



# Fluid–rock interactions in CO<sub>2</sub>-saturated, granite-hosted geothermal systems: Implications for natural and engineered systems from geochemical experiments and models

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Received 18 November 2013; accepted in revised form 16 June 2014; Available online 27 June 2014

## Abstract

Hydrothermal experiments were conducted and geochemical models constructed to evaluate the geochemical and mineralogical response of fractured granite and granite + epidote in contact with thermal water, with and without supercritical CO<sub>2</sub>, at 250 °C and 25–45 MPa. Illite ± smectite ± zeolite(?) precipitate as secondary minerals at the expense of K-feldspar, oligoclase, and epidote. Illite precipitates in experiments reacting granite and granite + epidote with water; metastable smectite forms in the experiments injected with supercritical CO<sub>2</sub>. Waters are supersaturated with respect to quartz and saturated with respect to chalcedony in CO<sub>2</sub>-charged experiments, but neither mineral formed. Carbonate formation is predicted for experiments injected with supercritical CO<sub>2</sub>, but carbonate only formed during cooling and degassing of the granite + epidote + CO<sub>2</sub> experiment.

Experimental results provide insight into the buffering capacity of granites as well as the drivers of clay formation. Metastable smectite in the experiments is attributed to high water–rock ratios, high silica activities, and high CO<sub>2</sub> and magnesium–iron concentrations. Smectite precipitation in supercritical CO<sub>2</sub>-bearing geothermal systems may affect reservoir permeability. Silicate formation may create or thicken caps within or on the edges of geothermal reservoirs. Carbonate formation, as desired for carbon sequestration projects coinciding with geothermal systems, may require extended periods of time; cooling and degassing of CO<sub>2</sub>-saturated waters leads to carbonate precipitation, potentially plugging near-surface production pathways. © 2014 Elsevier Ltd. All rights reserved.

## 1. INTRODUCTION

Shallow crustal rocks routinely undergo fluid–rock interactions driven by thermal, chemical, and/or physical disequilibrium. The resulting mass transfer has important implications for our understanding of processes such as hydrothermal alteration, ore formation, fault-zone permeability, and sedimentary diagenesis. Mass transfer is also especially important to geothermal operations since chemical reactions can impact production.

Our study comes at a time of renewed interest in conventional (convective) and non-conventional (conductive)

*Abbreviations:* scCO<sub>2</sub>, supercritical CO<sub>2</sub>; EGS, enhanced geothermal systems; CCS, carbon capture and storage; GWB, the Geochemist's Workbench; CPG, CO<sub>2</sub>-plume geothermal systems.

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<http://dx.doi.org/10.1016/j.gca.2014.06.015>

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geothermal systems for energy extraction (e.g., Moore and Simmons, 2013) as well as for sequestering carbon dioxide to mitigate concentrations of atmospheric carbon (e.g., Benson and Cook, 2005). Water is the naturally occurring fluid in geothermal systems and is commonly used as the working fluid for heat extraction. Supercritical CO<sub>2</sub> (scCO<sub>2</sub>) has also been proposed as a working fluid (Brown, 2000; Pruess, 2006). Non-conventional enhanced geothermal systems (EGS) (MIT, 2006) and conventional CO<sub>2</sub>-plume geothermal systems (CPG) (Randolph and Saar, 2011) are both targets for using scCO<sub>2</sub> as a working fluid, with the possible ancillary benefit of sequestering carbon dioxide.

One goal of a CO<sub>2</sub>-based geothermal system (whether EGS or CPG) is to dry out the reservoir over time by gradually extracting and displacing water. Another goal is to optimize heat extraction efficiency. The advantage of using scCO<sub>2</sub> compared to water is its low viscosity, large expansivity, and reduced reactivity with rock. In addition to increased productivity, scCO<sub>2</sub>-based geothermal systems may sequester carbon, especially by mineral and/or dissolution trapping (Benson and Cook, 2005) or by loss to surrounding country rock if permeability is high enough.

