



Effect of diagenetic phase transformation on the silicon isotope composition of opaline sinter deposits of Geysir, Iceland

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ABSTRACT

Detectible $\delta^{30}\text{Si}$ variations in present-day chemical silica deposits have stimulated the application of silicon isotopes to infer environmental conditions from ancient equivalents. Interpretations of $\delta^{30}\text{Si}$ signatures remain problematic in view of potential post-depositional changes, of which magnitudes and underlying mechanisms are largely unknown. A critical issue in the interpretation of isotope data from cherts concerns the extent to which early-diagenetic processes modify original $\delta^{30}\text{Si}$ signatures. Here, we report $\delta^{30}\text{Si}$ variations in opal-A, opal-A/CT and opal-CT from fossil sinter deposits in an active discharge apron in the Geysir geothermal field, Iceland. Opal-A samples show an average $\delta^{30}\text{Si}$ of $-0.7 \pm 0.2\%$, while opal-CT samples are isotopically lighter with an average $\delta^{30}\text{Si}$ of $-2.0 \pm 0.4\%$, implying a sizable shift of approximately 1.3% between the different phases. This shift can be explained by repetitive dissolution/re-precipitation processes, diffusive transport or temperature differences during phase transitions. On average, the fossil opal-A tends to be less negative in $\delta^{30}\text{Si}$ than amorphous silica that recently precipitated from the hydrothermal water. The difference can be attributed to primary variability in isotopic fractionation that accompanies precipitation out of spring water at the surface, or to a post-depositional release of surface ^{28}Si at the onset of diagenetic formation. Our results corroborate the perception that original silicon isotope signatures of silica, acquired during chemical precipitation from a saturated solution, may not be preserved in the geological record, and that post-depositional changes must be taken into account when interpreting data from ancient chert deposits.

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1. Introduction

A fundamental prerequisite for the interpretation of silicon isotope data from cherts with a silica precursor that precipitated directly from a fluid (orthochemical cherts) is insight into the extent to which original signatures, acquired when the silica was deposited, have been modified during diagenesis. Commonly, the pristine amorphous silica experiences a sequence of diagenetic changes until a more stable, crystalline form of silica is produced. With little or no burial, near-amorphous opal-A will gradually transform to paracrystalline opal-CT (α -cristobalite or α -tridymite) and/or well-ordered opal-C (α -cristobalite), and finally to microcrystalline quartz (e.g., Jones and Segnit, 1971; Herdianita et al., 2000; Lynne and Campbell, 2003; Lynne et al., 2005; Lynne, 2015). Despite observable microstructural changes during the transformation, the underlying mechanisms are poorly understood (e.g., Herdianita et al., 2000; Rodgers et al., 2004). The opal-A to opal-CT/C transition is accompanied by water loss, decreasing porosity and d-spacing, and increasing density (Rice et al., 1995; Herdianita et al.,

2000; Campbell et al., 2001; Rodgers et al., 2004; Jones and Renaut, 2007; Lynne, 2015). The onset of recrystallization might begin after ~20,000 years, so that most silica sinters should be in the form of microcrystalline quartz after a period of ~50,000 years (White et al., 1956; Herdianita et al., 2000). Other studies question this time span and consider that opal-A to quartz transformation proceeds in response to specific conditions that affect the deposit in question (Rodgers et al., 2004; Lynne et al., 2005, 2006, 2007; Lynne, 2015).

Silicon isotope fractionation can be induced by biological and abiotic reactions, whereby in both cases the light silicon isotope is preferably incorporated in the reaction product (e.g., Douthitt, 1982; Ding et al., 1996; De La Rocha et al., 1997, 2000; Georg et al., 2006, 2007, 2009; Opfergelt et al., 2009, 2010, 2011, 2012). Processes controlling the fractionation are incompletely understood. Suggested factors influencing silicon isotope fractionation include biological uptake (e.g., De La Rocha et al., 1997; Varela et al., 2004; Opfergelt et al., 2011; Hendry and Robinson, 2012; Sutton et al., 2013), weathering degree (e.g. Ziegler et al., 2005; Georg et al., 2007; Opfergelt and Delmelle, 2012; Pogge von Strandmann et al., 2012) and dissolution/re-precipitation processes (e.g. Basile-Doelsch et al., 2005; Ziegler et al., 2005; Georg et al., 2009; Opfergelt et al., 2011; Cornelis et al., 2014). Experimental studies on abiotic silicon isotope fractionation during silica removal

