

Coupling of serpentinization reactions and silica diffusion

Atsushi Okamoto^{1*}, Yuichi Ogasawara¹, Oyanagi Ryosuke¹, Noriyoshi Tsuchiya¹

¹Graduate School of Environmental Studies, Tohoku University

Silica activity is considered to be one of the key factors in controlling reaction paths and rates of serpentinization (e.g., Frost and Beard, 2007; Klein et al., 2009). There are many experimental studies on serpentinization during mantle peridotites and water interaction, focusing on extent of serpentinization, evolution of fluid chemistry and generation of hydrogen and hydrocarbons. However, all of these studies focused on bulk solid materials and solutions within the reaction vessel, and local changes of products and silica concentration gradient have not been clarified; therefore, the role of silica transport on progress of serpentinization is still unclear. In this study, we conducted hydrothermal experiments in the olivine (Ol)[?]orthopyroxene (Opx)[?]H₂O system at 250 degreeC and at a vapor-saturated pressure of 3.98 MPa to explore the role of silica diffusion in aqueous fluids during serpentinization. Olivine (Fo91), orthopyroxene (En92), or their composite powders (with Ol/Opx/Ol zones) were set in tube-in-tube vessels, and solution chemistry and the extent of serpentinization were analyzed in detail.

In the Ol[?]H₂O experiments, the product changed from serpentine + magnetite to serpentine + brucite + magnetite, accompanied by a Si-drop in the solutions, that is similar to the results of our previous observations (Okamoto et al., 2011). Serpentinization proceeded uniformly throughout the reaction tube, indicating that the supply of water was not the rate-determining process. In the Opx[?]H₂O experiments, orthopyroxenes were dissolved along the cleavages, and a small amount of serpentine + talc was formed. The silica activity of the solutions in the Opx[?]H₂O experiments was at the level coexisting serpentine + talc, that is 1[?]3 orders higher than in the Ol[?]H₂O experiments.

In the Ol[?]Opx[?]H₂O experiments, serpentinization proceeded in both the Ol and Opx zones, and white-colored reaction zone (2 mm thick) was developed at the boundary of the Ol and Opx zones. In the Opx zone, the extent of serpentinization was nearly constant, and mixture of serpentine + talc was formed. In the Ol zone, the total amount of H₂O revealed by thermogravimetry and SEM observations was most extensive along the boundary between the Ol and Opx zones, and it decreased gradually away from the boundary. Based on the Mg/Si values of the products, the ratio of talc in the products (M_{talc}/M_{talc}+M_{srp}) is highest in the Opx zone, and decreased steeply in the boundary zone, and talc was absent away from the boundary. The silica concentration in the bulk solution is lower than the stability field of talc, indicating the high Si gradient within the cm scale reaction tube.

Serpentinization in the Ol[?]Opx[?]H₂O experiments was modeled simply by coupled processes involving silica diffusion and three reactions: (1) a silica-consuming reaction after olivine, (2) a silica-releasing reaction after orthopyroxene and (3) talc-forming reaction after serpentine. We will discuss the kinetic model explain the reaction progress and evolution of solution chemistry, and the role of silica on the rate of serpentinization. Our experimental results represent an analogue of serpentinization in natural hydrothermal systems with a high porosity, and we suggest that the spatial variation of serpentine as a function of the distance from a source of silica could be a useful indicator of the relative magnitudes of reaction, mass transport, fluid flow as well as temperature during hydrothermal alteration of oceanic lithosphere.

Okamoto, A., Ogasawara, Y., Ogawa, Y., Tsuchiya N., 2011. *Chemical Geol.*, 289, 245-255.

Frost, B.R., and Beard, J.S., 2007. *J. Petrol.*, 48, 1351-1368.

Klein, F., Bach, W., Jons N., McCollom T., Moskowitz B., Berquo, T. 2009. *Geochim. Cosmochim. Act.*, 73, 6868-6893.

Keywords: serpentinization, silica diffusion, rate-determining process, olivine-orthopyroxene-H₂O