

Mass-transfer and rate-limiting process of serpentinization

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Serpentine minerals, which are produced by interaction between ultramafic rocks and fluids, contain about 13% water and are the greatest carrier of H₂O into the deep interior of Earth. Therefore, the volume and distribution of hydrated oceanic mantle are of special interest for understanding the global water circulation. There are several hydrothermal experiments on serpentinization in ultramafic rocks (Ol, Opx, peridotite)-water (seawater) interaction; however, these previous studies focused only on the extent of hydration of solids (Martin & Fyfe, 1970; Wegner & Ernst, 1983) or on the evolution of solution chemistry (e.g., Seyfried and Dibble, 1980; Allen and Seyfried, 2003). Therefore, the detailed reaction mechanism is still poorly understood, including evolution of the overall reactions, rate-limiting process, and resulting textures.

In this study, we conducted hydrothermal experiments in the olivine (Ol, Fo91) - orthopyroxene (Opx, En92) - H₂O system at 250 degreeC and vapor-saturated pressure (P_{sat}) for understanding the mechanism of serpentinization at oceanic lithosphere. At this temperature, high extent of hydration is expected for both Ol-H₂O and Opx-H₂O systems. The low-pressure condition of this study enables us to analyze both solution chemistry and the extent of hydration of the solid samples in detail. The main cylindrical reaction vessel (inner diameter 10.5 mm, height 100 mm) contains two sub-reaction tubes (inner diameter 4.5 mm, height 100 mm), in which the mineral powders (0.025-0.125 mm in size) are packed by meshes. We conducted three types of experiments in the Ol-H₂O, Opx-H₂O and Ol-Opx-H₂O (Opx layer is sandwiched by Ol layers) systems, respectively.

In the Ol-H₂O system, the reaction is divided into three stages. The Mg and Si concentrations increases (stage 1), then decreases (stage 2) and reaches the steady state (stage 3). The mineral assemblage also changes from serpentine (Srp) + magnetite (Mgt) at stages 1 and 2 to Srp + Mgt + brucite (Brc) at stage 3, that is consistent with the solutions, that change from stability field of serpentine to serpentine + brucite by drop of silica activity. The serpentine minerals occur as aggregate of fine-grained crystals (primarily lizardite, but chrysotile appear at stage 3), and discrete brucite crystals occur at the contact with olivine. The olivine commonly contains fractures filled by the products, that is similar to the natural mesh textures. In the Opx-H₂O system, the silica activity is 1 to 3 order higher than that in the Ol-H₂O system. The products are composed only of serpentine, and do not contain brucite, talc and magnetite. In contrast to serpentinization after olivine, the reaction occurs by pseudomorphic replacement of Opx. In the Ol-Opx-H₂O system, the Mg concentration in the bulk solution is similar to that of the Opx-H₂O system, whereas the Si concentration shows the similar behavior to the Ol-H₂O experiments. The serpentinization preferentially occurred in the Ol zone at the contact with the Opx zone.

At 250 degreeC, the hydration rate is greater in the Ol-H₂O system than in the Opx-H₂O system. The contrasting natures of solution chemistry and products suggest that the rate-limiting process during serpentinization in the Ol-H₂O, Opx-H₂O and Ol-Opx-H₂O systems are dissolution of olivine, precipitation of serpentine, and dissolution of orthopyroxene, respectively. Our results also indicate that hydrogen production, that is accompanied by the formation of magnetite, does not occur in the vicinity of Opx.

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