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Reactivity of geothermal reservoir rocks under temperature conditions found in the Upper Rhine Graben (Germany)

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Abstract

Deep seated fault systems play an important role for deep geothermal applications, particularly in volcanic inactive regions with low natural matrix permeability, such as the Upper Rhine Graben (URG, SW Germany). Fluid migration and heat transport occur through a network of connected fissures and faults, which generally are saturated with highly saline fluids. Alteration processes of primary mineral phases are a crucial for the evolution of the fissure permeability and thus, the lifetime of the geothermal reservoir. The aim of this experimental research is to investigate the interaction between reservoir rocks and synthetic close-to-nature brines to comprehend the alteration path from primary minerals to an alteration mineral assemblage. Primary reactions are intense quartz dissolution, chloritization of biotites in the granite samples, albitization of K-feldspar (granite and sandstones) and an extensive precipitation of highly euhedral analcime crystals on the surface of sandstone samples.

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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.

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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^{2,3}. 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 brines⁴⁻⁶.

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 °C⁴. All experiments lasted about 50 days.

One granite (MGr) and two different sandstone lithologies have been selected as representative lithologies for these reservoir horizons. The major mineral assemblage of the granite is quartz, K-feldspar, albite and biotite, the latter being partly chloritized. Main 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. Thus, the selected rock samples for the experiments represent the major modal composition of the reservoir rocks in the URG.

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.

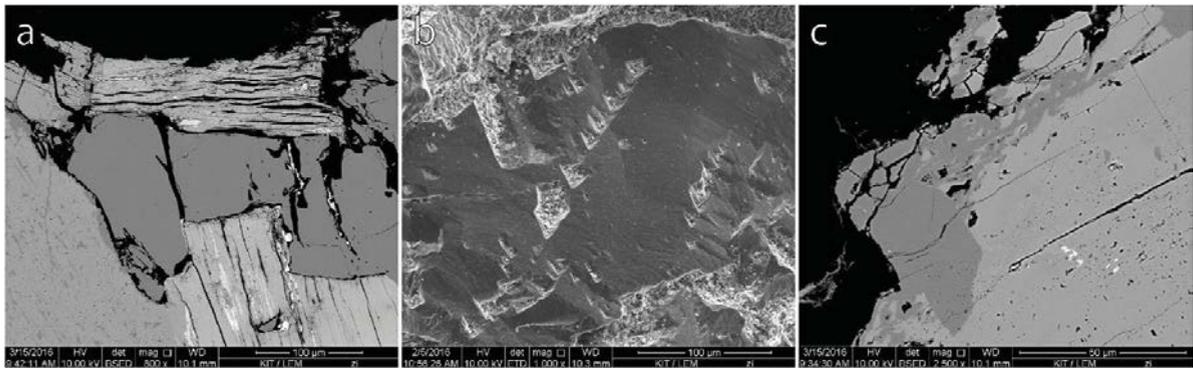


Fig. 1. SEM images (SE and BSE) of the granite sample after 260 °C experiment: (a) disintegrated rock fabric due to chloritization of biotite and quartz dissolution (dark grey); (b) triangular etch pits on the quartz surface; (c) albitization of K-feldspar at the mineral-fluid interface.

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 both 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 neighboring with few- μm small secondary clay minerals (Fig. 2c).

