European Geothermal Congress 2016
Strasbourg, France, 19-24 Sept 2016

Alteration products of reservoir rocks from the Upper Rhine Graben under geothermal conditions

Roman B. Schmidt[1], Kurt Bucher[2], Ingrid Stober[1]

[1] Karlsruhe Institute of Technology, Institute of Applied Geosciences, Adenauerring 20b, 76131 Karlsruhe, Germany

[2] University of Freiburg, Institute of Geosciences, Albertstraße 23b, 79098 Freiburg, Germany

roman.schmidt@kit.edu

Keywords: Fluid-rock interaction, geothermal energy, alteration, analcime, Upper Rhine Graben.

ABSTRACT

Deep seated fault systems play an important role in the utilization of deep geothermal energy, especially in non-volcanic enhanced geothermal systems with low natural permeability. In such settings, the fluid migration and heat convection take place through the connected fracture network that is often saturated with saline geothermal fluids, interacting with the primary and secondary mineral content of the host rock. The aim of this research is to comprehend the mineralogical alterations that occur, when the primary mineral assemblage is subjected to a synthetic geothermal fluid at temperatures above 200 °C.

Batch-type experiments with 2 molar Na-Cl solutions and cylindrical hard rock samples have been performed. Samples were taken from four different lithologies representing potential reservoirs for future geothermal exploration: Malsburg granite (crystalline basement), Tennenbach sandstone (Lower Buntsandstein) and Pfinztal sandstone (Upper Buntsandstein). Both, the solid samples as well as the fluid samples will be studied in order to draw a more general picture of the reaction process. The rock sample will be analyzed for dissolution processes, ion exchange reactions of primary minerals and precipitation of secondary minerals.

After extraction of the granite samples from the experiment, the surface mineral grains are disintegrated from the matrix. The main dissolution features that can be observed in SEM images are deep holes in quartz grains. The feldspars, however, do not show substantial dissolution, but are often covered with newly formed clay precipitates. Near-surface biotite grains are totally altered to greenish chlorite. The two sandstone samples show a different reaction behavior. Quartz dissolution is less pronounced than in the granite samples. While the feldspar remains mostly unaffected, muscovite grains show substantial alteration. The most prominent precipitate that can be found in the SEM images is analcime that occurs in spherical shape. It is usually associated with few-µm small hexagonal kaolinite.

1. INTRODUCTION

Deep seated fault systems play an important role in deep geothermal application, particularly in volcanically inactive regions with low natural permeability. Faults are crucial for convective heat transport and permeability within a fault zone is one decisive parameter for efficient geothermal utilization.

In our research site-specific URG aquifer rocks will be chemically and mineralogically characterized. Fluid-rock interaction experiments will be carried out to investigate alteration processes. This research serves the purpose to develop a better understanding and predict the efficiency of geothermal utilization in the URG.

Typical reservoirs of the Upper Rhine Graben comprise the crystalline basement and the stratigraphically younger Bunter, mostly composed of sandstones (Genter and Traineau 1992, Pauwels et al. 1993). In-situ fluids of the geothermal reservoir units are highly saline, with TDS up to 100-200 g/l and compositionally dominated by Na-(Ca)-Cl brines (Sanjuan et al. 2016, Stober and Bucher 1999, 2015).

2. METHODS

Batch-type experiments with several reservoir rocks were conducted in a stirred autoclave system (Limbo li, Buechi) to mimic geothermal conditions. A 2 molar Na-Cl fluid and cylindrical hard rock samples with a diameter of about 3 cm have been used. Experiments have been performed at 200 and 260 °C for every lithology, respectively, and under corresponding water vapor pressure. This is in the range of recorded reservoir temperatures for the URG, which is up to 260 °C (Sanjuan et al. 2016).

One granite (MGr) and two different sandstone lithologies have been selected as representative lithologies these reservoir horizons. The major mineral assemblage of the granite is quartz, K-feldspar, albite and biotite, the latter being partly chloritized. Main

