

HYDROTHERMAL ALTERATION EXPERIMENTS OF AMORPHOUS SILICATES: DEPENDENCE OF WATER/ROCK RATIO.

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Introduction: Carbonaceous chondrites such as CI, CM, CR, CV and Tagish Lake chondrite, have primitive chemical compositions, but have experienced aqueous alteration in their parent bodies. In order to examine the aqueous alteration process, many hydrothermal experiments using crystalline silicates and natural samples, such as chondrites and IDPs, have been performed (e.g., [1]). In contrast, circumstellar silicate dusts around young stars are mixtures of amorphous and crystalline silicates based on infrared astronomical observations (e.g., [2]). In addition, unique primitive carbonaceous chondrites, such as Acfer 094, have partly crystallized amorphous silicates as abundant component in matrix [3]. Therefore it is important to investigate aqueous alteration of amorphous silicate as well as crystalline silicates.

Based on the above consideration, we have performed hydrothermal alteration experiments of the mixture of amorphous silicates and crystalline silicates at 100-200°C [4]. In these experiments, we showed that saponite, serpentine, calcite and magnetite, which are found in aqueously altered chondrites, formed by alteration in the de-ionized water and the mineral assemblage obtained in the later stage of the alteration resembles to that of CM chondrite. In a series of the experiments, water to rock mass ratio (W/R ratio) was 332, although W/R ratios in aqueous alteration of carbonaceous chondrites are estimated to be lower and vary with their groups (e.g., [5]).

Experimental: In order to understand the effect of W/R ratio on aqueous alteration process of carbonaceous chondrites, we have carried out hydrothermal alteration experiments using Fe-free amorphous silicates. We synthesized glass of the system, SiO₂-MgO-Al₂O₃-CaO-Na₂O, with the CI chondrite chemical composition by quenching method. Three major elements, Fe, Ni and S, were excluded from the system for simplicity to avoid problems of controlling their redox states. The starting materials were ground to make micron-scale powders and were heated in de-ionized water at 150°C for 3 and 8 weeks with different W/R ratios ranging from 0.1 to 332 under water saturated vapor pressures (approximately 3.0 bars) in Teflon vessels sealed by a stainless jacket. After heating experiments, vessels were rapidly cooled to room temperature in water. Run products recovered from vessels were dried in vacuum at room temperature.

Run products were analyzed with powder X-ray diffraction (XRD; Geigerflex, Rigaku) and observed with field emission scanning electron microscope (FE-SEM; JSM-7001F, JEOL) with energy-dispersive X-ray spectroscopy (EDX; Inca, Oxford) and transmission electron microscope with EDX system (TEM).

Results and Discussions: XRD analysis showed two major phyllosilicates, serpentine and saponite were formed in alteration experiments at 150°C in de-ionized water. Unaltered glasses were existent in 3 weeks experiments, although more glasses were hydrated in 8 weeks experiments. Serpentine was formed at W/R ratio is equal to or higher than 0.5, but saponite was not formed at highest W/R ratio (322) and shorter duration and lower W/R ratio (3weeks and W/R ratio = 0.5). Hydration process progressed slower with decreasing of W/R ratio and little or no hydration proceeded at the extreme low W/R ratio (0.1).

FE-SEM/EDX observations showed that calcium composition normalized to silicon (Ca/Si) was decreased from starting materials in hydrated glasses. Na/Si indicate similar tendency then chemical composition of hydrated glass became Ca and Na poor. Phyllosilicate minerals which fill a gap among hydrated glass showed intermediate composition of saponite and serpentine.

These results probably indicate that phyllosilicates were formed in following process. At the early stage of alteration, Ca and Na were preferentially dissolved to water and hydrated glasses became serpentine. If the hydration process stops in this stage, serpentine only forms (3weeks and W/R ratio = 0.5 in our experiments). If water is very few, it is consumed in the hydration process. If moderate amount of water exists in the system, these elements are saturated in water and saponite and serpentine are formed by precipitation from water. If a large amount of water exists in the system, Ca and Na fully dissolve to water then hydrated glass should be serpentine (3weeks and W/R ratio = 332 in our experiments).

Serpentine is major phase in matrix of CM chondrites but saponite is rare. On the other hand saponite and serpentine are major phase in CI chondrites. In the present experiments, saponite was formed at later stage of alteration process or at relatively higher W/R ratio. These results are consistent with previous studies

which suggested CI chondrites are formed at higher W/R ratio than CM chondrites (e.g., [5]).

In addition to layer silicates, calcite was formed at W/R ratio is equal to or higher than 1.0. A relatively small amount of aragonite which is stable at higher pressure was formed at W/R ratio = 0.5. Micron scaled aragonites which were discovered in CM chondrites were possibly formed in the aqueous alteration process at lower W/R ratio.

References: [1] Ohnishi and Tomeoka, 2007. *Meteoritics & Planetary Science* 42: 49-61. [2] Honda et al., 2003. *The Astrophysical Journal* 583; 59-63. [3] Greshake, 1997. *Geochimica et Cosmochimica Acta* 61:437-452. [4] Noguchi et al., 2009. *Japan Geoscience Union Meeting 2009*. [5] Crayton and Mayeda, 1999. *Geochimica et Cosmochimica Acta* 63: 2089-2104