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Fluid Inclusions in the Deepest Part of the IDDP-2 Drill Core, Reykjanes Peninsula, Iceland

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ABSTRACT

We studied geothermal fluid, trapped as fluid inclusions in felsic minerals, from the bottom (>4500 m) of the IDDP-2 borehole, Iceland. Petrographic observations suggest that the fluid at this depth is separated into a vapor phase and a solute-rich brine. Based on fluid inclusion homogenization experiments, the temperature of the geothermal fluid is estimated to be 600 ± 20 °C and the pressure corresponds to near-hydrostatic conditions (~45 MPa) at the bottom of the borehole. The vapor phase is dominated by H_2O, containing minor CO_2, H_2S and H_2, whereas the solute-rich brine is dominated by Fe-K-chloride-hydroxides at room temperature and can contain up to 1.2 wt% Cu in the form of Cu-Fe-sulfide. Potassic silicate melt inclusions associated with the geothermal fluid are also present and they are the witnesses of melt-fluid interactions. Furthermore, they indicate that there is a danger of drilling into magma at similar depths at the Reykjanes peninsula.

1. INTRODUCTION

The Icelandic Deep Drilling Project (IDDP) investigates the economic potential of geothermal fluids at the deep roots of active geothermal systems (Friðleifsson et al., in press). This exploration includes investigating the feasibility of exploiting supercritical fluid for energy production and the extraction of economically valuable materials, such as base metals, directly from this fluid. Model calculations suggest that the power output of a geothermal well that uses superheated steam of supercritical origin can be an order of magnitude higher than the conventional geothermal wells (e.g., Albertsson et al., 2003). Furthermore, it is well-known that supercritical fluids might transport economically valuable elements such as Cu and Au in high quantities (e.g., Loucks and Mavrogenes, 1999, Hack and Mavrogenes, 2006) which, upon phase separation of the fluid, can precipitate to form high grade ore deposits.

The Reykjanes geothermal system is situated in southwest Iceland and is the surface manifestation of the processes operating in the Mid-Atlantic Ridge. The chemical composition of the geothermal fluid at shallow depth (<3000 m) is close to that of sea water (e.g., Arnórsson, 1978) with trace metal contents similar to or higher than those in black smokers (e.g., Harðardóttir et al., 2012, 2013). The IDDP-2 borehole was deepened in the Reykjanes geothermal field in 2016-2017 and reached ~4500 m vertical depth (total slant depth 4659 m, Friðleifsson et al., in press). The borehole was the continuation of the already existing production hole RN15, which originally was drilled to 2.5 km depth (Friðleifsson et al., in press). Drill cores were taken from depth ranges of 3000-3200 m, 4250-4310 m and 4630-4659 m for geochemical and petrophysical studies (Friðleifsson et al., in press). Due to the total loss of circulation below 3.1 km depth, these cores are the only rock samples available from the deeper part of this borehole.

This study focuses on the examination of fluid inclusions (small droplets of geothermal fluid trapped in minerals) from the bottom part (4630-4640 m) of the borehole in order to determine the physical state, temperature and composition of the geothermal fluid.

2. METHODS

2.1 Microthermometry

Microthermometric analyses were carried out using Linkam TMS600 and Linkam TS1500 heating stages at the Lithosphere Fluid Research lab, Faculty of Science, Eötvös University, Budapest. Doubly polished mineral chips were heated at a rate of 10°C/min to approximately 20°C below the phase transformation temperatures, after which the heating rate was changed to 5°C/min. The precision of the temperature readings of the TMS600 and TS1500 stages is 0.2° and 2°C, respectively. Phase transformations were determined by cycling the temperature 10°C below and above the phase transformation temperatures at a heating rate of 1°C/min. Microthermometric analyses were carried out on four samples (4635.11, A4637.80, N4635.25, G4634.48, where the numbers represent the slant depth in meters below the rig floor). In N4635.25, A4637.80 and G4634.48 we observed phase transformations and homogenization behavior of the solid-rich brine inclusions, whereas in 4635.11 and G4634.48 phase transformations were observed in the vapor-rich inclusions.

