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# RESEARCH LETTER

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#### **Kev Points:**

- Correlation of oxygen isotopes, H<sub>2</sub>O and P-T-fO<sub>2</sub> for Icelandic rhyolites
- Tectonic setting controls origin of "hot" (rift) and "cold" (off-rift) rhyolites
- Hot rift silicic magmas are possbile analogues for embryonic continental crust

#### **Supporting Information:**

- Readme
- · Supporting Information

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# Contrasting conditions of rift and off-rift silicic magma origin on Iceland

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**Abstract** Factors controlling the origin of silicic magmas on Iceland are poorly constrained. Here we present new data on  $H_2O$  content, pressure, temperature, oxygen fugacity, and oxygen isotope composition of rhyolites from Askja, Öræfajökull, and Hekla volcanoes. All these parameters correlate with tectonic (rift and off-rift) setting of the volcanoes. Askja rift rhyolites originate through extensive assimilation of high-temperature hydrothermally altered crust ( $\delta^{18}O < 2\%$ ) at shallow depths ( $\geq 1.8$  km). These rhyolites are hot (935–1008°C), relatively dry ( $H_2O < 2.7$  wt%), and oxidized (QFM = +1.4). Cooler (874–902°C), wet ( $H_2O = 4$ -6.3 wt%), and non-oxidized ( $\sim$ QFM to QFM-1) off-rift rhyolites (Öræfajökull, Hekla) originate through differentiation deeper in the crust ( $\geq 4$  km) with almost no or little assimilation of high-T, altered crust, as reflected by slightly lower to normal  $\delta^{18}O$  values (5.2–6‰). Although off-rift rhyolites predominate during the Holocene, older silicic rocks on Iceland primarily formed in a rift setting possibly analogous to the oldest continental crust on Earth.

#### 1. Introduction

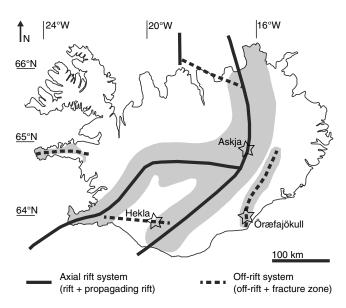
The origin of silicic magmas on Iceland, which is formed by a mantle plume interacting with a mid-ocean ridge, is widely debated. Fractional crystallization of primitive basalts and partial melting of hydrothermally altered basaltic crust are the two end-member models [Bindeman et al., 2012; Brophy, 2008; Hards et al., 2000; Jónasson, 2007; Kokfelt et al., 2009; Kuritani et al., 2011; Lacasse et al., 2007; Martin and Sigmarsson, 2007]. This long-lasting debate has been primarily focused on isotopic evidence for and against crustal assimilation. Only a few studies have provided data on the physicochemical conditions of the silicic magma origin, such as pressure (P), temperature (T), oxygen fugacity (fO<sub>2</sub>) [Gunnarsson et al., 1998; Portnyagin et al., 2012; Sigurdsson and Sparks, 1981], and pre-eruptive volatile content [Owen et al., 2013; Portnyagin et al., 2012] in Icelandic silicic magmas. The presently available data are insufficient to characterize the physicochemical conditions of the magma origin in relation to tectonic setting on Iceland. Here we evaluate the origin of silicic magmas formed in rift (e.g., Askja) and off-rift (e.g., Öræfajökull and Hekla) zones (Figure 1). Rift zones, where new crust is being generated through crustal spreading, are characterized by a high geothermal gradient due to the high magma flux. Magma flux and thus also geothermal gradient are much lower in off-rift zones, located beyond the influence of rift zone magmatism at the time of silicic volcanism [Martin and Sigmarsson, 2010].

Since proto-continents may have originated from oceanic plateaus, in some cases with islands, before the initiation of plate tectonics, Iceland with its thickened crust comprising a significant (up to 10% [Jónasson, 2007]) amount of silicic rocks provides the closest analogue to the earliest continental crust on Earth [Reimink et al., 2014]. Therefore, physicochemical conditions of silicic magma origin on Iceland may be similar to those during the proto-continental crust formation, which are otherwise difficult to estimate, and thus provide useful insights into the earliest Earth history.