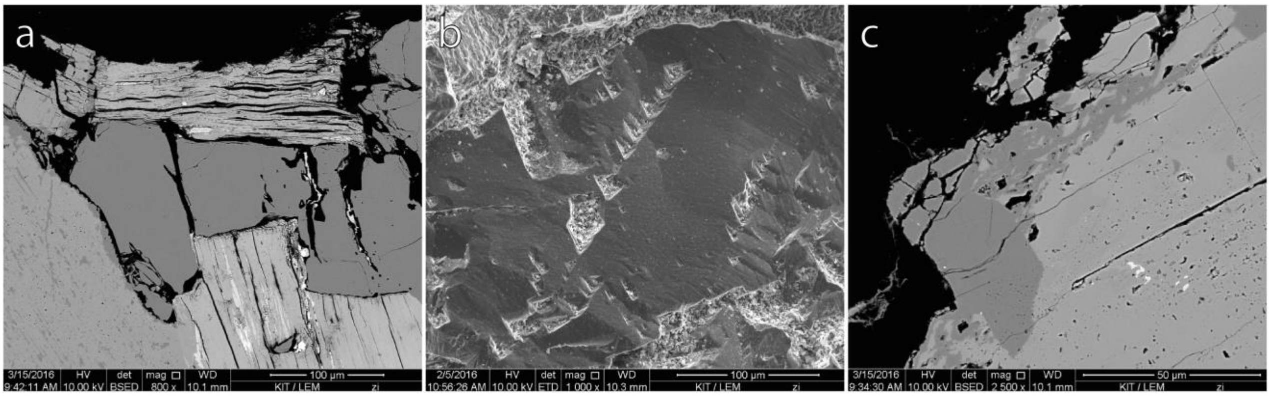

Figure 1: SEM images (SE and BSE) of the granite sample after 260 °C experiment: (a) disintegrated rock fabric due to chloritization of biotite and quartz dissolution (dark grey); (b) triangular etch pits on the quartz surface; (c) albitization of K-feldspar at the mineral-fluid interface.

phases of the fine-grained sandstone (Pfinztal sandstone, PfSst) is quartz, K-feldspar and relatively high amounts of muscovite. The second, Tennenbach sandstone (TenSst), is coarse grained with quartz and K-feldspar building up the framework and kaolinite, quartz and illite in the matrix.

2.1 Analytical Methods

Solid samples have been analyzed prior and after the alteration experiments with a special emphasis on dissolution of primary minerals and ion exchange reactions, as well as on precipitation of secondary minerals. For this purpose, SEM/EDX, electron microprobe and XRD have been applied. Fluid samples have been analyzed after the experiments by using ICP-MS, ICP-OES and IC.

3. RESULTS

3.1 Granite

Granite samples show intense alteration reactions after the experiments. The rock fabric is disintegrated through chloritization of biotites and extensive quartz dissolution (Fig. 1a). At 200 °C the (former) biotite grains near the surface protrude beyond the initially flat sample surface, evidencing a volume increase. In contrast, biotite after exposure to 260 °C show dissolution features and form morphological depressions. Quartz dissolution features are seen on surface images, as well as in thin section cross-cuttings. Wormhole-like and triangle shaped etch pits are reaching deep into sample interior (Fig. 1b). K-feldspars are subject to albitization on many parts of the sample surface (Fig. 1c). Secondary clay particles in the range of 1 µm can be found on entire sample surface. Additionally, an anhedral layer of analcime covers parts of the sample from the 260 °C experiment.

3.2 Sandstones

Sandstone samples show reaction features that are clearly different to those observed on granite samples. Quartz dissolution is less pronounced, while dissolution of matrix phases from the sandstones is intense. Incorporation of Na appears to be dominant process in the alteration of muscovite in PfSst samples. Albitization of K-feldspar occurs at near surface grains of the sandstone samples, similar as observed on granite samples. Analcime are the most abundant secondary mineral and can mainly be found on the entire surface independent of the substrate. It forms euhedral, spherical shaped analcime crystals cover all reacted sandstone samples at bother temperatures (Fig. 2a + b). The crystals often incorporate tiny Fe-oxides, which are primary in the matrix of both sandstones, PfSst but more pronounced in TenSst. Analcimes are often neighbouring with few-µm small secondary clay minerals (Fig. 2c).

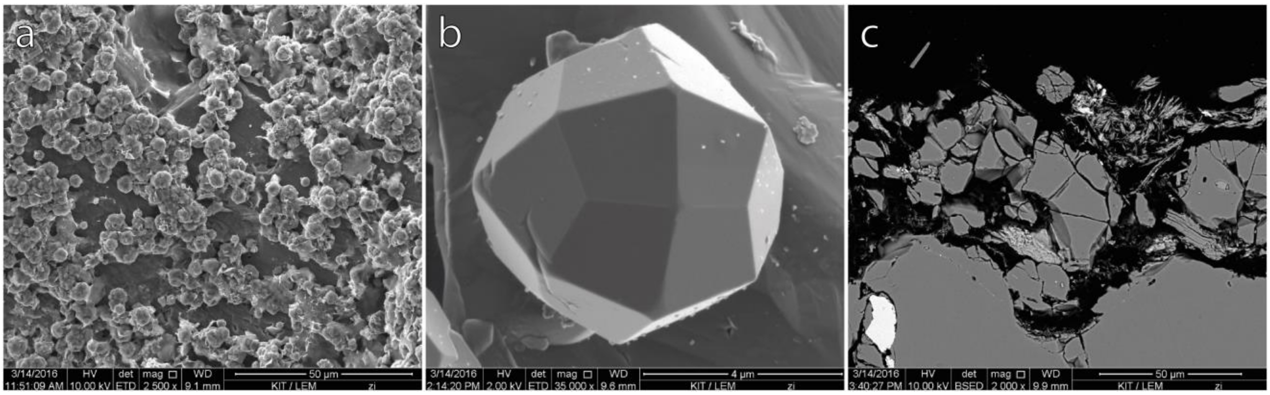

Figure 2: SEM images (SE and BSE) of analcime precipitation on sandstone sample after 260 °C experiments: (a) sample surface is covered by euhedral analcime (TenSst), (b) spherical analcime on PfSst, (c) BSE thin section image (PfSst) with partly dissolved sandstone matrix, quartz grains and secondary analcime.