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from solution have found a range between +0.5 and –5%, with suggested fractionation controls being the precipitation rate, surface complex formation or competition between kinetic and equilibrium isotope fractionation (Li et al., 1995; Delstanche et al., 2009; Geilert et al., 2014a; Oelze et al., 2014, 2015; Roerdink et al., 2015). Phase transformations during early-diagenetic processes in silica deposits are expected to be accompanied by shifts in silicon isotope compositions as this has also been observed for oxygen isotopes (e.g. Knauth and Epstein, 1976; Knauth and Lowe, 2003; Hren et al., 2009). Knowledge about the magnitude and direction of the entire shift between the amorphous silica precursor (probably opal-A in most cases) and the end product (usually crystalline quartz) is crucial for interpretations of ancient silica-rich deposits in which only the final quartz product is available for study.

Marin-Carbonne et al. (2012) documented significant micrometer-scale $\delta^{30}\text{Si}$ variations in micro-quartz of Archean and Proterozoic cherts, and attributed this to a sequence of dissolution of an amorphous silica precursor and subsequent crystallization of microquartz during diagenesis in a closed system. Along the same lines, Marin-Carbonne et al. (2014) ascribed $\delta^{30}\text{Si}$ differences between silica phases in samples of Eocene marine cherts to a similar diagenetic control. They found that microquartz had a mean $\delta^{30}\text{Si}$ value 1.65‰ lower than opal-CT, with each phase also showing a significant range on a micrometer scale. Tatzel et al. (2015) also described a >1‰ diagenetic shift in Pliocene–Pleistocene marine siliceous sediments, but in opposite direction, presumably associated with conversion of biogenic opal-A to opal CT.

This study documents silicon isotope compositions in sinter deposits, comprising opal-A, opal-A/CT and opal-CT, from a hot-spring setting in Iceland where microstructural changes accompanying opal phase transition were previously studied by Jones and Renaut (2007). Results provide new evidence for shifts in silicon isotope signatures accompanying the early-diagenetic transformation of amorphous silica, precipitated abiotically from a saturated solution, and offer a perspective on the significance of data obtained from cherts in the geological record.

2. Geological environment and sampling site

2.1. Geysir discharge apron

The high-temperature Geysir geothermal field is situated in the eastern vicinity of the main volcanic belt in southern Iceland (Arnósson 1970, 1985; Pálmason and Saemundsson, 1974). The object of this study is the Geysir discharge apron at the eastern rim of the field (Fig. 1). It stretches from a single, currently active vent pool over a distance of ca. 100 m to the NE and E, and ca. 25 m to the N, NW and S. The NE part of the apron is covered by soil and plants, whereas open sinter deposits dominate the S and NW parts (Jones and Renaut, 2010). The structure is almost entirely composed of opal, formed originally as siliceous sinter that precipitated from discharging hot spring water. Jones and Renaut (2007) subdivided a ca. 2–2.5 m thick vertical succession of deposits at the northwestern edge of the Geysir apron into five lithological units (A–E) based on field appearance, petrographic features and mineralogy. From tephrochronological, historical, geological and geomorphological information Jones et al. (2007) constructed a time frame for the accumulation of these deposits. An initial phase of sinter formation (presumably ca. 10,000 to 4000 years BP) started with deposition of non-laminated sinter and clay (unit A), which was followed by two units of finely laminated sinter. The lower unit B, consisting of opal-CT, shows a stratigraphically uninterrupted, sharp transition to unit C wherein opal-A dominates. The following phase (ca. 3300–2850 years BP) is marked by deposition of volcanic ash and minor intercalated sinter (unit D). After an interval without deposition, sinter formation resumed (ca. 800–900 years BP) and continued until the present day, probably associated with higher activity of the Geysir hot spring. This top layer (unit E) consists of opal-A. Throughout this paper we will