Based on compositions of melt inclusions, matrix glasses, and minerals, we estimated pre-eruptive volatile content,  $fO_2$ , P, T, and magma  $\delta^{18}O$  for two of the largest historic silicic eruptions on Iceland, 1875 A.D. Askja and 1362 A.D. Öræfajökull. These new data together with published results on Hekla H3 and H4 eruptions [Portnyagin et al., 2012] are used to elucidate possible correlations of the physicochemical parameters with magma composition and tectonic setting, and to propose a generalized model of silicic magma origin on Iceland potentially applicable to other regions of long-lived ocean-island volcanism during Earth history.

#### 2. Samples and Methods

This study focused on tephra fragments which were quickly quenched during eruption and thus provide the best possible material for studying phase relationships in silicic magmas before eruption, which can be reset



**Figure 1.** Map of Iceland with the neo-volcanic zone in gray and volcanic centers from this study given as stars.

by slow cooling. Tephra from the Plinian 1875 A.D. Askja eruption was sampled near Víti crater (Figure 1, N 65°2.78′ W 16° 43.25′). Samples of rhyolite pumice of the 1362 A.D. Öræfajökull eruption were collected from the southeastern foot of the volcano from the middle part of a 2 m thick tephra layer (Figure 1, N 63°53.52′ W 16°37.15′).

The samples studied were highly vesicular (75–85 vol%), sparsely phyric (Askja <0.5%, Öræfajökull 1–3% phenocrysts) pumice bombs of rhyolitic composition. Mineral phases are plagioclase, clinopyroxene, fayalitic olivine (in Öræfajökull), orthopyroxene (in Askja), ilmenite, magnetite, apatite, pyrhotite, and zircon (in Öræfajökull). Öræfajökull rhyolites are mineralogically very similar to Hekla rhyolites [*Portnyagin et al.*, 2012], whereas the Askja rhyolites are distinguished by the presence of orthopyroxene.

We performed a comprehensive microanalytical survey, which included analysis of all major mineral phases for major elements and volatiles in matrix glasses and melt inclusions (n = 160) hosted in plagioclase, pyroxene, and olivine by electron microprobe (major elements, S, Cl) and for selected inclusions by secondary-ion mass spectrometry (SIMS:  $H_2O$ ) and Fourier transform infrared spectroscopy (FTIR:  $CO_2$ ). The data were used to quantify the pre-eruptive volatile content, P, T, and  $FO_2$  conditions of the magmas. Oxygen isotope composition was analyzed by laser fluorination for phenocryst phases and glasses from the studied tephras. See the Supporting Information for analytical techniques and the complete data set.

#### 3. Results

Major elements of matrix glasses and melt inclusions are extremely homogenous for Öræfajökull but more heterogeneous in Askja magmas (Figure 2). In Öræfajökull pumice, melt inclusions ( $SiO_2 = 67.6-72.5$  wt%, n = 81) are found in extremely Fe-rich olivine ( $Fo_2$ ), plagioclase ( $An_{14-19}Ab_{74-78}Or_{7-9}$ ) and clinopyroxene ( $En_{3-5}Fs_{50-53}$  Wo<sub>44-45</sub>) and have major element compositions very close to that of matrix glass ( $SiO_2 = 68.6-72.0$  wt%, n = 21) (Figure 2), indicating entrapment of the host melt. Askja melt inclusions ( $SiO_2 = 69.2-75.7$  wt%, n = 79) in plagioclase ( $An_{43-61}Ab_{37-54}Or_{1-3}$ ), clinopyroxene ( $En_{39-41}Fs_{19-25}Wo_{38-41}$ ), and orthopyroxene ( $En_{59-61}Fs_{36-37}Wo_{3-4}$ ) are more and less evolved than matrix glasses ( $SiO_2 = 70.1-73.6$  wt%, n = 25) (Figure 2), indicating fractional crystallization associated with magma mixing prior to the 1875 Askja eruption.