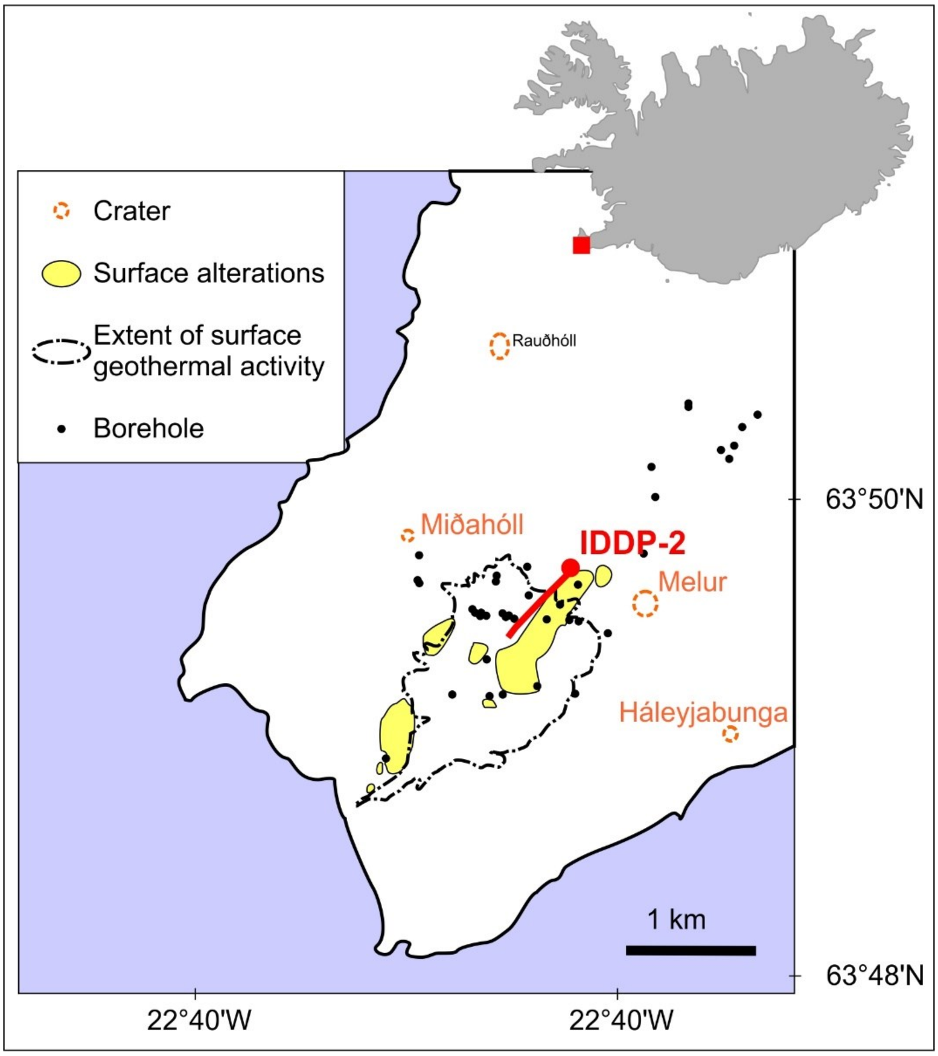

Figure 1: The location of the IDDP-2 borehole (the position and the direction of the drilling indicated in red) in the Reykjanes geothermal system, Iceland based on Freiðleifsson et al. (in press) and references therein.

2.2 Raman Microspectrometry

Raman microspectrometric analyses were carried out at the Research and Instrument Core Facility of the Faculty of Science, Eötvös University, Budapest (ELTE FS-RICF), at room temperature and during the heating experiments. During room temperature analyses, a 100x objective was used, whereas during high temperature measurements we used a 50x objective lens. Analyses were carried out by a Horiba LabRam HR spectrometer, applying a 532 nm (green) laser with a maximum laser energy of 130 W, 100 μm confocal pinhole, and 600 to 1800 grooves/mm gratings. Analyses ran for 30 seconds with 2-3 repetitions.

