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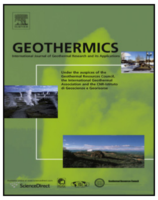

Compositions of hydrothermal silicates and carbonates as indicators of physicochemical conditions in the Reykjanes geothermal system, Iceland

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ABSTRACT

This paper presents results of a quantitative investigation of mineral chemistry in the Reykjanes geothermal system, Iceland, designed to assess the relative influence of various hydrothermal minerals on the observed bulk-rock mass changes of elements. Detailed petrography and electron microprobe analyses (n = ~1400) were performed on samples of rock cuttings from eleven wells that intersect different parts of the hydrothermal system. An important finding was the strong partitioning of barium into adularia, which becomes incorporated through "capture" substitution into the 12-fold coordination site, coupled with Al^{3+} substitution into the tetrahedral site. Adularia has average and maximum Ba contents of 0.8 and 3.5 wt.%, respectively. In addition, there is no relationship between Ba concentration in adularia and crystal size, occurrence type (replacement versus vein/vug), or rock composition. There is also no correlation between the Ba content of replacement adularia with that of the igneous plagioclase being altered, demonstrating that Ba concentration in the altered rocks of the Reykjanes geothermal system is being controlled by large-scale hydrothermal redistribution processes and not primary chemostratigraphic variability. Calcite exerts a major control on strontium proportions in the bulk rock, and contains up to 1 wt.% Sr. Concentrations of many minor and trace elements and some major elements in hydrothermal minerals at Reykjanes correlate positively with measured downhole temperature (to varying degrees), e.g.: Ba and Rb in adularia; $Al_{(iv)}$ in chlorite; Fe, Mg, Mn, and Sr in calcite; and the andradite component in garnet. The Fe/(Fe + Mg) of chlorite is unrelated to the precursor host rock ratio. Chlorite geothermometry, based on $Al_{(iv)}$ substitution and Fe/(Fe + Mg) content, predicts temperatures of 94–313 °C (average = 236 °C). These temperatures are similar to or less than the present-day temperatures measured for corresponding sample depths in all wells (with the exception of a shallow interval from RN10). This may indicate a prolonged and ongoing heating trend in the deep liquid-phase region of the Reykjanes reservoir.

1. Introduction

Chemical constituents in geothermal fluids are transported and incorporated into secondary minerals as major (>1 wt.%), minor (0.1–1 wt.%), or trace (<0.1 wt.%) components as a function of the intrinsic properties of the constituent (e.g., ionic potential) and the physiochemical conditions of the aqueous system. Dissolved components become integrated into mineral lattices as major elements or "impurities" following the rules set out by Goldschmidt (1937) and Ringwood (1955a,b), whereby ions of similar size, charge, and electronegativity are able to substitute for one another by "camouflage", "capture", or "admission" substitutions or find residence in voids and interstices present in mineral lattices. The degree to which this incorporation occurs is also a function of element availability (i.e., the statistical likelihood of an element interacting with the lattice of a precipitating mineral), kinetic variables (e.g., resulting from differing rates of precipitation), and independent system conditions such as temperature and pressure (which induce lattice expansion and contraction effects, and also affect the stability of aqueous species).

Because of the physicochemical controls on element substitutions, mineral-chemical data can be applied to the investigation of geothermal systems in order to: (1) gain insight into physicochemical conditions and processes occurring at the time the composition of the mineral was last "fixed"; (2) distinguish metasomatic signatures and specific alteration styles, and; (3) evaluate spatial variations of the data in relation to the hydrology of the system

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(e.g., proximity to upflow or feed zones; Gifkins et al., 2005). If data from downhole measurements or multiple mineral generations are available, then insight can also be gained into the physicochemical evolution of the system.

The controls of rock leaching and hydrothermal mineral buffers on the major element chemistry of geothermal fluids have been extensively studied in Icelandic geothermal systems (Arnórsson, 1978; Arnórsson et al., 1982; Arnórsson et al., 1983; Stefánsson and Arnórsson, 2000, 2002) and in other geothermal systems around the world (Giggenbach, 1980, 1981, 1984, 1988; and many others). The trace element chemistry of geothermal fluids has received much less attention, however, this aspect of Icelandic systems has been investigated by Hardardóttir et al. (2009), Kaasalainen and Stefánsson (2012), and Kaasalainen et al. (2015). An essential part of the fluid-rock interaction "story" in geothermal systems comes from investigations of altered rock through petrographic, lithogeochemical, and mineral-chemical studies. Petrographic studies of geothermal systems are numerous, and relationships of minerals to hydrothermal processes and physicochemical conditions, in many cases, are well understood (see reviews in Browne (1970, 1978, 1984), Elders (1977), and Reyes (2000)). Lithogeochemistry has been an under-utilized tool by the geothermal research sector and industry. Recently, however, a detailed field-wide lithogeochemical investigation was conducted on the Reykjanes geothermal system by Libbey and Williams-Jones (in review). Mineral-chemical data have been incorporated into an appreciable number of studies of active geothermal systems, however, most have restricted their consideration to major element chemistry and/or have utilized limited sample sets in terms of spatial distribution or mineral types (previous examples from the Reykjanes geothermal system include: Sveinbjörnsdóttir (1991), Lonker et al. (1993), Freedman et al. (2009), Marks et al. (2010, 2011), Raffone et al. (2010), and Fowler et al. (2015).