Equilibration temperatures were calculated for Askja and Öræfajökull rhyolites with the help of mineral-melt [*Putirka*, 2008] and magnetite-ilmenite [*Andersen et al.*, 1993] thermometers (Figure 3a). For Askja the thermometers yielded 935–1008°C (plagioclase-melt) and 886–993°C (ilmenite-magnetite). For Öræfajökull temperatures are 874–902°C (olivine-melt) and 836–928°C (ilmenite-magnetite) and tend to be lower than temperatures calculated for Askja. The temperatures for Öræfajökull rhyolites have similar temperatures to rhyolites from Hekla [830–880°C; *Portnyagin et al.*, 2012].

Concentrations of  $H_2O$  in melt inclusions determined with SIMS range from 0.1 to 2.7 wt% in Askja inclusions and 2.1 to 4.2 wt% in Öræfajökull inclusions (Figure 3b).  $CO_2$  was not detected in glasses. The highest  $H_2O$  concentrations in melt inclusions are considered to be minimum values and to provide the best estimate of pre-eruptive magma  $H_2O$  content, since magma degassing during ascent and possible diffusive water loss from inclusions [Portnyagin et al., 2008] can decrease the initial concentrations. Our measured water content in Askja melt inclusions is similar to  $H_2O$  reported in plagioclase-hosted melt inclusions ( $\leq$ 1.9 wt%)

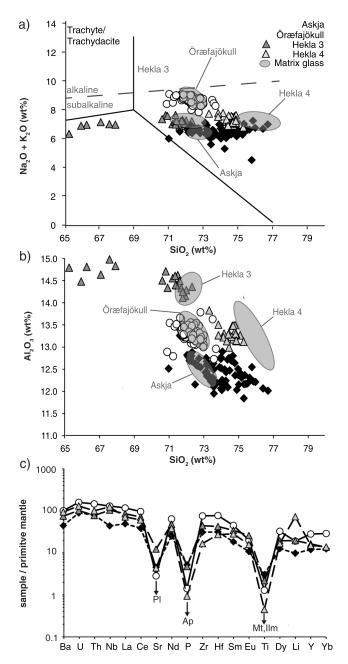


Figure 2. Major and trace element composition of melt inclusions and matrix glasses from Icelandic rhyolites. Both matrix glasses and melt inclusions have predominantly rhyolitic compositions (a). Close composition of matrix glasses and melt inclusions (a and b) indicates that no significant crystallization took place during magma accent and eruption. Matrix glasses were, however, significantly degassed with respect to H<sub>2</sub>O and likely CO<sub>2</sub> yet melt inclusions possibly preserved initial concentrations of volatiles. Lower Al<sub>2</sub>O<sub>3</sub> values in Askja melts compared to Hekla and Öræfajökull at given SiO<sub>2</sub> (b) correlate with lower primary water in Askja melt inclusions and testify that volatile content in melt inclusions is informative of pristine volatile content in magmas at depth. Similar Al<sub>2</sub>O<sub>3</sub> in matrix glasses and plagioclase-hosted melt inclusions suggest that no significant crystallization of plagioclase took place after entrapment of these inclusions. The Icelandic rhyolites have generally similar yet slightly distinctive patterns of trace elements for different volcanoes (plot c), all with deep minima of Sr, P, and Ti indicating strong control of plagioclase (Sr), apatite (P), and Fe-Ti oxides (Ti) on their composition. Data for Hekla are from Portnyagin et al. [2012]. Primitive mantle values are after Mcdonough and Sun [1995].

