

SCG060-10

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地殻におけるシリカ鉱物析出様式・速度に対するAlの影響 Effects of Al on kinetics of precipitation of silica minerals from aqueous fluids under crustal conditions

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Silica is a dominant component in the Earth's crust. Because of high solubility of silica in aqueous fluids and its dependency on P-T conditions, dissolution and precipitation processes of silica minerals play significant roles on the spatial and temporal distributions of fluids and rock strength in the crusts. An ubiquitous occurrence of quartz veins in the vicinity of seismogenic zones implies the importance of the sealing of fractures by quartz on the earthquake cycle.

In spite of the importance of the kinetics of silica precipitation, the complete expression of the rate equation has not been determined, except for precipitation rates on surface reactions (Rimstidt & Barnes, 1980). The difficulty in the estimates of the precipitation rate is arisen from the following reasons. First, although quartz is the most stable silica minerals in the crust, cristobalite and amorphous silica occur in the geothermal areas (Alekseyev et al., 2009). Second, precipitation of silica minerals occurs not only on quartz surfaces but also via nucleation processes in fluids. Third, trace elements including Al³⁺, Na⁺ and K⁺ in solutions affect on the species and kinetics during silica precipitation (Okamoto et al., 2010). Feldspars are dominant constituents of the crust, and thus the effects of these minor components are crucial for considering the silica precipitation in the crust.

In this study, we conducted the hydrothermal flow-through experiments to investigate the overall precipitation rate of silica minerals and the effects of Al in the solutions under crustal conditions (430 C and 31 MPa). The experimental apparatus is similar to that in Okamoto et al. (2010). For precipitation of silica minerals, we used a blank vessel that does not include any rock/mineral substrates. The Si-supersaturated solutions (300-350 ppm, C/Ceq = 3-3.5) were prepared by dissolution of quartz at 350 °C, and the concentration of Al in the input solution was controlled by dissolution of albite or granite with different temperatures. The Al and Na (and K in the case of granite) included in the input solutions from 0 to 7 ppm, and the atomic ratio of Al and Na were unity, that is same as the stoichiometry of albite.

The experiments in pure Si solution revealed that the precipitation via nucleation in fluids was approximated as the first-order reaction that is the same as the precipitation on the pre-existing quartz surfaces. Activation energy of precipitation of silica minerals from the solution was estimated to be 39 kJ/mol. In solutions in absence of Al, amorphous silica precipitated. With increasing Al and Na contents in the solutions, the dominant silica mineral systematically changes from amorphous silica, cristobalite to quartz. An important observation is that the logarithmic precipitation rate increased linearly with increasing the Al concentration. Combining these results and the surface reaction rate obtained in the previous study, we obtain the empirical full expression of the rate equation on the silica precipitation that is a function of the degree of supersaturation, temperature, pre-existing quartz surface area, water volume, and Al concentration. One of the implications of this rate equation is that dominant precipitation mechanism changes from surface growth on quartz to precipitation via nucleation, with increasing fracture aperture or decreasing quartz mode in the wall rock in the crust. This is consistent with the observations of natural quartz veins. We will discuss the spatial distribution of silica precipitation and its relationship to the fluid flow in the crusts.

References: Rimstidt, J. D. and Barnes, H. D., 1980, *Geochim. Cosmochim. Acta.*, 44, 1683-1699.; Alekseyev, V. A., Medvedeva, L. S. and Starshinova, N. P., 2009, *Geochem. Int.*, 47, 731-735.; Okamoto A., Saishu H., Hirano N. and Tsuchiya N., 2010, *Geochim. Cosmochim. Acta.*, 74, 3692-3706.

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