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Fluid origin and evolution of Cu-Pb-Zn mineralization in rhyolite breccias in the Lón area, southeastern Iceland

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ABSTRACT

Iceland, the landward extension of the Mid-Atlantic Ridge, hosts dilute, predominantly meteoric hydrothermal systems that rarely form base metal (Cu-Pb-Zn) mineralization. One occurrence of Cu-Pb-Zn mineralization in intrusive rhyolitic breccias is in the Lón area of southeastern Iceland. Petrographic, electron-probe, fluid inclusion, stable isotope, and U-Pb zircon dating analyses on samples from Lón constrain the conditions and timing of sulfide mineral formation.

Observations of outcrops and hand samples suggest that hydrothermal fluids precipitated chalcopyrite, sphalerite, galena, quartz, epidote, chlorite and calcite in rhyolitic breccia pipes and adjacent basalt flows. The mean salinity of liquid-dominated, multi-phase fluid inclusions in quartz coeval with chalcopyrite inclusions in quartz is 4.2 wt% NaCl, and the mean trapping temperature is 332 °C, which is consistent with the results of $\delta^{34}S$ geothermometry of coexisting galena and chalcopyrite. Calculated $\delta^{18}O$ and δD values of fluids in equilibrium with epidote coeval with chalcopyrite range from -5.2 to -2.7 ± 0.1‰ and from -35.9 to -27.7 ± 0.1‰, respectively. The $\delta^{18}O$ values of fluids in equilibrium with quartz coeval with chalcopyrite are up to 5‰ larger than those of fluids in equilibrium with late stage quartz precipitated after chalcopyrite. The U-Pb crystallization age of magmatic zircons in the rhyolite breccia is 2.6 ± 0.1 Ma, significantly younger than the proximal 3.7 to 7.3 Ma silicic intrusions of southeastern Iceland.

Our results indicate that early-stage, mineralizing fluids derived from a mixture of meteoric water, seawater, and a minor magmatic water component exsolved from an evolved anatexis-produced melt. Late-stage fluids were derived exclusively from meteoric waters. Although anatectic dehydration melting of altered basalt produced millions of years of felsic magmatism in southeastern Iceland, only hydrothermal fluids that derived from a mixture of meteoric water, seawater, and brine exsolved from a highly evolved melt concentrated base metals in significant quantity to produce base metal sulfide mineralization.

1. Introduction

Hydrothermal base metal (Cu-Pb-Zn) mineralization is rare in Iceland, which is a landward extension of the Mid-Atlantic Ridge (Fig. 1A; Jolley and Bell, 2002). Icelandic igneous activity is the product of superimposed mantle plume and mid-ocean ridge magmatism (Sigmarsson and Steinthórsson, 2007), and the oldest rocks exposed on the eastern and western flanks of the island are less than ~16 Ma (Saemundsson, 1979). Active and fossil hydrothermal systems in Iceland, which are pervasive across the island and range greatly in temperature, are typically highly dilute and derive their fluids from meteoric waters (Kaasalainen and Stefánsson, 2012; Kaasalainen et al., 2015). The generally low Cl content of Icelandic thermal waters results in very low (ppb or less) concentrations of base metals (Stefánsson et al., 2017). A notable exception is the modern Reykjanes hydrothermal system, where saline seawater-derived fluids precipitate metal sulfide minerals in basalt (Hardardóttir et al., 2009).

The island's paucity of base metal deposits correlates with the relative scarcity of evolved magma bodies (Oskarsson et al., 1982), which in other geologic settings may exsolve metal-rich ore-forming hydrothermal solutions (Hedenquist and Lowenstern, 1994). Limited bimodal silicic and basaltic magmatism occurs in a number of central volcanic complexes, and recent isotopic studies of Icelandic silicic rocks support the hypothesis that silicic magmas are derived largely from dehydration melting of hydrothermally altered basalts (Bindeman et al., 2012; Pope et al., 2013; Padilla, 2015). Several modern hydrothermal systems hosted in bimodal rhyolites and basalts, including Krafla, Námafjall,

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Torfajökull, and Geysir dissolve base metals in concentrations that are much higher (up to 10 s to 100 s of ppb) than average Icelandic geothermal systems, but that are insufficient to precipitate base metal minerals in appreciable quantity (Kaasalainen and Stefánsson, 2012). Hydrothermal base-metal sulfide minerals are interpreted to have precipitated in felsic igneous rocks in ancient tectonic rift settings and comprise a major type of ore deposit known as volcanogenic massive sulfide (VMS) deposits (Galley et al., 2007). VMS deposits hosted in felsic rocks occur globally but few are reported in modern or recent hydrothermal systems (Sillitoe, 1982).