CO<sub>2</sub>-rich fluids, however, are chemically aggressive and could impact reservoir permeability and porosity by dissolution and precipitation reactions. Because there are few natural laboratories (CO<sub>2</sub> reservoirs) where the evolution of fluid and hydrothermal alteration can be assessed, we have used hydrothermal experiments and geochemical models to: (1) quantify fluid–rock interactions in granite-hosted hydrothermal systems, with and without scCO<sub>2</sub>; and (2) bridge the gap that often exists among field observation, experimental results, and geochemical predictions. Our results elucidate processes such as the buffering capacity of felsic rocks, drivers of metastable clay formation, and tungsten ore formation. Results also clarify fluid–rock interactions in scCO<sub>2</sub>-based EGS or CPG as well as in high temperature granitic or arkosic reservoirs targeted for carbon capture and storage (CCS).

Many experimental efforts have been made over the past 50 years to elucidate hydrothermal fluid–rock interactions for felsic igneous rocks, and all offer unique contributions to our understanding of natural and engineered systems (Appendix A). The associated batch and flow-through experiments were conducted at temperatures and pressures ranging from 20 to 600 °C and 1.4–150 MPa with distilled water, or less commonly, dilute Na–Cl, Na–HCO<sub>3</sub>–Cl, or Na–Ca–HCO<sub>3</sub>–Cl water. A few used scCO<sub>2</sub> at the onset of each experiment. To date, we are unaware of any studies using both scCO<sub>2</sub> and a realistic groundwater chemistry. We are also unaware of any that allow a system to approach a steady-state prior to introducing scCO<sub>2</sub>. Our experiments were designed to explore such conditions.

A commonality observed in the referenced studies is the frequent precipitation of smectite as a secondary mineral. Based on our knowledge of natural systems, smectite and mixed-layer illite–smectite are characteristic of temperatures up to ~180 and ~220 °C, respectively (e.g., Henley and Ellis, 1983), and therefore, experimental observation of smectite formed at temperatures higher than ~180–

220 °C begs explanation. To our knowledge, this discrepancy is not discussed in the literature, and because of its critical implications for geothermal systems, we review and discuss the discrepancy in the context of experimental systems, geochemical models, and natural systems.

We present experimental data to evaluate aqueous geochemistry and mineralogical relationships in water–granite ± scCO<sub>2</sub> and water–epidote–granite ± scCO<sub>2</sub> systems at 250 °C and 25–45 MPa. Granite and epidote–granite experiments provide a baseline understanding for fluid–rock interactions in fresh and altered rock, respectively. We also construct thermodynamic models and compare calculations/predictions to our experimental results, and then compare our experimental and model data to natural and engineered systems. Although beyond the scope of this paper, a kinetic model is also under development. Important discussion topics include: (1) paths to equilibrium including the extent to which experiments approach equilibrium and the sequence of water–rock reactions; (2) results in the context of natural systems, including factors that govern smectite formation; and (3) implications for engineered systems.

## 2. METHODS AND MATERIALS

### 2.1. Experimental design and setup

Hydrothermal experiments were conducted in rocking autoclaves and flexible Au–Ti reaction cells (Dickson cells) (Seyfried et al., 1987). Each gold cell ports directly to a valve that allows for periodic fluid sampling without perturbing the experiment. Aqueous samples were collected approximately every 1, 2, 5, 14, and 28 days for each stage of an experiment (pre- or post- CO<sub>2</sub> injection). We also analyzed unreacted water and minerals (i.e., prepared for but not included in the experiment) as well as the very final reacted water and minerals (i.e., recovered at the conclusion of the experiment).

Our experiments and geochemical simulations emulate the geothermal conditions, aqueous geochemistry, and mineralogy of Roosevelt Hot Springs, Utah (Ward et al., 1978; Capuano and Cole, 1982), a well-characterized, magmatically-heated geothermal system. Basing simulations and experiments on such a system allows us to relate results to similar settings. With respect to CO<sub>2</sub>-based EGS or CPG, our experiments are representative of conditions when the reservoir water is saturated with CO<sub>2</sub> and has not yet been displaced by scCO<sub>2</sub>. Experiments have initial and final water:rock ratios of ~20:1 and 10:1, respectively; this emulates the high water:rock ratio in a localized zone of fractured granite.

The granite is a mixture of powdered (75%, <45 μm) and chipped (0.1–0.7 cm) research-grade quartz, plagioclase feldspar (oligoclase), potassium feldspar (K-feldspar), and minor biotite. Instead of using actual granite, this approach allowed us to avoid alteration and accessory minerals, thus simplifying analysis of our modeling and experimental results. Two experiments also include 50% epidote, and simulate fluid–rock interactions in EGS reservoirs stimulated by fracturing along pre-existing zones of weakness

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