2.3 Electron Microprobe Analyses

Electron microprobe analyses of silicate melt inclusions were carried out by Jeol JXA-8230 SuperProbe at the Institute of Earth Sciences, University of Iceland. Operating conditions were: accelerating voltage of 15 kV, beam current of 5 nA, and a beam size of 4 to 7 μm, depending on melt inclusion size. Analyses were carried out by WDS spectrometry, where Na was analyzed first to minimize the effect of the electron beam. Natural and synthetic standards provided by Astimex and the Smitsonian Institute were used for calibration. Additionally, we used ATHO rhyolitic glass as secondary standard.

2.4 Focused Ion Beam Scanning Electron Microscopy with Energy Dispersive Spectrometry– Slice and View (FIB-SEM-EDS)

The FIB-SEM-EDS analyses were carried out on a FEI Quanta 3D Dual Beam SEM at the Research and Instrument Core Facility of the Faculty of Science, Eötvös University, Budapest (ELTE FS-RICF). The EDS analyses and SEM imaging were done using 15-20 kV acceleration voltage and 23.7-480- pA beam current, depending on magnification. The surface above the selected fluid inclusion was coated with a ~1μm layer of platinum in order to protect the inclusion from the high current Ga^+ ions used for milling the three trenches around the inclusion. Low current Ga^+ ions were used for slicing the inclusion; the distance between the slices were ~150 to 250 nm, depending on the suspected size of the smallest solid phases within the inclusions previously identified by Raman mapping. High resolution secondary electron (SE) and backscattered electron (BSE) images were taken on every newly exposed surface (slice) of the inclusion in order to calculate the exact volume proportion of the different phases. Exposed solid phases were identified using point or area measurements with a high resolution EDAX SDD (Silicon Drift Detector).

3 RESULTS

3.1 Petrography and Composition of Fluid Inclusions at Room Temperature

Fluid inclusions were studied in felsic segregation veins (Zierenberg et al., 2020) composed of plagioclase+quartz+biotite+opaque(s). Based on petrographic observations, we distinguished both primary and secondary (Roeder, 1984) fluid inclusion assemblages (FIAs) in plagioclase and quartz. Primary FIAs occur along growth zones of the minerals, whereas secondary FIAs form planes along healed cracks (Figure 2a). These planes crosscut grain boundaries. Therefore, one can assume that they formed after the crystallization of the felsic veins. As the secondary FIAs are more representative of the current geothermal fluid than the primary ones, this study focuses only on their composition. We observed three types of inclusions in the secondary FIA planes, which we refer to as Type1, Type2 and Type3 inclusions throughout the text.

Type1 inclusions are vapor-rich and are generally composed of a large dark vapor bubble surrounded by a thin liquid film (Figure 2b). Occasionally, these inclusions contain a small opaque phase. At room temperature the bubble phase in these inclusions is dominated by CO_2 with minor H_2S and traces of H_2, whereas the liquid film is low salinity or pure water. Based on the volume proportion of the liquid film and the vapor bubble, and assuming that the liquid phase at room temperature is pure water, the density of the Type1 inclusions is estimated to be ~0.07 (±0.04) g/cm^3 based on 12 inclusions. The density is likely to be somewhat higher as the liquid probably contains minor chloride component(s).