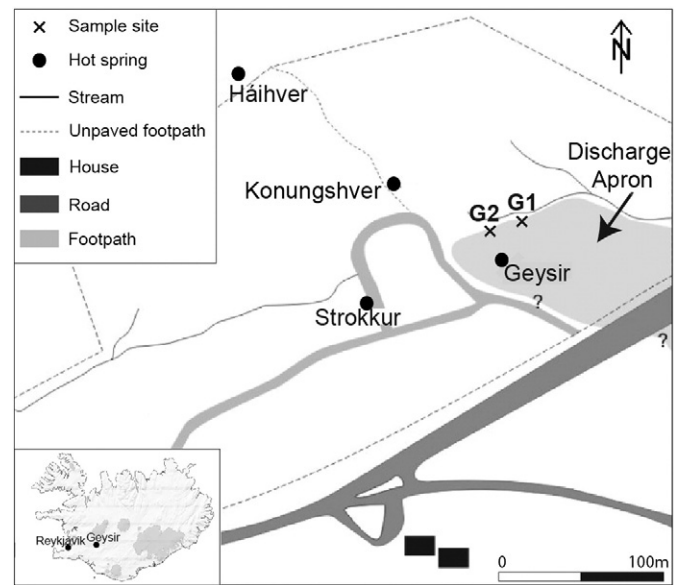


Fig. 1. Geysir discharge apron with locations of sampling sites G1 and G2.

use the term “fossil” for any of the silica deposits of units A–E, as opposed to sinter that has recently been deposited from thermal water issued at the currently active pool.

In recent times, the 20 m wide pool issues slightly alkaline (pH = 9.1), low-salinity, bi-carbonate water at an approximate rate of 9 l per second (Torfason, 1985; Pasvanoglu et al., 2000; Geilert et al., 2015). The outflow area is mainly an unchanneled sheet flow, forming small sinter terraces. During the sampling campaign in June 2012, the pool water had a temperature of 90 °C whereas it decreased to 20 °C over a 40 m distance along the outflowing stream. Concentrations of aqueous SiO_2 everywhere exceeded 500 ppm, implying oversaturation with respect to amorphous silica.

2.2. Sampling and sample descriptions

Opals selected for this study are from units B, C and E, which were collected at two different sites, termed G1 and G2 (Figs. 1 and 2, site coordinates in Table 1), where Jones and Renaut (2007) investigated microstructures associated with the transition from opal-A to opal-CT. Three samples were taken from site G1. Sample 12-Gey-26 is composed of porous layers with alternating dark (up to 5 mm thick) and light gray (≤ 2 mm) layers. Locally, a fatty glamor akin to conchoidal fractures is visible, and some parts show signs of strong weathering. Some of the lighter gray laminae show internal, palisade-like structures. Sample 12-Gey-27 contains relatively thick layers of white, porous material (~10 mm thick), intercalated with thinner (≤ 2 mm), dense layers of dark-gray material. In general, the white layers do not show an internal structure, although palisade-like structures exist in some parts. Sample 12-Gey-29 has a more weathered appearance than other samples and shows variable brownish-ochre colors with barely visible lamination. The deposit is partly made up of cemented small spheres with pores and cavities, and with a palisade structure in denser parts.

Seven samples were taken from site G2. Opals 12-Gey-16 to 12-Gey-23 are generally well preserved and show laminations of dark gray and whitish-ochre layers. Dark gray layers (≤ 3 mm) are homogeneous, dense and show the fatty glamor of conchoidal fractures. They alternate with porous, whitish-ochre irregular layers (up to 5 mm), in some parts showing fibrous structures (12-Gey-19). Opals 12-Gey-24 and 12-Gey-25 from the surface of the site are highly porous and laminated with thin light gray (≤ 3 mm) and thicker (5–10 mm) dark gray layers.

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