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Experimental study on calcite precipitation in hydrothermal environments

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The crustal fluids are commonly characterized by the compositions in C-H-O system, mainly composed of H₂O, CO₂ and CH₄, and thus transport of these fluids and precipitation of carbonate are important for the global carbon cycle. To reduce greenhouse gas (CO₂, CH₄ etc) in the atmosphere, the carbon storage underground has been tried; however, it is considered to be difficult to precipitate calcite in reasonable timescale. In contrast, calcite veins are very common in the oceanic crusts, metamorphic rocks, and accretionary prisms. For example, calcite + quartz veins occur ubiquitously in the Shimanto belt. The solubility of calcite decreased with temperature, that is the opposite trend of quartz; and thus how calcite precipitated in the conditions that quartz also occurs is puzzling. In spite of its importance, the experimental studies on the calcite precipitation are very limited. Most experiments are carried out under near room temperature and controlled by pH change or synthetic CO₂ saturated fluids (Lee & Morse, 1999), that are far from natural conditions of calcite-vein formation. To best of our knowledge, there are no experimental studies on calcite precipitation under hydrothermal conditions (>100 degree C).

The purpose of this study is to understand the controlling factors on calcite precipitation under conditions of calcite-vein formation (fluid compositions, P-T conditions, host rock types). The solubility of calcite increases with decreasing temperature, with increasing fluid pressures, and with increasing concentration of NaCl (Ellis, 1963). What is the most controlling factor that enhances the calcite-vein formation at the conditions of the Shimanto belt is unknown. As a first step, we conducted hydrothermal flow-through experiments to precipitate calcite at 300 degree C and 30 MPa by using the temperature dependency of solubility.

Before the precipitation experiments, because a reliable solubility data on calcite at elevated temperature is lacking, we tested how amount of Ca ions dissolved from calcite in the flow-through system at 30 MPa with temperature range from 100 to 400 degree C. The Ca concentration path-through the vessel is highest at 100 degree C, and it decreased with increasing temperature.

The experimental apparatus of the precipitation experiments is composed of two reaction vessels; in the first vessel, the super-saturated solutions were prepared by dissolution of limestone sand (1-2 mm in size) in the distilled water at 100 degree C. In the second vessel, seven limestone substrates (5x5x15 mm) were set along the flow-path. The limestone is composed of fine grained aggregate of calcite (<0.03 mm). The temperature of the precipitation vessel was set to be 300 degree C. The fluid flow rate was 1 ml/min. After the run of 240 h (10 days), the total increase of weight of limestone substrates was 0.051 g. Observation of the surface morphologies of the substrates by SEM and thin sections by optical microscope reveal that euhedral calcite crystals with size of 0.02-0.03 mm grew from the calcite in the substrates.

Our results suggest that calcite veins could be formed around 300 degree C, if fluids saturated with calcite at lower temperature would be brought accompanying with subduction of slabs. However, our experimental conditions deviate from that of quartz and calcite vein formation, because the temperature increase leads to the dissolution of quartz, so that it cannot be co-existing of quartz and calcite in the same veins. Second, the source of Ca and CO₃²⁻ would be the host sedimentary or basaltic rocks in the Shimanto belt. We will try to precipitate calcite with using other factors, including pressure dependence on the solubility, the host rock type.

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