Based on optical microscopy, Type2 inclusions are composed of four different solid phases, a vapor bubble, ± a minor liquid phase (Figure 2c, d). The shape of the vapor bubble in these inclusions is always irregular indicating that the liquid phase, if present, should be minor as the vapor fills the space remaining after the crystallization of the solid phases. Solid phase 1 (S1) is green to yellow in plane polarized light (Figure 2c) and is strongly anisotropic with crossed polars. At room temperature, this mineral has two characteristic Raman bands at 3451 and 200 cm^{-1}. Additional smaller bands are observed in some of the spectra at 140 and 385 cm^{-1}. The approximate stoichiometry of this phase is $(Fe,Mn)_3(Na,K)_2Cl_4(OH)_4$ based on FIB-SEM-EDS (Figure 2d). Solid phase 2 (S2) is also yellowish-green in plane polarized light (Figure 2c) and seems to be isotropic with crossed polars. This mineral has one characteristic Raman band measured at 3457 cm^{-1} and its approximate stoichiometry is $(Fe,Mn)(Na,K)_2Cl_3(OH)$ (Figure 2d). Solid phase 3 (S3) is an isotropic and transparent mineral (Figure 2c, d), commonly showing cubic crystal habit with no characteristic Raman bands and is a halite-sylvite solid solution with minor Fe- and Mn-contents, dominated by the halite component. Solid phase 4 (S4) (Figure 2c, d) is a Cu-Fe sulfide with characteristic Raman bands at 381, 405 and 450 cm^{-1}, which makes this mineral structurally similar to molybdenite (Windom et al., 2011). Based on FIB-SEM-EDS analyses, we identified further minor phases in these inclusions, such as Fe-dominated chloride-hydroxide (similar in composition to akageneite), a Ba- and a Ca-rich chloride phase, and apatite.

Type3 inclusions are silicate melt inclusions (MI, Figure 2e, f), which are composed of a colorless silicate glass and a vapor bubble. The size of the vapor bubble is variable in these inclusions and sometimes they contain S1 (Figure 2e). Type3 inclusions are rare and they are occasionally found in the secondary FIAs with Type1 and Type2 inclusions (Figure 2e). Based on electron microprobe analyses, these inclusions are rhyolitic in composition with SiO_2 content of 79 wt.%, total alkali content of 6 wt.%, and 0.27 wt% Cl. Their K_2O/Na_2O ratio is ~1.5. This is the first report of felsic melt compositions from the Reykjanes Volcanic System, and to our knowledge the first report of potassic ($K_2O_{mol}/Na_2O_{mol}>1$) melt compositions from Iceland.

3.2 The Temperature of the Geothermal Fluid Based on Heating Experiments

The fluid temperatures at the time of trapping of Type1 and Type2 inclusions were determined by heating experiments combined with Raman microspectrometry. In Type1 inclusions the liquid film has optically disappeared at 550°C. However, liquid H_2O was observed in the Raman spectra up to at least 590°C at the edges of each studied inclusion (Figure 3). No liquid water signal was observed in four out of five spectra at 600°C. However, in one fluid inclusion there is a broad shoulder at Raman shift of ~3540 cm^{-1}, corresponding to liquid H_2O. Based on these observations, we conclude that Type1 inclusions are homogenized into the vapor phase at a temperature of 600 ± 10°C.

Phase transformations in Type2 inclusions were observed in the following sequence: S1 dissolves in each inclusion at temperatures between 175 and 180°C, S2 dissolves between 220 and 240°C, S3 dissolves at 377 to 390°C, whereas S4 dissolves at 600 ± 20°C (Figure 4). At 600°C Type2 inclusions are composed of a vapor bubble and a liquid brine phase (Figure 4). Note that the size of the vapor bubble is significantly smaller at 600°C than at room temperature. None of the Type2 inclusions homogenized below 715°C. As phase transformations happen in very narrow temperature ranges in each studied brine inclusion (n=32 in 7 FIAs in 4 thin sections), we conclude that the volume proportions of these phases are similar in the brine, regardless of the sample studied. Furthermore, the dissolution of S4 solid phase in Type2 inclusions and the disappearance of the liquid film in Type1 inclusions happen at 600 ± 20°C, which suggest that the temperature of the geothermal fluid should be close to this value. Complete homogenization of the Type2 brine inclusions do not happen at this temperature as they contain accidently trapped vapor phase of the Type1 inclusions in various quantities due to the phase separation at 600°C at >4500 m depth. Independent mineral geothermometers from the paragenetically latest stage of mineralization and alteration also suggest present day in-situ temperature is close to 600°C (Zierenberg et al., 2020).

