



Formation of silica particles from supercritical fluids and its impacts on the hydrological properties in the crust

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Silica is a dominant constituent of the Earth's crust and the solubility of quartz varies significantly as a function of water density and temperature (e.g., Manning, 1994). In particular, in the volcanic areas with high geothermal gradient, the solubility of quartz increases with depth, and has a maximum at around 350 °C, and then drops drastically near the critical point of water and then increases again. Based on the quartz solubility profile along the deep drilling well at Kakkonda geothermal field in Japan, Saishu et al. (2014) suggested that the silica precipitation provides significant influence on the formation and maintenance of the permeable-impermeable boundary within the crust. Around the hydrological boundaries, the fluid pressure could fluctuate between lithostatic and hydrostatic, and the drop of fluid pressure causes silica precipitation. Weatherley and Henley (2013) proposed that at the time of earthquakes, fluid pressure drops to below the level of lithostatic pressure at the fault jogs, and that such flash vaporization produces gold-quartz veins. However, the mechanism of silica precipitation at such extreme condition are poorly understood.

We conducted the flash experiments of the high-silica aqueous solution ($\text{Si} = 9.1 \text{ mmol/kgH}_2\text{O}$), which was created by dissolution of granite sand at 370 °C, from 261, 353, 400, 450 °C at 36 MPa. All runs show the isothermal decompression P-T paths from liquid to vapor (261, 353 °C) or supercritical fluid to vapor (400, 450 °C) within 5 seconds. We collected the silica precipitates by an alumina filter placed at the outlet of the reactor, and found the formation of a large amount of spherical particles of amorphous silica. The size distribution of silica particles show the modal value of 500-900 nm, and some particles showed a cauliflower-like roughness, indicating that these particles are formed via nucleation and aggregation of amorphous silica during vaporization of water droplets. The batch experiments with using amorphous silica produced in the repeated flash experiments revealed that the amorphous silica changed to quartz within a few days at the supercritical conditions. These results of our experiments suggest that the short-lived amorphous silica could be formed easily in the high-temperature crusts in response to a fluctuation of fluid pressure. One of the characteristic features of silica nanoparticles is their high mobility in fluid flow, which is different from silica precipitation as quartz overgrowth from the vein walls. As the results, silica particles can clog faults effectively, and the clogging and break of pore throats by silica particles could cause the oscillation of permeability during the fluid flow in the crusts.

[1] Manning, C.E., 1994. *Geochim Cosmochim Acta*, 58, 4831-4839.

[2] Saishu, H., Okamoto, A., Tsuchiya, N., 2014. *Terra Nova*, 26, 253-259.

[3] Weatherley, D.K., Henley, R.W., 2013. *Nature Geosciences*, 6, 294.