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Hydrothermal experiments on calcite precipitation via water-rock interaction

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Calcite veins are very common within crusts 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. Also, the fluid inclusions in the Shimanto veins are composed of CH4, with exception of CO2-dominant inclusions found in the southern parts of the Muroto Peninsula, that belongs to the Tertiary Shimanto belt (Lewis, 2000). 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?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 or pH, 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. We conducted two types of hydrothermal flow-through experiments for calcite precipitation by (1) temperature change and (2) water-rock interaction at constant P-T. In both experiments, the P-T condition for calcite precipitation is 300 ?C and 30 MPa.

In the first experiments, the supersaturated solutions were prepared by dissolution of limestone sand (1-2 mm in size) in the distilled water at 100 ?C. In the precipitation 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 ?C. The fluid flow rate was 2.5 ml/min. After the run of 240 h (10 days), the total increase of weight of limestone substrates was 0.051 g. Observations by SEM and by optical microscope reveal that epitaxial grwoth calcite from substrate crystals with size of 0.02-0.03 mm.

In the second experiment, we used NaHCO3 solution (pH 8.4) as input solution. In the preparing vessel, sands of sandstone, mudstone or basalt from the Shimanto belt were set to dissolve Ca and other cations. In the second vessel, four limestone substrates as the same size as the first experiment. The measured fluid flow rate was 2.59 ml/min. After the run of 240 h, the total increase of weight of substrates was 0.037 g. Observation of the surface of the substrates by SEM and EDS reveal that calcite crystals with size of 0.01 mm precipitated together with clay minerals and apatite.

Our results suggest that calcite veins could be formed at high temperature around 300 ?C, in higher pH fluids, if fluids saturated with calcite by Ca from host rocks and CO32- in the crustal fluids. The possibility of the formation of quartz and calcite vein is also suggested from the precipitation of silicate and calcite from natural rock samples at the same temperature. The source of Ca and CO32- would be the host sedimentary or basaltic rocks in the Shimanto belt.

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