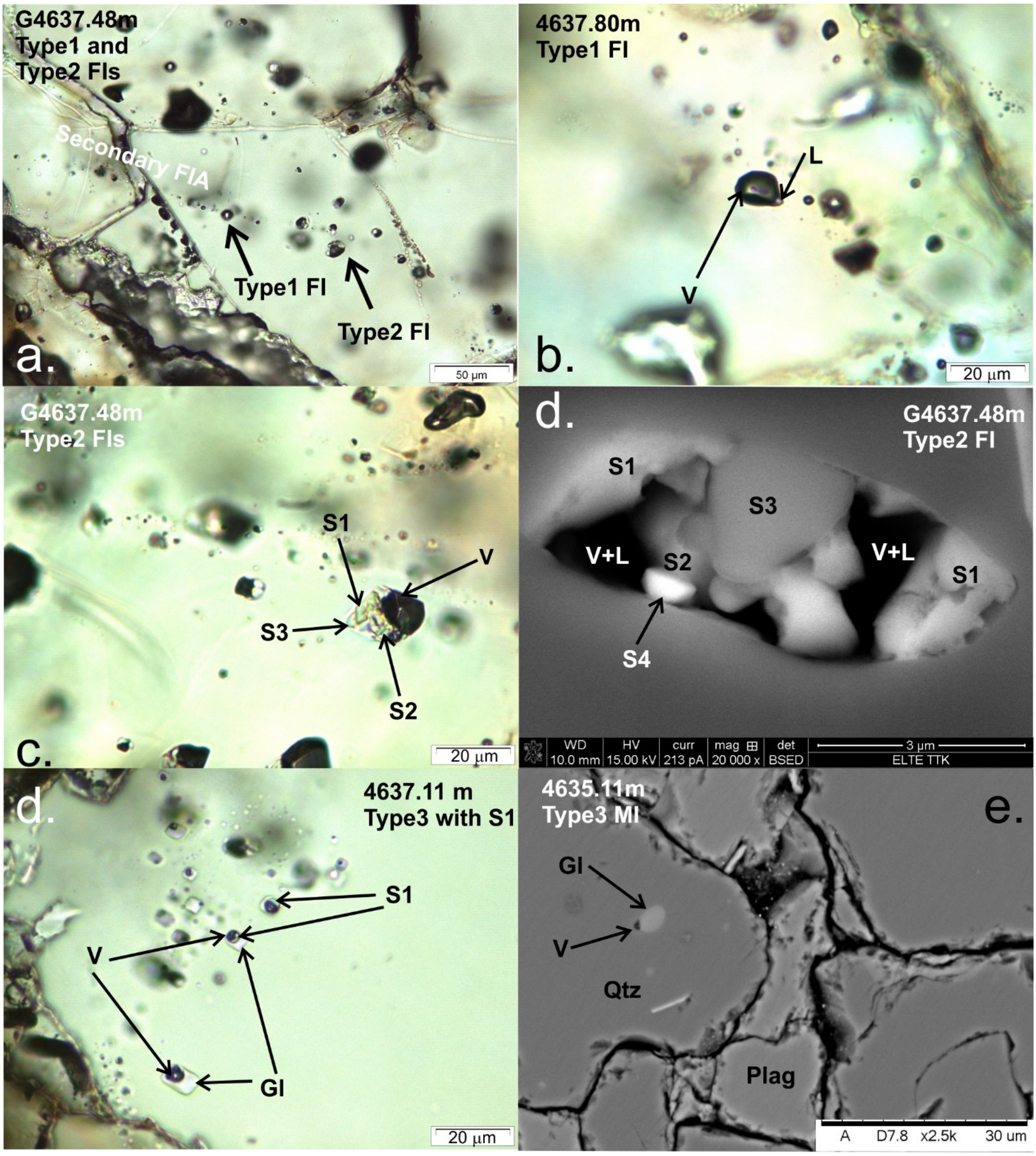

Figure 2: Petrographic features of fluid inclusions from felsic veins of the IDDP-2 borehole in the Reykjanes geothermal system, Iceland. a) Photomicrograph of Type1 and Type2 fluid inclusions (FI) in a secondary fluid inclusion plane, plane polarized light; b) Photomicrograph of Type2 fluid inclusion composed of liquid film (L) and vapor bubble (V), plane polarized light; c) Photomicrograph of Type2 fluid inclusion with various solid (S1-S3) and a vapor (V) phase, plane polarized light; d) Back scattered electron image of various solid phases (S1-S4) exposed in a Type2 fluid inclusion, FIB-SEM image; d) Photomicrograph of Type3 silicate melt inclusions composed of felsic silicate glass (Gl), Vapor (V) and S1 solid in quartz, plane polarized light; e) Back scattered electron image of Type3 melt inclusion (MI) in quartz (Qtz), Gl-silicate glass, V-vapor bubble, Plag-plagioclase.

3.3 The Composition of the Geothermal Fluid at >4500 m Depth

We determined the composition of the homogeneous Type1 inclusions based on the Raman spectra collected at 600°C using the method developed by Dubessy et al. (1989). These inclusions are dominated by H_2O vapor (~97.5 mol%), containing ~1.5 mol% CO_2, minor H_2S and traces of H_2.

Salinity in Type2 inclusions is estimated to be 45-46.6 wt% $NaCl_{equiv}$ based on halite (S3) dissolution temperatures (Bodnar and Vityk, 1994). A more precise determination of Type2 brine compositions was done based on the volume proportions and chemical composition (from FIB-SEM-EDS analyses) of each phase within these inclusions. As the amount of liquid water is difficult to determine with any of the techniques used, we calculated a maximum solute content. This was derived assuming all pore space observed in FIB-SEM