

Evolution of fluid chemistry during basalt-water and basalt-seawater interactions revealed by hydrothermal flow-through experiments

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Recently, several deep drilling projects for exploiting the supercritical geothermal reservoirs in the world. For example, in 2017, the Island Deep Drilling Project (IDDP-2) reached 427 °C and 34 MPa, which exceeds the critical point of seawater. The drillcores revealed that the intense water-rock interaction under greenschist to amphibolite facies metamorphism. Although such deep drilling provides us direct information of active metamorphic processes. however, it is still unclear whether conventional petrological analyses could apply to reaction processes in open systems at high temperature hydrothermal alteration at seafloor. There have been few experiment studies on the water-rock interaction under supercritical conditions with some exceptions (Yardley et al. 2014).

In this study, we conducted hydrothermal flow-through experiments on the basalt-H₂O, basalt-NaCl.aq, basalt-seawater systems under the sub or supercritical conditions. The temperature and pressure were set to be 400 °C and 37 - 39 MPa, and the input solutions was flowed at constant flow rate (0.1 ml/min) for 1 -6 days. We placed a powder of MORB (Island, 250-500 μm), which is composed of plagioclase (Pl), olivine (Ol) phenocrysts and clinopyroxene (Cpx), Pl groundmass, with lessor amount of volcanic glass, in a Ti inner tube (L=275 mm, I.D.= 4.35 mm). During the run, the solution was continuously pumped into the reaction vessel, and the output solution was collected every few hours. The concentration of Si, Al, Ti, Fe, Mg, Ca, Na, and K in the output solution was analyzed by ICP-AES.

In the basalt-H₂O experiment, the products varied with distance from the inlet. At the inlet and middle of tube, Ca-rich Pl (An# 0.98) formed around primary Pl grain with producing abundant pores, whereas chlorite was dominantly formed around Ol and Pl at the middle to outlet of inner tube. The solution chemistry evolves with time: Si concentration was initially high and then decreases toward the steady state. pH, Na and Al increases with decreasing Si, whereas the Ca, Mg and Fe concentrations were low. These features indicate the dissolution of volcanic glass at the initial stage, and then the solution chemistry was controlled by preferential leaching of albite component of plagioclase.

In the basalt-saline water (2.5 wt% of NaCl) experiment, we observed pH rises accompanying the decrease Si concentration, indicating the leaching of Si and dissolution of Pl similar to the basalt- H₂O experiment. One of the large differences between the two experiments was the extremely high Na concentration of saline water, resulting in the leaching of Ca (Ca concentration of the output solution was 40 to 80 times that of H₂O experiment), or breakdown of Ca-bearing minerals. In the basalt reaction with artificial seawater, which contains a lot of Mg²⁺, SO₄²⁻, CO₃²⁻ ions, revealed that the significant amount of precipitation of sulfate minerals occurred in the pre-heating parts of the vessel (before pass through the fluid reached the basalt sand), and that chlorite was formed throughout the reaction tube, which clogged the reaction vessel in a day. In contrast to the H₂O and saline water experiments, pH decreased to 3. In spite that we conducted the experiments with the same rock sample (basalt sand) and the same P-T conditions, the products and solubilities after the water-rock interaction were quite different in response to the input solutions, probably because the open system, in which new fluids are continuously supplied,

is never rock-buffered. We will analyse the complicated reaction systems which control the evolution of solutions and minerals, based on the analyses of speciation of aqueous species, saturation of minerals and variation of replacement textures of minerals.

Reference

Yardley, B. W., Harlov, D. E., & Heinrich, W. (2010). Rates of retrograde metamorphism and their implications for crustal rheology. *Geofluids*, 10(1-2), 234-240.

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