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Porosity and permeability evolution induced by precipitation of silica under hydrothermal conditions

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Abstract

Water-rock interaction provides significant influences on evolution of hydrological properties within the crusts. Especially, dissolution and precipitation of silica in fractures play essential roles on permeability evolution. In this study, we conducted hydrothermal-flow through experiments on silica precipitation within porous media (alumina inner tube packed with alumina balls) for investigating how a fracture is clogged by silica precipitation. We used high Si supersaturated solution for precipitation under vapor (390 ˚C and 20 MPa) or supercritical condition (420 ˚C and 30 MPa), and the porosity structure was examined by X-ray CT. Under the vapor condition, fine-grained quartz was nucleated and immediately deposited on the bottom. In contrast, under the supercritical condition, spherical silica particles, which are composed of opal-A, opal-C and quartz, were formed in fluids, transported and covered the surface of alumina balls uniformly. In the latter case, a characteristic fluid pressure (permeability) oscillation was observed; which is probably explained by repeated clogging and break of pore throats.

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Keywords: silica, precipitation, hydrothermal experiments, fluid pressure oscillation

1. Introduction

Permeability within the crust is not static parameter, but it changes dynamically[1]. The permeability change has been often considered to link with the seismic cycle[2]; when earthquake rupturing occurs permeability increases immediately, and it decays gradually by sealing and/or healing processes. However, such a dynamic cycle has not been verified experimentally. Silica is one of the dominant constituents of the Earth's crusts, and its solubility

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changes as a function of pressure and temperature[3]; therefore, dissolution and precipitation of silica minerals provide significant impact on the hydrological and mechanical properties of the Earth's crust. In the geothermal area, where geothermal gradient exceeds 100 ˚C/km, the deep drilling projects revealed that the crust is divided into permeable zone in the shallower part and impermeable zone in the deeper; the boundary of permeable and impermeable zones is located at 3-5 km depth[4,5]. Based on the quartz solubility in water along the deep wells, the permeable and impermeable boundary is consistent with the drop of silica solubility; therefore, silica precipitation controls the hydrological properties of the Earth' crusts[6,7]. Several authors[7] revealed that this drastic drop of silica solubility corresponds to the phase transition of water from liquid to supercritical conditions. There have been limited experimental studies on permeability evolution during water-rock reactions under hydrothermal conditions in rock fractures. Most of these previous experiments were dissolution experiments under relatively low temperatures (<200 ˚C)[8]. Therefore, it is still unclear how porosity structure and permeability evolve during the dissolution and precipitation of minerals at deep crustal environments. In this study, we conducted hydrothermal flow-through experiments for silica precipitation under sub to supercritical conditions, especially focusing on how silica precipitation clogs the fracture from high Si supersaturated solution.

2. Experiments

The fundamental apparatus similar to that in[9], is briefly explained here. The apparatus consists of two reaction vessels; first one is the dissolution vessel for preparing the high Si aqueous solutions, and second one is the precipitation vessel (Fig. 1a). In the dissolution vessel, granite + quartz sand (1-2 mm size) was dissolved in distilled water at around the critical point of water. The fluid with high Si solution is brought into the supercritical or vapor conditions. The precipitation vessel has a tube-in-tube structure where alumina inner tube (4 mm inner diameter, and 100 mm length) is placed in the main vessel, which was made of stainless steel. In the alumina inner tube, alumina balls with 1 mm size were closely packed as an analogue of torturous flow path within the fracture. After each run, the alumina inner tube was taken out for evaluating of distribution of silica precipitates and porosity along the flow path. The resolution of X-ray was 10 μm. During the experiments, the fluid pressures at inlet and outlet of the precipitation vessels were monitored, and the solutions were taken periodically to identify the concentration of silica as well as minor components such as Al, Na and K.

We conducted two-series of experiments. In the first experiment, silica precipitation was caused in response to phase transition from liquid to vapor at 20 MPa, and in the second experiments from liquid to supercritical fluid (SCF) at 30 MPa (Fig. 1a). The durations of the vapor and SCF experiments were 67 and 30 h, respectively.

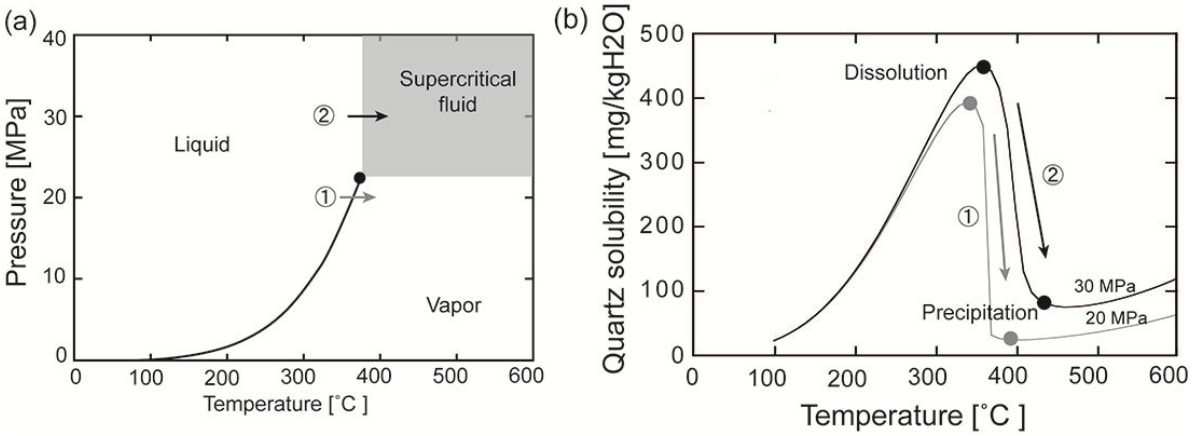

Fig. 1. (a) P-T conditions, and (b) quartz solubility in water for silica precipitation experiments in response to phase transition of H_2O (1) from liquid to vapor and (2) from liquid to supercritical fluids.