images (Figure 2d) is low density vapor in these inclusions. As all inclusions contained a large vapor bubble and only minimal H_2O (Figure 2a and c), we suggest that the brine composition is close to this estimate. Thus, we propose that the brine percolating at >4500 m depth in the Reykjanes geothermal system might contain up to 26 wt% Fe, 13 wt% K, 5 wt% Na, 37 wt% Cl, 0.6 wt% S and 1.2 wt% Cu. The low estimated H_2O content (~17 wt%) suggests that this brine might be very similar to salt melt inclusions reported from the porphyry Cu and Au ore deposit of Biely Vrch Slovakia (Kodera et al., 2014). Zierenberg et al. (2017) report that a yellow salt formed on cut core surfaces due to evaporation of pore fluid in the rock. The yellow salt was examined by EDS and determined to be dominantly composed of K, Fe and Cl with less abundant Mn, Zn and Cu, accompanied by halite crystals.

4 DISCUSSION

4.1 The Physical State of the Geothermal Fluid >4500m depth

The major aim of the IDDP project is to drill into the supercritical region of an active geothermal system. Borehole logs suggest that the pressure at the bottom of the IDDP-2 hole might reach ~35 MPa and the estimated borehole temperature at the same depth is ~535 (±50) °C based on the Horner method (Friðleifsson et al., in press). These estimates suggest supercritical conditions if the geothermal fluid is pure H_2O or dilute saline fluid similar to sea water (Driesner, 2007), which dominates the shallow part of the Reykjanes geothermal system (Arnórsson, 1978). Based on our study, the fluid temperature at the bottom of the borehole should be close to 600 ± 20°C. These temperature estimates are similar to the lower estimates of Zierneberg et al. (2020), determined based on alteration minerals in the same core material.

