thermodynamic data used for describing the above mineral assemblages should be critically evaluated in future work.

It is furthermore important to acquire data describing the kinetics of $H_2S$ precipitation into minerals, as it is not reasonable to describe it only through thermodynamics. Additional sulfur-bearing minerals such as mackinawite (FeS) and greigite ($Fe_3S_4$) might also be important in simulating $H_2S$ sequestration in basaltic aquifers: studies have shown these minerals to be stable under anoxic conditions, which persist in Hellisheidi (e.g., Cahill et al., 2000; Benning et al., 2000).

## CONCLUSIONS

Reactive transport simulations carried out in this study indicate *in situ* $CO_2$ and $H_2S$ mineral sequestration in basalts to be a viable option in reducing gas emissions from geothermal power plants, as they predict fast mineralization of injected gases.

## ACKNOWLEDGMENT

We would like to thank Einar Örn Þrastarson, Trausti Kristinsson and Magnús Þór Arnarson for their never-ending contribution to CarbFix and SulFix. We are furthermore grateful to all our CarbFix collaborators for a challenging and fruitful cooperation since 2007. This work was funded by Reykjavík Energy, the 7th Framwork Programme of the EC (project no. 283148) and Geothermal Research Group GEORG (09-02-001).

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