Progress of hydration in olivine-H$_2$O and orthopyroxene-H$_2$O systems at Psat

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Hydration of ultramafic rocks (serpentinization) commonly occurs in mid-ocean ridges, and the extent and distribution of hydrated mantle plays an important role on the global circulation of H$_2$O fluids. Although there have been several experimental studies on serpentinization (e.g., Martin and Fyfe, 1970; Seyfried et al., 2003), these studies focused only on the H$_2$O content in the products or solution chemistries. Therefore, fundamental kinetics and mechanism of serpentinization is still poorly understood. In this study, we conducted hydrothermal experiments on serpentinization to clarify the relationship between the temporal evolution of solution chemistry, progress of hydration reaction and textural developments.

The starting materials were powders of olivine (Fo$_{90}$, Fa$_{10}$) or orthopyroxene (En$_{65}$, Fs$_{35}$) with size of $<$0.125mm. The powders and the distilled water were set in the batch type vessel (8.8 cm$^3$) made of the stainless steel with water/rock ratio of 1.0. The temperatures were 250 degreeC at the liq-vap saturation curve. The duration was up to 1008 hours. After the experiments, the solid samples were dried at 90 degreeC for one day, and then were analyzed by X-ray diffractometry and Thermogravimetry. The morphology and cross sections of the products were observed by Scanning electron microscope. The solutions were analyzed by ICP-Mass Spectrometry. The pH values of the solution after the experiments were 7.6?8.5 at room temperature. In both systems, hydration reactions proceeded, but show quite different features in the products and solutions. In the Opx-H$_2$O experiments, chlorite formed on the surfaces of opx grains. The Si concentration increased with time toward 193.8 ppm, whereas the concentration of Mg was 3.42 ppm. The total H$_2$O content was less than $<$0.7 wt.% even at 1008 h. The formation of chlorite is different from the common occurrence of talc after orthopyroxene in natural peridotites. The reason is not clear, but one possibility is that higher Fe content in opx used in this study than that in the oceanic peridotites (En $>$ 80). In the Ol-H$_2$O experiments, the products were composed of serpentine, magnetite with or without brucite. The H$_2$O content of the samples increased with time, and reached 3.9 wt.% in 1008h. According to the solution chemistry, the progress of serpentinization is divided into three stages. The concentrations of Si and Mg in solutions increased toward 3.66 and 20.54 ppm, respectively (stage 1), and then decreased drastically toward 0.29 ppm and 0.28 ppm at 504 hours, respectively (stage 2). After 504 h, the solution chemistry was nearly stable (stage 3). Accompanying with the change in the solution chemistry, the products varied as follows: After 336 hours (stage 2 and 3), brucite started to form at the contact with olivine within serpentine rims. Also, only lizardite occurs at stage 1 and 2, whereas chrysotile formed with lizardite. According to activity diagram in Mg-Si-O-H system, the solutions at stage 1 and 2 corresponds to the stability filed of serpentine, and a drop in Si concentration indicates the shift from the serpentine stability field to serpentine+brucite stability filed, that is consistent with the change of the products. These results suggest that serpentinization after olivine is not always simple as Ol+H$_2$O=Serp+Br, but the reactions evolve with time. In common peridotites, Ol and Opx coexisted, and hydrothermal alteration occurs heterogeneously. Our experimental results indicate that olivine-H$_2$O and opx-H$_2$O system show a quite different fluid compositions (Si and Mg) and different rate of hydration. We will conduct further experiments with Ol+Opx+H$_2$O systems, and discuss how hydration proceed in the oceanic seafloors.


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