If a single supercritical fluid was present in the Reykjanes geothermal system in these conditions, we should observe only a single fluid inclusion type. These inclusions, if saline, should homogenize during heating experiments to the liquid phase, with shrinkage of the vapor bubble first and a fading meniscus at the homogenization temperature (Klyukin et al., 2019). Instead, we observe the presence of vapor-rich (Type1) and brine inclusions (Type2), occasionally associated with silicate melt (Type3) inclusions within a single FIA (Figure 2). Furthermore, Type1 inclusions homogenize into the vapor phase at 600 ± 10°C, whereas brine inclusions are not homogenized below 715°C. The highly variable proportion of vapor bubble within Type2 inclusions suggest that the geothermal fluid is not a single supercritical fluid but is separated to a low density vapor-rich fluid dominated by H_2O and a salt-rich brine at these conditions. The phase transformations in the coexisting vapor (Type1) and brine (Type2) inclusions can be approximated by the properties of the well-studied $NaCl-H_2O$ system (based on Bodnar and Vityk, 1994, Driesner, 2007, Bakker, 2018). Halite dissolution in the brine at 377-390°C and vapor homogenization at 595-597°C happens if these fluid inclusions were simultaneously trapped in their host minerals at pressures between 50 and 60 MPa (Driesner, 2007, Bakker, 2018). We must, however, note that the presence of $FeCl_2$ in a saline fluid changes the position of the critical curve in the P-T-x space (Steele-MacInnis et al., 2015). As the critical curve in $FeCl_2-H_2O$ system has a smaller dP/dT slope compared to that in $NaCl-H_2O$ or $KCl-H_2O$ systems, the entrapment pressure of the studied fluid inclusions might be slightly lower than the estimated value based on the pure $NaCl-H_2O$ system. Therefore, we suggest that the fluid entrapment pressure is close to hydrostatic conditions (45 MPa) and might be somewhat higher than those measured at the bottom of the borehole.

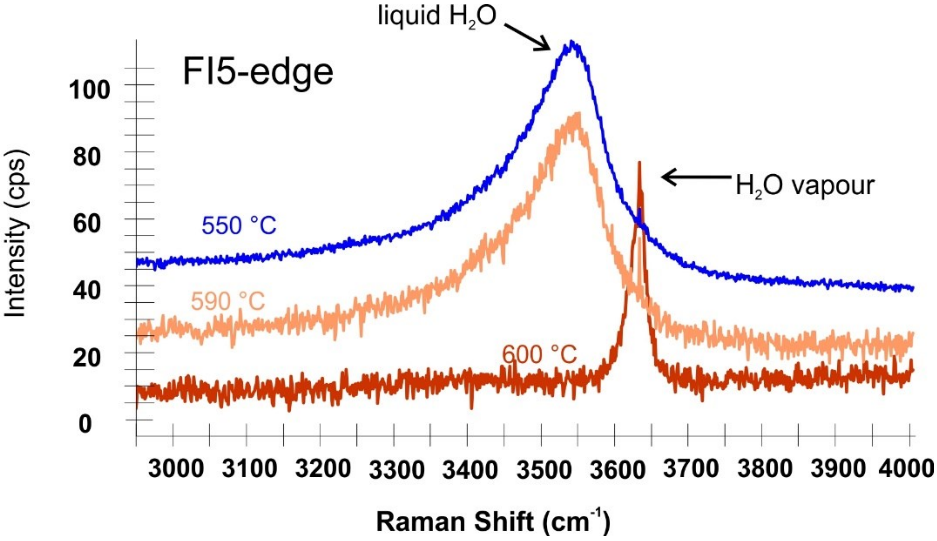

Figure 3: Raman spectra of liquid and vapor H_2O analyzed at the edge of a Type1 fluid inclusion. Note that although no liquid film was optically visible above 550°C, it was detected up to 590°C in all fluid inclusions.