Fossil silicic/mafic intrusive complexes exposed in southeastern Iceland (Fig. 1B; Cargill et al., 1928) have been dated between 3.7 and 7.3 Ma (Martin et al., 2011; Padilla, 2015), and within the Lón area of this region (Fig. 1B), there are rhyolite breccia pipes that contain hydrothermal Cu-Zn-Pb-sulfide mineralization (Jankovic, 1970a, 1970b; Vigdís Hardardóttir and Gestur Gíslason, *Personal communication*). Field mapping, petrographic observations, fluid inclusion heating and freezing measurements, and O-H-S stable isotope analyses were undertaken to characterize the base metal sulfide mineralization, and to reconstruct the composition and origin of the hydrothermal solutions that precipitated Cu-Pb-Zn minerals in rhyolite breccias at Lón. U-Pb crystallization ages of magmatic zircons in the rhyolite breccia and a rhyolite dike proximal to the breccia pipes were measured to constrain the timing of hydrothermal mineralization with respect to nearby intrusive centers. Our results suggest that rhyolitic melts produced through anatectic dehydration melting of hydrothermally altered basalts in Iceland facilitated the concentration of base metals in local hydrothermal fluids by exsolving magmatic fluids that mixed with meteoric water and seawater.

2. Geologic background

2.1. Geologic and tectonic setting

As a consequence of the confluence of the Mid-Atlantic Ridge and Iceland mantle plume, Iceland hosts numerous active and fossil magma-hydrothermal systems. Active high-temperature geothermal systems are prevalent in the Neo-Volcanic Zone, and glacially eroded remnants of fossil hydrothermal systems are exposed in the western and eastern margins of the island (Fig. 1A; Arnórsson, 1995). Here we focus on Cu-Pb-Zn mineralization hosted in silicic magmatic rocks in the Lón area of eastern Iceland (Fig. 1B).

Active geothermal systems on the Reykjanes peninsula (Fig. 1A) provide the only other significant occurrences of base metal sulfide mineralization in Iceland (Hardardóttir et al., 2009). The Reykjanes hydrothermal system is hosted in basalt with seawater-derived geothermal fluids dissolving up to 100 s of ppm of various base metals (Hardardóttir et al., 2009; Marks et al., 2010; Fowler and Zierenberg, 2016), with elevated concentrations of Fe (9–140 ppm), Cu (14–17 ppm), Zn (5–27 ppm), and Pb (120–290 ppm) (Hardardóttir et al., 2009).

Elsewhere in Iceland, geothermal fluids are largely derived from meteoric waters (Ármannsson, 2016; Stefánsson et al., 2017). High-temperature meteoric geothermal fluids in Iceland have lower concentrations of solutes than most extant hydrothermal fluids worldwide, with total dissolved solids in the range of 1000 to 2000 ppm (Arnórsson, 1995). Since basalts in Iceland generally have low concentrations of base metals (~50–200 ppm; Cargill et al., 1928; Schilling et al., 1983), many transition metals occur in ppb concentrations in Icelandic high-temperature geothermal fluids (Kaasalainen and Stefánsson, 2012; Kaasalainen et al., 2015). In typical Icelandic geothermal fluids, Cu and Ni have average concentrations <1 ppb, Pb <10 ppb, and Fe <100 ppb and are systematically undersaturated with respect to sulfide minerals (Kaasalainen et al., 2015).

Although dominantly meteoric systems hosted in rhyolites such as Krafla, Námafjall, Geysir, and Torfajökull have concentrations of base metals that are greater than other meteoric-derived thermal waters, they are too low to precipitate base metal sulfide minerals in appreciable quantity. Maximum temperatures in deep geothermal drill holes reach up to ~440 °C (Axelsson et al., 2014; Elders et al., 2014; Fridleifsson et al., 2014).

