

Hydrothermal alteration experiments of MgO-SiO₂ amorphous silicates

TAKAHASHI, Ryohei¹ ; TSUCHIYAMA, Akira^{1*} ; MIYAKE, Akira¹ ; KAWAMURA, Katsuyuki²

¹Division of Earth and Planetary Sciences, Graduate School of Science, Kyoto University, ²Graduate School of Environmental and life Science, Okayama University

Hydrothermal alteration is one of the important processes for solid evolution in the early solar system. So far, many experiments have focused on reaction between aqueous fluids and meteorite itself or crystalline silicates, such as olivine and pyroxene (e.g., [1, 2]). Reaction with amorphous silicates is also important because interstellar dust as original building blocks of planetesimals is mostly amorphous [3] and primitive amorphous silicates are present in cometary dust as GEMS (e.g., [4]) and some carbonaceous chondrites (e.g., [5]). Hydrothermal alteration experiments with amorphous silicates were also carried out using natural GEMS [6] and synthetic materials with CI composition [7]. However, any experiments using amorphous silicates in a simple system have not been made. In this study, hydrothermal alteration experiments of amorphous silicates in the system MgO-SiO₂-H₂O were carried out to understand basic features of hydrous layer silicate formation.

Nanoparticles of amorphous silicates ~10 nm in size with nearly enstatite and forsterite compositions (E and F: Mg/Si = 1.15 and 2.02) were synthesized by the induction thermal plasma method as starting materials. Intermediate compositions were also prepared by mixing E and F (ME, M and MF: Mg/Si=1.25, 1.5 and 1.75). Three types of experiments were made. (1) Synthetic experiments: Starting materials and pure water (water/rock ratio: W/R=5.0) were sealed and heated at 50, 100, 150 and 200°C or held at room temperature for 0 to 1344 hrs. The run products were dried in vacuum and examined by powder X-ray diffraction (XRD) and SEM/EDX. (2) In-situ experiments: Starting materials mixed with pure water (W/R=5.0) were exposed in water-saturated atmosphere at 60°C and repeatedly measured by in-situ powder XRD (total duration: 9-292.5 hrs.). The final run products were dried in vacuum and measured again by powder XRD. (3) Swelling experiments: some dried run products were immersed in ethylene glycol, glycerol or water and measured by powder XRD to check swelling.

Layer silicates formed in all the run products even at room temperature. Brucite and sometimes magnesite were formed in runs using F. XRD reflection peaks of the layer silicates are weak and broad particularly for the (001) peak suggesting that they are thin and have low crystallinity. The (001) lattice spacing and the chemical compositions of the run products together with the results of the swelling experiments indicate that the layer silicates are disordered mixed layer minerals of stevensite (Mg-smectite) or vermiculite, serpentine and talc. The (001) peak becomes sharper to some extent and shift to higher 2θ (smaller lattice spacing) with increasing run duration or temperature except for runs using E. The crystallinity and degree of mixing order also changed by drying process. The experimental results can be explained by that aqueous solutions of high degree of supersaturation with respect to layer silicate minerals formed by instantaneous dissolution of metastable amorphous silicate nanoparticles with high reactivity, and disordered mixed layer minerals metastably formed from these solutions. The presence of disordered mixed layer minerals in some carbonaceous chondrites, such as in CI [8], indicates that they formed from amorphous silicates. It should also be noted that we could not discuss hydrothermal conditions based on the present features of layer silicate minerals in meteorites, as they could be different from those during hydrothermal alteration.

[1] Jones and Brearley (2006) GCA, 70: 1040. [2] Ohnishi and Tomeoka (2007) MAPS, 42: 49. [3] Kemper et al. (2004) ApJ, 609: 826. [4] Bradley et al. (1994) GCA, 58: 2123. [5] Greshake (1997) GCA, 61: 437. [6] Nakamura et al. (2005) MAPS, 40: A110. [7] Noguchi et al. (2008) MAPS, 43: A177. [8] Tomeoka and Buseck (1988) GCA, 52: 1627.

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