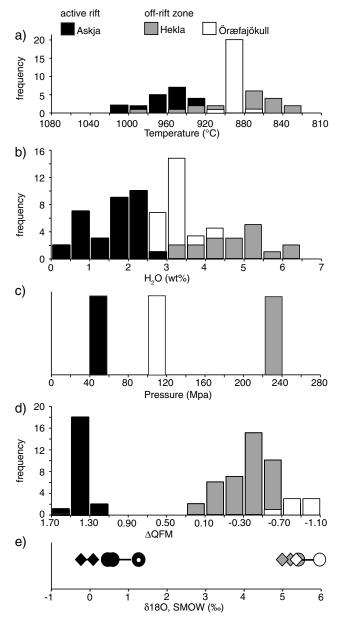


Figure 3. The range of physicochemical conditions and compositional parameters for silicic Icelandic magmas. (a) Plagioclase-liquid (Askja) and olivine-liquid temperatures (Hekla, Öræfajökull) in the presence of H<sub>2</sub>O [Putirka, 2008]. (b) H<sub>2</sub>O content in melt inclusions measured with SIMS, (c) equilibrium pressures calculated with VolatileCalc [Newman and Lowenstern, 2002], (d) oxygen fugacity estimated with coexisting ilmenite-magnetite crystals [Andersen et al., 1993], and (e)  $\delta^{18}$ O compositions of plagioclase (diamonds) and matrix glasses (circles) measured by laser fluorination. Data for Askja rhyolites are from this study (white dot) and from Bindeman et al. [2012].

from Clark [2012]. For Öræfajökull our data indicate much higher H<sub>2</sub>O values than reported previously on melt inclusions in pumice [≤2 wt%, Sharma et al., 2008].

The minimum crystallization depths were determined from the composition of melt inclusions with the highest H<sub>2</sub>O content by estimating the pressure of saturation of the melts with pure H<sub>2</sub>O fluid during the entrapment [Newman and Lowenstern, 2002]. The minimum pressures are estimated to be 51 MPa for Askja and 117 MPa for Öræfajökull (Figure 3c), which correspond to depths of >1.8 km for Askja and >4 km for Öræfajökull. If 100 ppm of CO<sub>2</sub> was present in the magmas (detection limit of our FTIR analyses), the last pressure of equilibration would be 15 MPa (or 500 m depth) greater. These depths are at the shallow limit for the depth of magma chambers from geophysical data under these volcanoes [Kelley and Barton, 2008], as is expected for the last depths of equilibration of magmas.

Oxygen fugacity during magma crystallization was estimated from the composition of magnetite and ilmenite inclusions coexisting in clinopyroxene and olivine phenocrysts [Andersen et al., 1993] and is reported as ΔQFM, reflecting the deviation of logfO2 from quartz-fayalitemagnetite equilibria for a specific temperature also estimated from magnetite-ilmenite equilibria. The data indicate contrasting redox conditions for Askja and Öræfajökull. While Öræfajökull melts crystallized at fO2 below quartzfayalite-magnetite (QFM) equilibria  $(\Delta QFM = -0.9 \pm 0.2, 1s.d., n = 7)$ , Askja melts are more oxidized ( $\Delta QFM = 1.4 \pm 0.1$ , 1s.d., n = 21) (Figure 3d).

Oxygen isotopes were determined on plagioclase phenocrysts and fresh matrix glasses. Rhyolites, formed by fractional

crystallization of MORB-like basaltic melts, have  $\delta^{18}$ O glass ~ 6% and plag ~ 5.7% [Bindeman et al., 2008]. Öræfajökull exhibit normal  $\delta^{18}$ O values (6.0% in glass, 5.4% in plagioclase) and Hekla slightly lower than normal  $\delta^{18}$ O (H3: 5.2% in plagioclase; H4: 5.5% in glass, 5.0% in plagioclase). Askja samples have very low  $\delta^{18}$ O values (1.3‰ in glass), in good agreement with recent data from *Bindeman et al.* [2012] requiring shallow crustal re-melting of zircon-saturated low- $\delta^{18}$ O silicic rocks (Figure 3e).