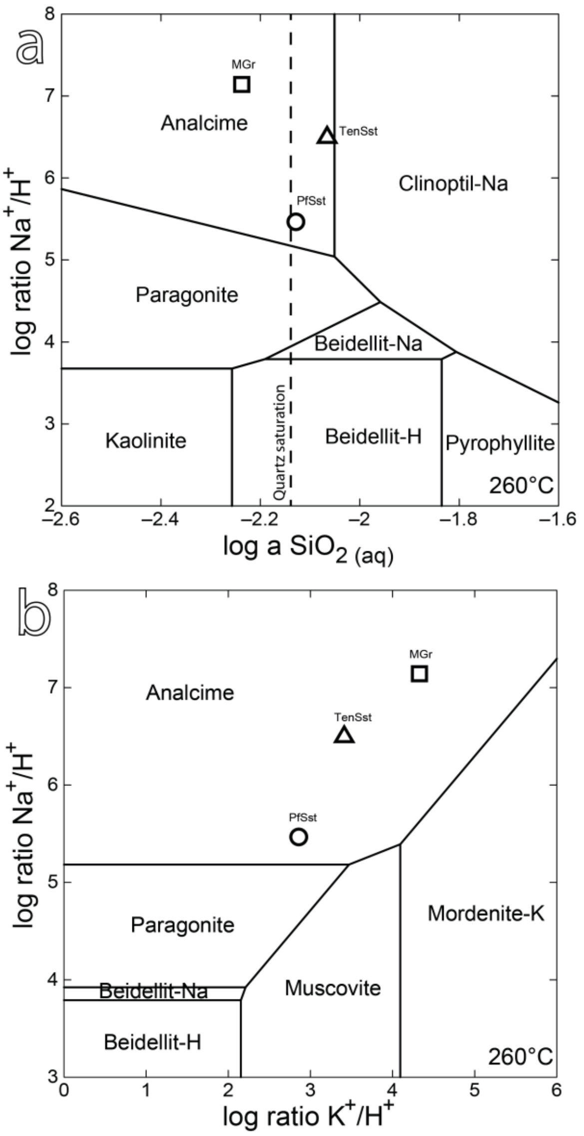

Figure 3: Activity diagrams calculated with GWB (Bethke 1996, GWB dataset thermos.tdat) with data points of the experimental fluid. Conditions were temperatures of 260 °C and corresponding water vapor pressure. Al was balanced with kaolinite. (a) log a(Na^+)/a(H^+) vs. log a($SiO_{2(aq)}$); (b) Log a(K^+)/a(H^+) vs. Log a(Na^+)/a(H^+) ($SiO_{2(aq)}$ = Quartz saturated).

3.3 Fluid

The rock alteration is also preserved in the fluid composition after the experiment. Elevated concentrations of potassium reflect albitization of K-feldspars in sandstone as well as in granite samples, and furthermore the progressing chloritization of biotites in the experiments with granite. Quartz dissolution is recognized in silica concentrations in the fluid in the range of quartz saturation (Fig. 3a). Main cation concentrations plot in the stability field of analcime (Fig. 3).

3. CONCLUSIONS AND OUTLOOK

The experiments show that alteration reactions are strongly lithology-dependent with distinct alteration paths. Several mineral-mineral and mineral-fluid reactions could be identified for the different lithologies. Future studies will establish a more process-based understanding of the alteration reactions by also quantifying the reactivity of single mineral phases and their contribution to the bulk process.

REFERENCES

Bethke, C.: Geochemical Reaction Modelling. *Oxford University Press*, Oxford. (1996).

Genter, A. and Traineau, H.: Borehole EPS-1, Alsace, France: preliminary geological results from granite core analyses for Hot Dry Rock research, *Scientific Drilling*, (1992), 3, 205-214.

Pauwels, H., Fouillac, C. and Fouillac A. M.: Chemistry and Isotopes of Deep Geothermal Saline Fluids in the Upper Rhine Graben - Origin of Compounds and Water-Rock Interactions. *Geochimica et Cosmochimica Acta*, **57**, (1993), 12, 2737–2749.

Sanjuan, B., Millot, R., Innocent, C., Dezayes, C., Scheiber, J. and Brach, M.: Major geochemical characteristics of geothermal brines from the Upper Rhine Graben granitic basement with constraints on temperature and circulation, *Chemical Geology*, **428**, (2016), 27–47.

Stober, I. and Bucher, K.: Deep Groundwater in the crystalline basement of the Black Forest region, *Applied Geochemistry*, **14**, (1999), 237-254.

Stober, I. and Bucher, K.: Hydraulic and hydrochemical properties of deep sedimentary aquifers of the Upper Rhine Graben, Europe, *Geofluids*, (2015).

Acknowledgements

This project is supported by the German Federal Ministry for Economic Affairs and Energy (BMWi, no: 0325623C).