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Redox reactions during sandstone flow-through experiments at geothermal conditions

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Abstract

Copper ions (Cu^+ or Cu^{2+}) in formation fluids can be reduced by carbon steel of the wells casing at geothermal conditions resulting in native copper ($\text{Cu}(0)$) precipitation, and oxidation and dissolution of iron (Fe). Indeed, high amounts of $\text{Cu}(0)$ were observed at the bottom of a production well at the geothermal site Groß Schönebeck (Germany). It was expected that this reaction may also reduce the permeability of the reservoir rock close to the well casing. This study aimed to simulate this reaction at laboratorial conditions with special focus on the effect of reaction temperature and kinetics.

A flow-through experiment was designed to simulate the conditions in a geothermal reservoir and well: a saline, and Cu-containing solution flowing through a sandstone media in contact with a steel capillary representing the well casing. Cu and Fe concentration as well as pH-, and redox values were monitored over time. After the experiment, pore spaces were characterized by X-ray computer tomography and by microprobe analysis using EDX on thin sections.

Precipitation of Cu (without Fe) occurred within the sandstone near the wall and bottom of the capillary. The reaction-induced precipitation reduced the flow rate, but did not reach equilibrium after 49 h. While the brine salinity had little impact on the reaction kinetics, an increasing temperature from 25 to 80°C strongly enhanced the overall reaction.

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1. Introduction

Water-rock interactions at geothermal conditions are often difficult to predict because access to representative samples at great depth is difficult and even the simulation of those processes at laboratorial conditions is challenging due to elevated temperatures and pressures. However, the geochemical processes assumed to happen at the

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geothermal site in Groß Schönebeck (North German Basin) are of high relevance and therefore a thorough understanding of the processes is important to predict and possibly prevent those reactions.

Briefly, at the geothermal research laboratory Groß Schönebeck a well doublet was drilled into a Permian sandstone and volcanic rock reservoir (1). The fluid at around 4100 m depth is a Ca-Na-Cl brine at a temperature of about 150 °C that contains 265 g/L total dissolved solids^{1,2}. During hydraulic tests in 2011 and 2012, a strong decrease in rock permeability was observed over time³. Simultaneously, it was found, that the production well was clogged with scaling material that consisted predominantly of native copper, together with barite, laurionite, and magnetite⁴. Based on these observations it was assumed that native copper forms as corrosion product by the reaction of Cu-containing fluid with the carbon steel liner at depth⁴. It was further hypothesized that this reaction also occurs within the reservoir rock on the outer side of the steel tube, where the Cu precipitates would clog the pores⁴. This could be an explanation for the observed reduction in productivity rates³. To verify and quantify this, a laboratorial experiment was developed to simulate the reaction in dependence of temperature and salinity.

2. Materials and Methods

The set-up of the flow-through laboratorial experiments consists of a steel capillary (representing the well casing) that was first inserted into a pre-drilled rock sample (representing the geothermal reservoir; Fig. 1). The assembly was placed into a 5 l container (“reservoir container”) yielding synthetic brines of various compositions. The capillary was connected to a tube through which the brine was pumped (peristaltic pump) into a second container (“collecting container”). The initial pumping rate was adjusted to 6 rpm (60 ml/h) but needed to be re-adjusted in all experiments over time due to variable flow-through conditions that established during some experiments. Both containers were continuously stirred (magnetic stirrers) and heated to the desired temperature. The brine and the entire system were permanently Ar purged to keep the experiment oxygen-free. Altogether, eight experiments were performed that varied with respect to temperature (25°, 50°, or 80 °C), brine composition (with and without chloride salts), and type of steel (RS carbon steel or stainless steel 1.4307; Table1). For each experiment a “fresh” rock sample (cylindrical sandstone, 50 mm in length, 25 mm in diameter) and steel capillary (30 mm length; 6 mm outer diameter; 3 mm inner diameter) were used. The rock material was a Fontainebleau quartz arenite consisting of 99.98 % of quartz^{5,6} with a permeability of ca. $100 \cdot 10^{-15} \text{ m}^2$. The synthetic brine always contained 1 mM CuCl_2 with a background of either deionized water or deionized water with 2 M NaCl and 1.5 M CaCl_2 . The samples were immediately acidified to $\text{pH} < 2$ and total Fe and Cu were measured by inductive coupled plasma optical emission spectroscopy (ICP-OES). Before and after each experiment, both the rock samples and the capillary were dried at 105 °C and weighed. After the experiments, the capillaries were separated from the rock and rock samples were transected perpendicularly to the capillary.

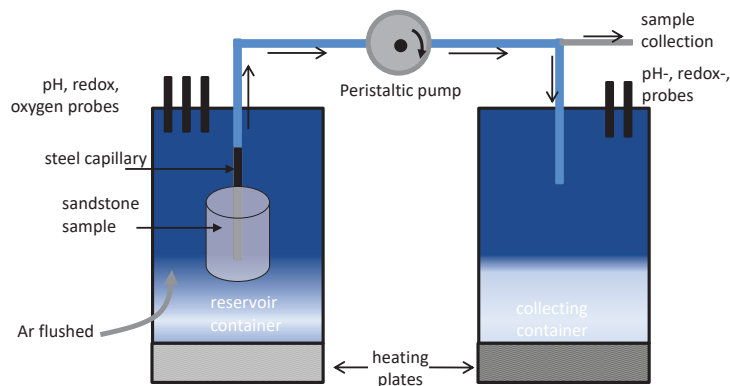


Fig.1. Experimental set-up: the fluid is pumped from the reservoir container (filled with synthetic, Ar-flushed brine) through the rock-steel capillary assembly. Redox-, pH-, and dissolved oxygen are continuously monitored.

Polished thin sections were prepared after embedding the rock samples in epoxy resin. They were analyzed by

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