#### 4. Discussion

#### 4.1. Water-Rich and Water-Poor Icelandic Silicic Magmas

Even though Icelandic rhyolites have often been described as "dry" due to the typical absence of amphibole in their phenocryst assemblage [Jónasson, 2007], recent melt inclusion studies showed that the magmas have much higher water content than previously believed [Hekla ≥6.2 wt%; Portnyagin et al., 2012; Torfajökull, ≥4.8 wt%; Owen et al., 2013]. Our new data from Askja (≥2.7 wt% H<sub>2</sub>O) and Öræfajökull (≥4.2 wt% H<sub>2</sub>O) show that the range of  $H_2O$  content in silicic Icelandic magmas is large. Taking all available data for silicic magmas into account, Hekla has the highest H<sub>2</sub>O, Torfajökull and Öræfajökull have intermediate H<sub>2</sub>O, and Askja has the lowest H<sub>2</sub>O measured directly in melt inclusions. By analogy with explosive island-arc volcanism with high pre-eruptive H<sub>2</sub>O content in the magmas, the high water contents in the Icelandic silicic magmas drove these highly explosive eruptions and not interaction with ice or ground waters [Portnyagin et al., 2012].

Partial melting of hydrated basalts or hydrothermally altered silicic material could produce silicic rocks on Iceland [Bindeman et al., 2012; Gautason and Muehlenbachs, 1998; Hattori and Muehlenbachs, 1982; Macdonald et al., 1987; Martin and Sigmarsson, 2007]. Meteoric waters and hydrothermally altered rocks on Iceland have low  $\delta^{18}$ O [Bindeman et al., 2012; Gautason and Muehlenbachs, 1998]. Interaction of magmas with hydrothermally altered (and thus likely hydrated) crust in Iceland is therefore expected to produce H<sub>2</sub>O-richer rhyolites with low  $\delta^{18}$ O. Very low  $\delta^{18}$ O in Askja rhyolites provide strong evidence for a large contribution (tens of %) from shallow crustal melting of low-ô<sup>18</sup>O, hydrothermally altered crust to explain isotopic mass balance [Bindeman et al., 2012]. Therefore, the relatively low pre-eruptive H<sub>2</sub>O content in the rhyolites is unexpected.

Because H<sub>2</sub>O solubility in magmas depends strongly on pressure [Moore et al., 1995], the paradoxically low  $H_2O$  contents of low- $\delta^{18}O$  Askia magmas are likely to reflect  $H_2O$  saturation at low pressure and therefore shallow silicic magma storage (≥1.8 km) beneath the rift. In contrast, Hekla fractionated at ~8 km depth [Portnyagin et al., 2012]. Higher pressures favored higher water solubility, resulting in higher melt water contents [Portnyagin et al., 2012]. Therefore, H<sub>2</sub>O content in Icelandic magmas appears to largely reflect magma storage depths and does not serve as a good proxy for the amount of assimilated hydrated material. The low- $\delta^{18}$ O values in magmas thus serve as a barometer, suggesting shallow storage conditions.

#### 4.2. Reduced and Oxidized Icelandic Magmas

Icelandic rhyolite magmas are thought to be reduced, having crystallized at oxygen fugacity close to or below QFM [Carmichael, 1991]. In this respect, the magmas are clearly different from typical island-arc rhyolites, which are normally strongly oxidized ( $fO_2 > QFM = +1$ ) [Ghiorso and Evans, 2008]. Based on high H<sub>2</sub>O but low fO<sub>2</sub> in Hekla magmas, Portnyagin et al. [2012] proposed that the major difference between Icelandic and island-arc silicic magmas is in oxygen-redox conditions, not in water content of the magmas. Our new data, however, show that redox conditions of Icelandic rhyolites are not uniformly reduced, as previously thought.

The off-rift Öræfajökull rhyolites crystallized at reduced conditions ( $\Delta QFM = -0.9 \pm 0.2$ ), even more reduced than Hekla rhyolites [H3:  $\Delta QFM = -0.4 \pm 0.2$ , H4:  $\Delta QFM = -0.1 \pm 0.3$ ; Portnyagin et al., 2012]. Reduced conditions appear to be typical for Iceland and likely reflect the redox conditions in deep Icelandic crust and that of mantle-derived basalts.