The current study employs a large petrographic and mineral-chemical dataset for silicate and carbonate phases from the Reykjanes geothermal system to augment understanding of system-wide variations in mineral chemistry, physicochemical evolution, and the specific mineralogical controls on bulk-rock metasomatic signatures described in Libbey and Williams-Jones (in review). A particular aim of this study is to increase the understanding of barium incorporation into hydrothermal minerals, as lithogeochemical data from Reykjanes have shown this element to be particularly enriched in the upflow zone (i.e., mass additions of >400 mg/kg$_{\text{PrecursorRock}}$), and there is evidence that it may be useful for making system mass/volume estimates. A companion study by Libbey and Williams-Jones (2016) details the sulfide mineralogy and mineral chemistry in the Reykjanes geothermal system.

2. The Reykjanes geothermal system

The Reykjanes geothermal system is a basalt-hosted, seawater-dominated active hydrothermal system situated at the tip of the Reykjanes peninsula in southwest Iceland, which represents an on-shore expression of the Mid-Atlantic Ridge (Fig. 1a). Surface manifestations and altered ground at Reykjanes cover an area of $\sim$1 km^2, and the natural heat flow of the system is estimated to be 130 ± 16 MW$_{th}$ (Friðriksson et al., 2006). The upper 1 km of the Reykjanes system is dominated by hyaloclastite tuff, breccias, pillow basalts, and tuffaceous and marine sediments. The deeper parts of the system are composed primarily of basaltic lavas and diabase dikes (Marks et al., 2010). Altered and hydrothermally-cemented tuffaceous and sedimentary successions at depths of 400–800 m

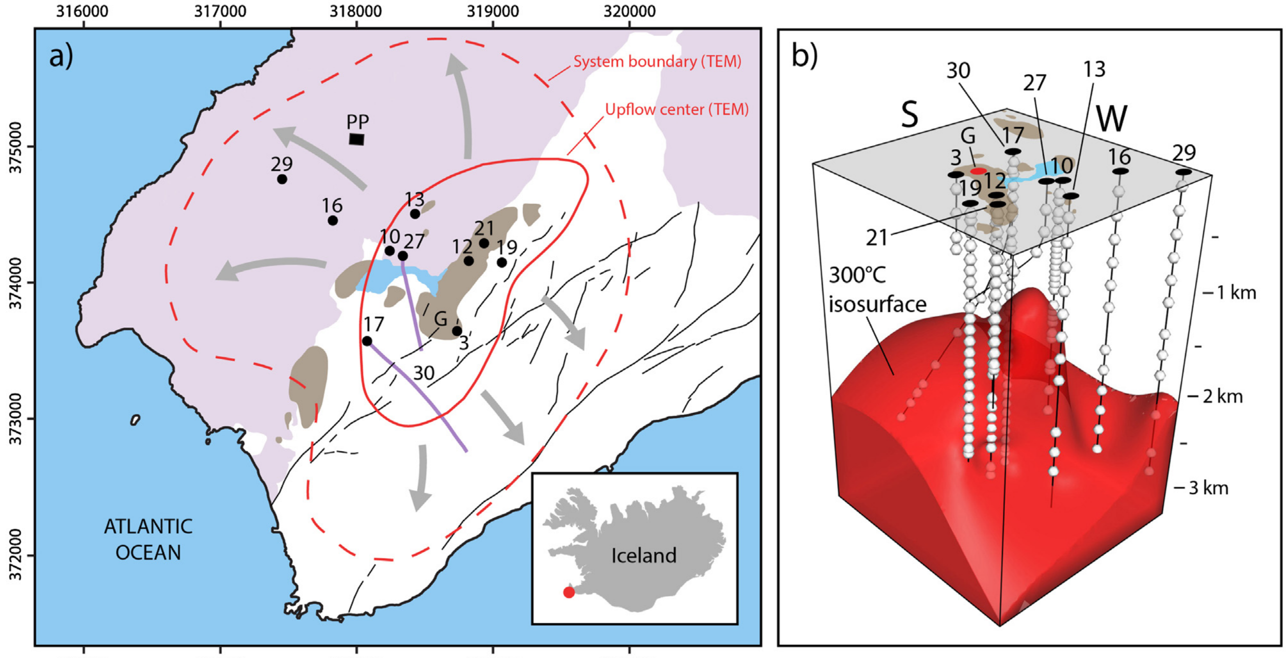

Fig. 1. (a) Location map of the Reykjanes geothermal field showing the system boundary (which corresponds to where the low resistivity cap reaches a depth of 800–1000 m), the upflow center (as defined by TEM—transient electro-magnetics), and the inferred outflow paths (grey arrows; TEM studies by Karlsdóttir, 2005), fractures/faults (black lines; Sæmundsson, 2011), areas of altered ground (brown), lavas younger than surface fracturing/faulting ($\sim$800 to $\sim$2000 years old; Sæmundsson, 2011), and the "Grey Lagoon" brine discharge pool (blue in center). PP = Reykjanes geothermal power plant; G = Gunnuhver thermal area. The map projection is ISN93. (b) 3D well map showing sample locations from which polished thin sections were prepared (spheres), the measured 300 °C isosurface (established from measurements in 17 wells), areas of altered ground (brown), the Gunnuhver thermal area (red dot), and the "Grey Lagoon" brine pond (blue). Well paths and temperature data were provided by HS Orka and ISOR. Figure modified from Libbey and Williams-Jones (2016). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)