Low total concentrations of dissolved solids in meteoric derived geothermal fluids are attributed to the low concentration of Cl (Stefánsson et al., 2017) derived from basalt dissolution and magmatic gases (Stefánsson and Barnes, 2016). Slightly elevated concentrations of Cl, however, occur in some hydrothermal systems hosted in rhyolite like Geysir and Torfajökull (Stefánsson et al., 2017). Silica is commonly the most abundant dissolved solid, Na the most abundant cation, and dissolved gases such as CO_2, H_2S, and H_2 may be locally present in concentrations exceeding all other dissolved components (Arnórsson, 1995). In typical meteoric-dominated systems, fluid components such as H_2S and CO_2 are commonly derived from water-basalt interactions (Stefánsson et al., 2017). Geothermal fluid compositions are modified to varying degrees by boiling and mixing with surface, glacial, and magmatic fluids, but the overall signature of dilute meteoric derived geothermal fluids is prevalent in high-temperature geothermal systems of the Neo-Volcanic Zone (Arnórsson, 1995; Pope et al., 2009, 2014).

Deep drilling in active geothermal systems reveals systematic hydrothermal mineral zoning with increasing depth and temperature, ranging from zeolite to amphibolite facies of hydrothermal metamorphism (see summaries in Arnórsson, 1995; Bird and Spieler, 2004; Franzson et al., 2008). With increasing depth and temperature, hydrothermal alteration of basalts have been characterized by the following zones: the Smectite-Zeolite Zone, Mixed Layer Clay Mineral Zone, Chlorite Zone, Chlorite-Epidote Zone, Epidote-Amphibole Zone, and Amphibole Zone. Similar hydrothermal mineral zones occur in fossil hydrothermal systems such as those at Breiddalur (Walker, 1963), the Reydarfjordur deep drill hole (Exley, 1982; Mehegan et al., 1982; Viereck et al., 1980) and Geitafell (Fridleifsson, 1983, 1984, 1986), all located in eastern Iceland (see Fig. 1).

The Lón area of southeastern Iceland (Fig. 1) contains chalcopyrite-galena-sphalerite mineralization in rhyolitic breccia pipes that are exposed over areas of 100 s of m^2 (Jankovic, 1970a). Average reported concentrations from rock chip line samples for Cu, Pb, and Zn in Lón are 1800 ppm, 800 ppm, and 700 ppm, respectively (Jankovic, 1970a). Similar Cu-Pb-Zn minerals are hosted in minor quantities in mm-scale fractures in felsic dikes in Fossadalur, several km from the Lón area, and the Thverá, Njörfatindar, and Reydarfjall areas proximal to Lón rhyolites that have Cu-Pb-Zn concentrations up to 1000 ppm (Jankovic, 1970b).

The Lón area (Fig. 1) is within the Southeast Icelandic Intrusives Suite (Padilla, 2015). This area of southeastern Iceland (Fig. 1B–C) has a long history of silicic magmatism and includes the 3.7 Ma Vesturhorn silicic intrusive complex (Martin et al., 2011), 6.4 Ma Austurhorn silicic intrusion (Padilla et al., 2014), and the ~7.3 Ma Reydarfjall silicic intrusion (Padilla, 2015). Previous work on Austurhorn, the most thoroughly studied silicic body in southeastern Iceland, suggested that the silicic magmas had been produced by fractional crystallization (Furman et al., 1992). More recent work has shown that the silicic magmas of Austurhorn, as well as those of the proximal intrusions of Reydarfjall and Slaufrudalur, were produced by anatectic dehydration melting of hydrothermally altered basalt (Padilla et al., 2014; Padilla, 2015), much like the majority of silicic magmas in Iceland (Bindeman et al., 2008, 2012; Pope et al., 2013), which range in $\delta^{18}O$ from −1 to 10‰ with a mean value of ~3‰ (Pope et al., 2013). Accordingly, primary magmas from the mantle ascended through the Icelandic crust