In contrast, magnetite-ilmenite pairs from Askja pumice indicate oxidized conditions ( $\Delta QFM = 1.4 \pm 0.1$ ). Late-stage oxidation of Askja magmas has been proposed on the basis of Fe<sup>+2</sup>/Fe<sup>+3</sup> Mössbauer spectroscopy measurements in glasses [Helgason et al., 1992]. This process, however, cannot alter the composition of ilmenite and magnetite inclusions in silicate minerals, which we measured in this study, that are protected from later oxidation. Therefore, the oxidation state estimated in coexisting magnetite and ilmenite pairs is likely to reflect pristine compositions of the Askja magma before eruption.

ΔQFM values estimated for Icelandic silicic magmas and oxygen isotope composition of minerals and glasses appear to be very different for rift and off-rift systems. While rift melts (Askja) are oxidized ( $\Delta QFM = 1.4$ ) and have low  $\delta^{18}$ O, reflecting assimilation of hydrothermally altered, more oxidized shallow crust, off-rift melts (Hekla, Öræfajökull) are reduced ( $\Delta$ QFM to QFM -1) and show normal to slightly lowered oxygen isotope values (5.2-6%). Oxygen isotope composition of Icelandic silicic rocks is widely believed to reflect the extent of assimilation of hydrothermally altered crust [Bindeman et al., 2012; Condomines et al., 1983; Hattori and Muehlenbachs, 1982; Martin and Sigmarsson, 2010; Muehlenbachs et al., 1974; Sigmarsson et al., 1991]. Our

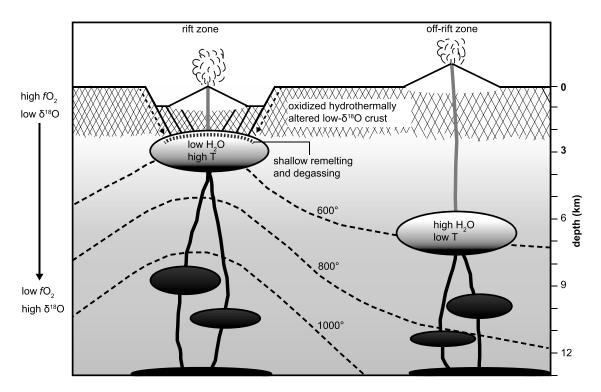


Figure 4. Model for origin of silicic magmas in rift and off-rift settings on Iceland. Isotherms are shown by using data from *Jones and Maclennan* [2005]. See details in text.

study documents for the first time that this process is also responsible for the large variations in the redox state of Icelandic silicic magmas.

Our data also provide evidence that oxygen fugacity exerts a major control on the stability of olivine and orthopyroxene in silicic magmas. Olivine is stable in magmas crystallizing at  $fO_2 < QFM$  (Hekla, Öræfajökull) and orthopyroxene at  $fO_2 > QFM$  (Askja), in agreement with data from quartz-bearing rhyolites [Ghiorso and Sack, 1991].

# 4.3. Tectonic Control on the Conditions of Silicic Magma Origin

Martin and Sigmarsson [2010] proposed a tectonic control on the conditions of silicic magma origin based on oxygen and radiogenic isotopes. They proposed that silicic Icelandic magmas in rift zones originate from partial melting of hydrated metabasaltic crust followed by fractional crystallization ± assimilation, whereas off-rift volcanoes generate silicic magmas mainly by fractional crystallization of basaltic magmas, due to the lower geothermal gradient. Only the present tectonic setting, however, is relevant for the youngest Quaternary silicic magmas from a center, since older rocks may have formed in a different setting, as demonstrated for Snæfellsnes Peninsula where lavas >5.5 Ma formed in a rift setting and younger lavas in an off-rift setting [Martin and Sigmarsson, 2010].

