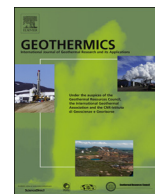




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Understanding amorphous silica scaling under well-constrained conditions inside geothermal pipelines



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ABSTRACT

Amorphous silica is a common precipitate in modern and ancient hot springs and in geothermal power plants, yet the corresponding precipitation rates and mechanisms are still highly debated, primarily due to the plethora of parameters that can affect the reactions in natural waters. Here, we report the results from a first ever industrial-scale time-resolved (1 day to 10 weeks) study of silica precipitation conducted at the Hellisheiði geothermal power plant (SW-Iceland). We show that such in-work pipelines of a geothermal power plant are ideal environments to investigate silica precipitation because the physicochemical conditions are well constrained and constantly monitored. Our results document that amorphous silica forms via two distinct precipitation modes: (1) the fast deposition of continuous botryoidal silica layers and (2) the growth of 3D fan- or ridge-shaped silica aggregates. The continuous layers grow by heterogeneous nucleation and subsequent surface controlled growth by monomer addition. In contrast, the 3D aggregates form through homogeneous nucleation of silica nano- and microparticles in solution, followed by deposition and cementation on the surface of the botryoidal layer. From the time-resolved data, silica precipitation rates of over $1 \text{ g m}^{-2} \text{ day}^{-1}$ are derived. Over time, this deposition of silica on pipelines and fluid handling equipment is detrimental to geothermal power production. Our data does not only help improve our understanding of silica precipitation from geothermal fluids, but the determined silica precipitation mechanisms and rates help improve mitigation strategies against silica scaling inside in-work geothermal power plants.

1. Introduction

Silica (SiO_2) is the most common chemical compound in the Earth's crust and a major component in most geothermal reservoirs. The maximum concentration of silica in geothermal fluids depends on the reservoir temperature and is controlled by quartz solubility or, if the temperature is below 110°C , by the solubility of chalcedony (Arnósson, 1975; Fournier and Rowe, 1966). When these geothermal fluids rise through the crust and emerge at the Earth's surface, rapid cooling results in supersaturation with respect to amorphous silica and precipitation. This sinter formation was studied in numerous modern and ancient terrestrial settings (Braunstein and Lowe, 2001; Cady and Farmer, 1996; Handley et al., 2005; Jones and Renaut, 2004; Konhauser et al., 2004, 2001; Mountain et al., 2003; Tobler et al., 2008) and recently, evidence for ancient silica sinter formation has even been

documented on Mars (Preston et al., 2008).

Identical to their natural analogues, amorphous silica precipitation also occurs in geothermal power plants, where fluids are rapidly cooled during energy production, resulting in precipitation of amorphous silica. This so called “silica scaling” is common in high-enthalpy geothermal power plants around the world e.g. Iceland, New Zealand, Japan, the USA, the Lesser Antilles and El Salvador (Dixit et al., 2016; Gunnarsson and Arnósson, 2003, 2005; Harrar et al., 1982; Meier et al., 2014; Mroczek et al., 2017; Padilla et al., 2005; Rothbaum et al., 1979; Yokoyama et al., 1993). In such systems, amorphous silica precipitation occurs in fluids characterized by a wide range of total silica concentrations (250–900 mg/L), temperatures ($20\text{--}200^\circ\text{C}$), pH (7.2–10.2), total dissolved solid concentrations (1300–93'000 mg/L) and different types of geothermal power plants (e.g. flash steam and binary). Because of the ubiquity and importance of this natural process

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to renewable energy production and sinter formation, amorphous silica precipitation was studied extensively in the laboratory. Amorphous silica forms via the condensation of silica monomers (H_4SiO_4) into Si-O-Si bonds (Iler, 1979), through polymerisation that can either occur at an interface (e.g., minerals, bacteria or plant matter) where it is described as “heterogeneous nucleation” or in the bulk fluid (“homogeneous nucleation”) (Benning and Waychunas, 2007). In both cases, once silica nuclei have reached a critical size (< 0.5 to 2 nm, Iler, 1979; Noguera et al., 2015; Tobler et al., 2009), they grow spontaneously by the addition of silica from solution. Monomers are the dominant growth species (Bohlmann et al., 1976; Bremere et al., 2000; Mroczek and McDowell, 1988) due to their predominantly neutral charge ($K_a \sim 10^{-8.8}$ at 120 °C) (Fleming and Crerar, 1982; Seward, 1974) in the slightly alkaline pH regime of silica-rich geothermal waters. In contrast, silica polymers and nuclei have a higher dissociation constant ($K_a > 10^{-8}$) (Dugger et al., 1964; Hair and Hertl, 1970), resulting in an overall negative surface charge. Their attachment to existing silica particles or surfaces and the aggregation of silica particles in solution in the absence of bridging cations will thus be limited by electrostatic repulsion. Experimental evidence showed that silica polymerisation, nucleation and growth are enhanced at slightly alkaline pH, elevated temperature, medium to high ionic strength (especially the presence of Al and Fe) and high total silica concentrations (Alexander et al., 1954; Crerar et al., 1981; Fleming, 1986; Gallup, 1997; Goto, 1956; Gunnarsson and Arnórsson, 2005; Icopini et al., 2005; Iler, 1979; Kitahara, 1960; Tobler and Benning, 2013; Weres et al., 1981).