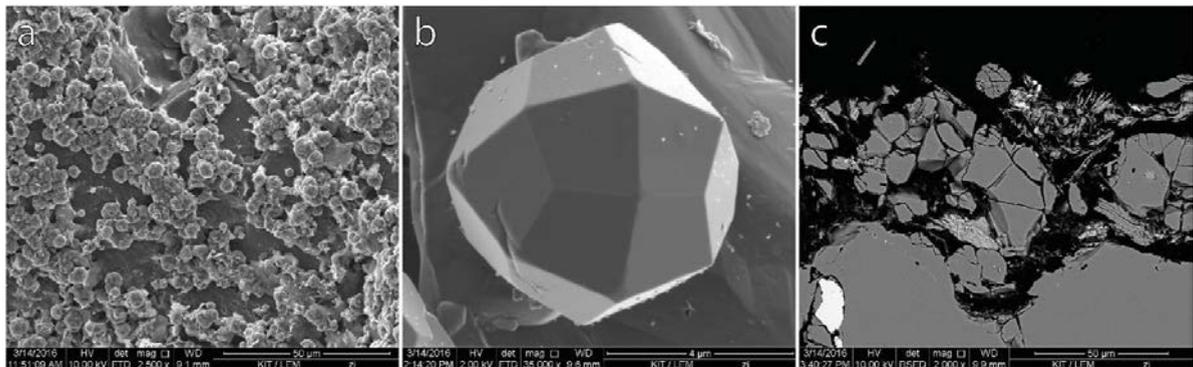


Fig. 2. SEM images (SE and BSE) of analcime precipitation on sandstone sample after 260 °C experiments: (a) sample surface is covered by euhedral analcime precipitates (TenSst), (b) spherical analcime crystal in PfSst sample, (c) BSE thin section image (PfSst) with partly dissolved sandstone matrix,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

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).

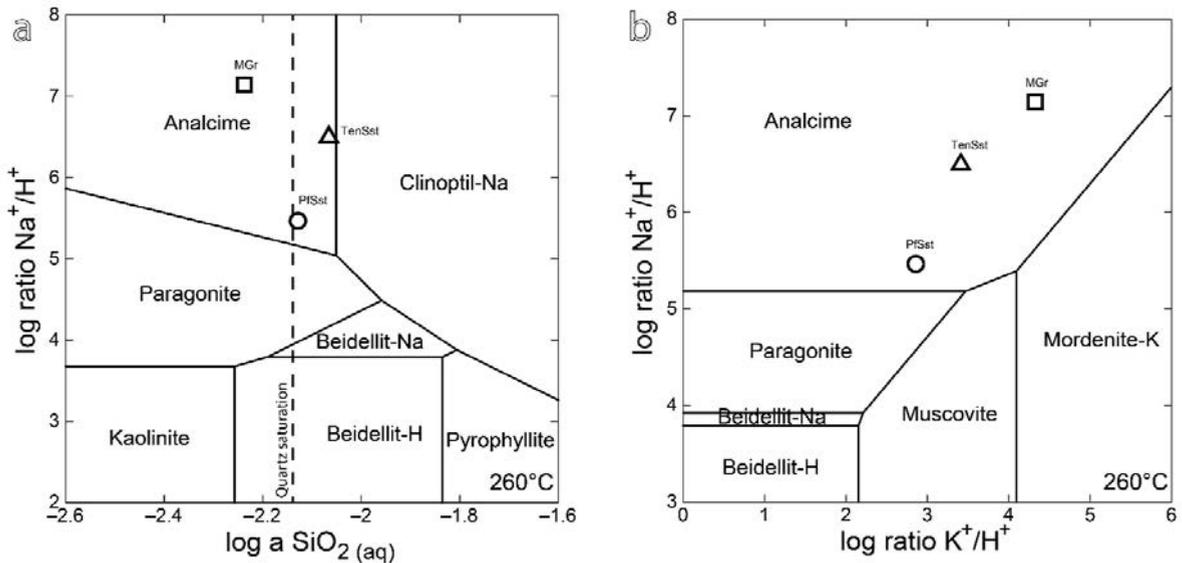


Fig. 3. Activity diagrams calculated with *GWB*¹ (*GWB* dataset *thermos.tdat*) with data points of the experimental fluid. Conditions were temperatures of 260 °C and corresponding water vapor pressure. Al was balanced with kaolinite. (a) $\log a(\text{Na}^+)/a(\text{H}^+)$ vs. $\log a(\text{SiO}_{2(\text{aq})})$; (b) $\log a(\text{K}^+)/a(\text{H}^+)$ vs. $\log a(\text{Na}^+)/a(\text{H}^+)$; ($\text{SiO}_{2(\text{aq})}$ = Quartz saturated).

4. Conclusion and Outlook

The experiments show that alteration reactions are strongly lithology-dependent with distinct alteration paths. Nevertheless, steady state conditions have not been reached. Thus, for example crystallization of analcime consumed earlier formed secondary clay minerals. But they are still partly present in direct proximity to the analcime crystals.

Several mineral-mineral and mineral-fluid reactions could be identified for the different lithologies. Both, volume increasing (e. g. analcime precipitation) and decreasing (e.g. quartz dissolution) alteration processes occur at the same time. So, at this state of the study it is not possible to determine definite permeability changes in the fractured geothermal systems. 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.

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