Here we demonstrate that the physicochemical conditions of silicic magma origin are related to geotectonic setting (Figure 4). Rift rhyolites from Askja originate at high T (>900°C) in a shallow ( $\geq$ 1.8 km) magma chamber and exhibit strong evidence (low  $\delta^{18}$ O and high  $fO_2$ ) for extensive interaction with high-T, hydrothermally altered and oxidized crust (Figure 4). Recent data on oxygen isotope composition of olivine from Askja basalts [*Hartley et al.*, 2013] suggest that Askja magmas assimilate hydrothermally altered crust already during early stages of fractionation. It is therefore likely that rift magma series ranging from basalts to rhyolites, as exemplified by Askja, originate by assimilation and fractional crystallization (AFC) processes [*Sigurdsson and Sparks*, 1981] at relatively high assimilation rate (0.1–0.3, assuming low  $\delta^{18}$ O of altered crust and 90% fractionation from basalt), favored by the high geothermal gradient under rift zones [*Martin and Sigmarsson*, 2010].

In contrast, recent water-rich rhyolites in off-rift zones (Hekla, Öræfajökull) originated at lower T (<900°C) and deeper depths (up to 8 km) (Figure 4). The magmas show only modest or no obvious signs of interaction with metabasaltic crust as evident from their near-normal  $\delta^{18}$ O and low  $fO_2 \le QFM$ . Even though deeper depths

are generally more favorable for crustal recycling [Reiners et al., 1995], it is more difficult for cooler off-rift magmas to melt and assimilate unaltered (dry) metabasaltic and gabbroic rocks than for hotter rift lavas to assimilate altered metabasalt on Iceland. At Öræfajökull we can exclude major AFC, since the crust beneath Öræfajökull has a different isotopic composition from the Quaternary basalts and rhyolites, which have identical isotopic compositions. Assimilation to form Öræfajökull rhyolites is also inconsistent with the extreme heterogeneity of the rhyolites. Therefore, we propose that silicic magmas in off-rift zones, which have low geothermal gradients, are primarily formed through fractional crystallization of initially low-H<sub>2</sub>O and low  $fO_2$  basaltic parental magmas at low assimilation rates (<0.1).

### 5. Conclusion and Implications

Different tectonic settings on Iceland appear to exert a primary control on the location of silicic magma chambers in the crust and on the chemistry of the silicic magmas. Shallow chambers with hot and low-H<sub>2</sub>O magma may be favored under rift zones and deeper chambers with colder and H<sub>2</sub>O-rich magmas under off-rift volcanoes. The Icelandic crust appears to be strongly zoned having low- $\delta^{18}$ O and being oxidized at shallow depths. The silicic magmas formed under the rift evolve in an open system and appear to assimilate hydrothermally altered metabasaltic crustal rocks, which changes both the oxygen isotope composition and redox state of the silicic magmas. Our results therefore show that P-T-fO2 conditions of silicic magma on Iceland are strongly related to their tectonic setting, composition, and the thermal and oxidation state of the crust.

Improved understanding of silicic magmatism can not only help decipher past geodynamic settings on Iceland, characterized by rift- and off-rift zones, but also may be applicable to formation of rhyolites on other ocean island with thickened crust formed through plume-ridge interaction (e.g., Galapagos Islands) and possibly to formation of the embryonic continental crust [Reimink et al., 2014].

Tonalitic gneisses from the Acasta Complex in Canada afforded a rare opportunity for Reimink et al. [2014] to investigate the composition and possible conditions of origin of pre-4.0 Gyr silicic crust. Unlike tonalitetrondhjemite-granodiorite (TTG) suites predominating in Archaean complexes, the oldest known silicic rock units from the Acasta Complex have compositions similar to Icelandic andesites (icelandites) and are proposed to have formed via magmatic assimilation of preexisting high-T altered and oxidized basaltic crust at shallow depths. Thus, our new data for rift-related Icelandic rhyolites (Askja) may provide a possible estimate of P, fO<sub>2</sub>, and minimum T conditions for the proto-continental crust, which are otherwise difficult to retrieve directly from relic zircons in strongly and multiply metamorphosed Archaean rocks.

## Acknowledgments

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