These physicochemical factors also affect amorphous silica precipitation from naturally occurring geothermal fluids, where they are in competition with one another and it is often difficult to isolate the dominant parameter in any given fluid. In addition, microbial activity in hot springs (Mountain et al., 2003; Tobler et al., 2008) and high flow rates in geothermal power plants (Meier et al., 2014) make the understanding of these systems highly challenging. Therefore, most laboratory findings cannot be directly transferred and applied to silica precipitation from natural geothermal fluids (Carroll et al., 1998) and although a number of field studies investigating silica sinter formation around hot springs (e.g. Braunstein and Lowe, 2001; Handley et al., 2005; Jones and Renaut, 2004; Konhauser et al., 2004; Mountain et al., 2003; Tobler et al., 2008) give insights into what happens once geothermal solutions reach the Earth surface, they do not address processes that govern formation of amorphous silica inside geothermal power plants. This is despite the fact that in-production geothermal power plants represent systems with very well constrained physicochemical conditions that are thus ideal sites to investigate silica precipitation. Such studies would not just help quantify real world silica precipitation but would also inform silica scaling mitigation strategies, thus improving efficiency of geothermal energy production and reduce costs.

The main reason for the dearth of on-site studies lies in the difficulties in accessing both fluids and silica scales during energy production, without affecting normal operations. A few studies aimed to circumvent these problems by conducting experiments in bypass systems from the main production (Carroll et al., 1998; Dixit et al., 2016; Harrar et al., 1982; Mroczek et al., 2017; Rothbaum et al., 1979). However, the conditions in such bypass systems are most often markedly different from inside in-work pipelines and the bypass is often less well constrained, resulting in data that are only partly applicable to the in-production systems.

To change this landscape, we have for the first time conducted a detailed study inside actual in-work geothermal pipelines of a high enthalpy geothermal power plant at Hellisheiði, SW-Iceland. We monitored silica precipitation for up to 10 weeks using stainless steel scaling plates deployed at different positions within the pipelines and characterized the precipitated solids, as well as fluid composition and the physicochemical conditions, under which precipitation occurred. Our results reveal that two largely independent pathways control silica precipitation. We observe the formation of a continuous, botryoidal

Table 1

Duration and starting/end dates of individual scaling plate deployments. The duration of the heat exchangers in early October 2014 (after the 10 week and before the 2 week deployment) was part of regular (every 4–6 months) and scheduled maintenance at the Hellisheiði power plant to remove the accumulated silica scales.

	Deployment [days]	Start date	End date
1 day	1	02/02/2015	03/02/2015
3 days	3	16/03/2015	19/03/2015
1 week	7	27/10/2014	03/11/2014
2 weeks	14	03/11/2014	17/11/2014
4 weeks	28	23/06/2014	21/07/2014
6 weeks	41	03/02/2015	16/03/2015
10 weeks	72	21/07/2014	01/10/2014
Cleaning of heat exchangers		06/10/2014	07/10/2014

layer of silica on the scaling plate surfaces and the growth of 3D structures consisting of silica particles that nucleate homogeneously in the fast travelling fluid and are then deposited and cemented to the silica layer. Through this work, we evaluate the first ever amorphous silica precipitation rates and mechanisms inside in operando pipelines of an active geothermal power plant.

2. Materials and methods

2.1. Field deployments

The time-resolved deposition of amorphous silica was studied on stainless steel scaling plates (5.4×2 – 2.5 cm) deployed for between 1 day and 10 weeks (Table 1) at four different locations within the pipelines of the Hellisheiði power plant (Fig. 1): (1) several metres before the heat exchangers, (2) several metres after the heat exchangers, (3) several tens of metres downstream of location 2, after a bypass with fluid that had not passed through the heat exchangers that then re-joined the main pipeline and before mixing with condensed steam (equivalent to almost pure water) and (4) ~ 1300 m downstream from location 3, at the Húsmúli re-injection site. These locations were chosen because they differed with respect to physicochemical conditions of the fluid (temperature, flow rate, silica concentration etc.) thus allowing us to study how these parameters affected silica precipitation. It is worth noting that due to operational constraints the time resolved deployments were not done in order of deployment length (Table 1).

At each location, scaling plates made from S316 stainless steel (Fig. 1B–D) were attached to a sampling rod and inserted into the fast flowing geothermal fluid through valves in the pipeline walls. The surfaces of the plates were aligned to be parallel to the flow. The power plant operators monitor the temperatures and flow rates at each of the chosen sampling locations hourly to identify changes in production parameters and as a guide for when maintenance (e.g., heat exchanger cleaning) is required. These continuous datasets helped make sure that the scaling plate deployments were started/finished during periods where no maintenance was required. At the end of each deployment, the plates were removed from the fluid, gently rinsed with distilled water to prevent the precipitation of salts during evaporation of the geothermal fluid and then dried at 40 °C for up to 16 h. For short deployments (1 day, 3 days and 1 week), the scaling plates were weighted pre- and post-deployment in order to determine the mass of precipitated silica. For deployments of 2 weeks or longer this was not considered feasible as in most cases larger amounts of silica precipitated on the plates and some of the accumulated silica would have been either lost because of the fast fluid flow rates (fluid flow 280 to 430 L/s) or during scaling plate handling post removal.

At the beginning and end of each deployment, the fluid at each location was sampled. It was cooled down to ambient values (21 – 27 °C) using a ~ 5 m stainless steel coil and temperature and pH were

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