4.2 Interaction of Geothermal Fluid and Metabasaltic Crust >4500 m Depth

The potassic composition of the rhyolite (Type3) melt inclusions and the occasional presence of S1 in their bubble phase (Figure 2d) clearly shows that the silicate melt interacted with the K-rich brine in the system. Although rhyolites and even alkali rhyolites with a total alkali content up to 11.5 wt% are not uncommon in Iceland (Jónasson, 2007 and references therein), they are always sodic with $K_2O/Na_2O<1$. Therefore, the elevated K_2O/Na_2O ratio in these melt inclusions must be the result of mixing between a sodic rhyolite formed by either crustal anatexis or fractional crystallization of tholeiitic basalt (Zierenberg et al. 2020, Jónasson, 2007, and references therein) and the Fe-K-rich brine percolating at >4500 m depth.

In the shallow part of the Reykjanes geothermal system the geothermal fluid has salinities similar to sea water, with an Na/K ratio of 6.5 (e.g., Arnórsson 1995), although fluid inclusions studies suggest varying salinity between 0.5 and 7.6 wt% $NaCl_{equiv}$ (Franzson et al., 2002, Fowler et al., 2015). At >4500 m depth and ~600 °C the fluid composition is markedly different. The brine is dominated by KCl over NaCl, with Na/K of 0.5, and the salinity in the brine of over 40% $NaCl_{equiv}$. This change in fluid composition can be attributed to the metamorphism of the original tholeiitic basaltic and gabbroic crust of the Reykjanes peninsula in the presence of seawater. At the shallower/cooler part of the system, the original plagioclase is commonly albitised, depleting the fluid in Na relatively to K. Similarly, the replacement of original igneous clinopyroxenes (Na_2O~0.2 wt%, e.g., Marks et al., 2010) with hornblende (Na_2O~1.6 wt%, K_2O~0.2 wt%, Marks et al., 2010) would lead to a progressive decrease in the Na/K ratio in the geothermal fluid, leaving behind a K-rich brine. These reactions also lead to elevated Fe content in the fluid.

Figure 4: Phase transformations in Type2 fluid inclusions during heating experiments. Photomicrographs were taken in plane polarized light at a) room temperature, b) 179 °C (S1 already dissolved), c) 195 °C, d) 350 °C (S2 already dissolved), e) 400 °C (S3 already dissolved), f) 600 °C (S4 already dissolved). Note the change of the shape of vapor bubble between room temperature and 179 °C.

Similar high temperature hypersaline fluid associated with silicate melt inclusions have been described from porphyry Cu and Au ore deposits from Biely Vrch (Slovakia, Kodera et al., 2014), Cerro de Pasco, (Peru, Rottier et al., 2016), Grasberg (Papua province Indonesia, Mernagh and Mavrogenes, 2019). The brine inclusions studied here might contain Cu at wt% level, and these high Cu concentrations are potentially coupled with elevated Au, Ag and Pb contents, as observed in the shallower part of the geothermal system (Harðardóttir et al., 2012, 2013). Pyrrhotite and intermediate solid solution Cu-Fe sulfides are part of the alteration assemblage in the deep cores (Zierenberg et al., 2020). This suggests that the IDDP-2 borehole not only sampled geothermal fluid that has the potential to produce an order of magnitude higher energy than the conventional geothermal wells (Albertsson et al., 2003), but it may have sampled an actively forming high grade ore deposit. These ore deposits should be widespread in the non-accessible Mid-Oceanic Ridges worldwide and are analogous to volcanogenic massive sulfide (VMS) ore deposits in ophiolites.

4 CONCLUSIONS

The geothermal fluid at the bottom part (>4500 m) of the IDDP-2 bore hole reaches 600°C and is separated into a H_2O-rich vapor and a Fe-K-Cl-rich brine with Cu concentration potentially over 1 wt%. The high temperature H_2O-dominated steam might serve the geothermal industry with high enthalpy fluid for electricity production, whereas the brine transports and deposits Cu-rich ores below 4500 m depth. The potassic composition of silicate melt inclusions and their occasional presence with the fluid inclusions suggest that silicic melt has interacted with the brine at this shallow depth. Therefore, there is a danger of drilling into magma at similar depths at the Reykjanes peninsula.

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