Formation of silica particles in vent fluids under supercritical and vapor conditions

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The precipitation of minerals (sulfides and silica) in seafloor hydrothermal vents is conventionally thought to be caused by rapid cooling due to mixing of hot hydrothermal fluids and cold seawater, but the formation mechanism of mineral particles is still unclear. The Kuroko deposits, one of the typical massive sulfide deposits in association with rhyolitic volcanism, are distributed along the western side of the central mountains of NE Japan. Some Kuroko deposit shows a concentric strcuture similar to that of chimneys in submarine hydrothermal deposits (Shimazaki and Horikoshi, 1990), where characteristic bipyramidal quartz grains occur along with sulfides. To understanding the physical and chemical conditions in the chimneys, in this study, we conducted a circulating hydrothermal experiment of silica precipitation and discussed the formation process of silica particles under supercritical and vapor conditions.

The silica precipitation experiments were conducted using an flow-through apparatus simulating a submarine hydrothermal vent, in which a high-silica aqueous solution (Si = 300 mg/kg(H_2O), Al = 5 mg/kg(H_2O)) prepared by quartz + albite, was flowed vertically upward. A 25 cm long alumina inner tube was installed in the reaction vessel, and 24 stainless steel nets were placed at 1 cm intervals. Silica minerals were precipitated by heating the reaction tube along the flow path and adhered to the nets in the alumina tube. After the experiment, the nets were removed, and the products were observed using a scanning electron microscope and were identified by the micro Raman spectroscopy. We conducted three runs at supercritical condition of 25 MPa and 350–430 ℃ (6 h, 12 h) and vapor condition of 8.6 MPa and 200–300 ℃.

For the experiments under supercritical conditions (25 MPa, 350–430 ℃), systematic changes of silica minerals were observed along the flow path from 350 ℃ to 430 ℃. In the 12-hour experiment, quartz was observed at 10–14 cm from the inlet and cristobalite at 15–18 cm. In the 6-hour experiment, cristobalite was dominant, but quartz was observed at 14–15 cm from the alumina tube inlet, and quartz was observed emerging from the interior of the cristobalite at 14–15 cm from the inlet. A thin section was prepared and mapped by Raman spectroscopy, which showed the presence of quartz inside the cristobalite. These results suggest that silica first nucleated as amorphous silica with low interfacial energy, and the silica minerals aggregated downstream and transformed into more stable cristobalite and quartz (Okamoto et al., 2015). Quartz has a high interfacial energy and is difficult to nucleate homogeneously, which may have caused heterogeneous nucleation inside the cristobalite. For the experiments under saturated vapor pressure conditions, the fluid pressure was set at 8.6 MPa and the reaction tube was heated from 200 ℃ to 300 ℃ along the flow path. Very fine-grained amorphous silica was observed at 20–24 cm from the alumina tube inlet. This confirms previous findings that amorphous silica is instantly produced as nano- to micron-sized spherical particles by nucleation and aggregation during evaporation of water droplets (Amagai et al., 2019). These results suggest that the formation of bipyramidal quartz requires the continuous supply in liquid or supercritical fluids.

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