

## Effects of Al and Na on mineralogy of silica deposits from hydrothermal fluids

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Silica is one of the most dominant components in the Earth's crust, and is characterized by high solubility with respect to water that is sensitive to temperature. An ubiquitous occurrences of the quartz vein in seismogenic depth of the subduction zones implies the importance of the quartz precipitation in fractures on earthquake cycle. However, the mechanism of quartz vein formation is still poorly understood. One of the difficulties is arisen by the fact that silica can precipitate from hydrothermal fluids as metastable polymorphs such as opals. Hydrothermal fluid contains various minor elements derived from the crustal rocks: and thus it is possible that kinetics of silica precipitation is affected by these minor elements. Our previous study (Okamoto et al., 2010) revealed that the mineralogy of silica from pure Si solutions are different from that from impure solutions including minor amounts of Al, Na and K derived from granite .

In this study, we conducted the hydrothermal flow-through experiments at 430 C and 31 MPa to investigate the effect of Al and Na on precipitation of silica minerals. . We use a blank vessel for precipitation of silica minerals without rock/mineral substrates. The Si-supersaturated solutions (300-350 ppm,  $C_{Si}/C_{Si,Qtz,eq} = 3-3.5$ ) were prepared by dissolution of quartz at 350 C, and the concentration of Al and Na in the input solution was systematically changed by dissolution of albite at different temperatures. The concentrations of Al and Na in the input solutions range from 0 to 7 ppm, and the atomic ratio of Al and Na were unity that is same as the stoichiometry of albite.

With increasing Al and Na concentration in the input solutions, the dominant silica mineral systematically changes from amorphous silica, cristobalite to quartz. The atomic ratio of difference of Al and Na between input and output solutions,  $dC_{Al}/dC_{Na}$ , was 1.06. The modal abundances of individual silica minerals were estimated from XRD spectra by using the internal reference addition method. With increasing Al concentration in the input solution, the modal abundance of amorphous silica monotonically decreases from 100 to 15 wt.%, whereas that of quartz increases from 0 to 82 wt.%. Cristobalite becomes dominant at the intermediate Al concentration (1-3 ppm) in the input solutions. In the experiments with low Al concentrations ( $C_{Al} < 3.0$  ppm) in the input solution, the contents of Al<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O of the products (amorphous silica and/or cristobalite) are less than 0.06 wt%, and no systematic relationship between Na and Al content. On the other hand, in the experiments with high Al solution ( $C_{Al} > 3.0$  ppm), the products contains Al<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O are contained up to 0.36 wt.% and 0.25 wt.%, respectively. These lines of evidences suggest that Al<sup>3+</sup> coupled with Na<sup>+</sup> substituted for Si<sup>4+</sup> in quartz in our experiments.

Natural silica sinters, which form at shallow levels (< 1 km depth) of the crust, are composed mainly of opaline silica (amorphous silica and cristobalite) with lesser amounts of quartz. In contrast, the hydrothermal quartz veins do not contain the relic of other silica polymorphs, expect for chalcedony. Such contrasting mineralogy of silica deposits probably reflect the concentration of Al and Na in the solution. Because solubility and morphology of precipitates are quite different between quartz and amorphous silica, the minor amount of Al and Na in the hydrothermal fluids would affects the location and amount of silica deposits at the Earth's crust.

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