Formation of low-crystalline magnesium silicate in natural alkaline environment and flow-through experiments

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INTRODUCTION

Magnesium silicate formation at low temperature $(25-200^{\circ})$ is receiving a lot of attention in various research fields. Sepiolite and serpentine, crystalline Mg-clay minerals, are known to be formed in MgO-SiO₂-H₂O systems at low temperature. However, some studies have been suggesting that low-crystalline magnesium silicate recently should be formed in this low-temperature condition. These kinds of low-crystalline silicates have actively been studied by many reaserchers and engineers. For instance, calsium and alminium silicates are known as C-S-H (calcium silicate hydrate) and allophane/imogolite, respectively. These are studied for both natural minerals and synthesized materials. On the other hand, studies of the low-crystalline magnesium silicate are still in their infancy and little is known. Therefore, the nature of the low-crystalline magnesium silicate has not been clear yet. Since the magnesium silicate can be utilized in various engineering fields (e.g., improved oil recovery, CO₂ geological storage, geothermal power generation, concrete degradation, mineral clogging in radioactive waste disposal, treatment of environmental pollutions, etc.), the current reserch gaps in the understanding of the low-crystallline magnesium silicate formation need to be addresed so as to contribute positively to the above engineering technologies. Moreover, this study can also be applied to the field of planetary science because low-crystalline magnesium silicate should be observed even on surface of Mars and in interstellar silicate dusts.

ANALYSES OF NATURAL PRODUCTS

Firstly, this study confirmed that low-crystalline magnesium silicate forms in nature. Samples of precipitates were collected from the surface where fluid flows out from serpentinite rocks around Kamuikotan metamorphic belts, Hokkaido, Japan. Although a clear XRD pattern from the bulk samples was not observed, electron microscopy (Field emission scanning electron microscope and Transmission electron microscope; TEM) clearly identified the phase. TEM-energy dispersive X-ray spectrometry indicated that the low-crystalline phase was composed of Mg, Si, and O. Moreover, electron diffraction with TEM showed diffuse halo rings from some particles in the samples, showing the formation of low-crystalline phase. Furthermore, the fluid samples participating in formation of the precipitates were collected and analyzed. From the results, the fluid samples were alkaline (9.50 < pH < 10.67) and saturated with low-crystalline magnesium silicate phase (magnesium silicate hydrate reported by Lothernbach et al., 2018).

FLOW-THROUGH EXPERIMENTS

Secondly, to demonstrate the above formation, flow-through experiments in open system were conducted. Alkaline solution with 1.5 mM of dissolved silica was flowed into MgO powder in the reaction cell. TEM observation revealed the low-crystalline magnesium silicate formation as well. Moreover,

dissolution rate of Mg-bearing mineral and formation rate of magnesium silicate were obtained from dissolved silica and magnesium concentrations in the output solutions. These suggested that the magnesium silicate formation was extremely rapid under the oversaturated condition, and that the dissolution of Mg-bearing mineral is the rate-determining step during formation of the low-crystalline magnesium silicate. Although the flow-through experiments used only MgO as a Mg-bearing mineral, the above discussion can be applied to all of Mg-bearing minerals such as olivine and pyroxene because dissolution rate of MgO is greater than any other Mg-bearing minerals. Therefore, the flow-through experiments broaden the kinetical understanding of low-crystalline magnesium silicate formation not only at the Kamuikotan metamorphic belts, but also to other areas where the magnesium silicate is formed from the supply of Mg-bearing minerals in rocks/soil.

Keywords: Magnesium silicate, Transmission electron microscope, Kinetics, Kamuikotan metamorphic belts, Serpentine, Thermodynamic calculation