AQUEOUS ALTERATION EXPERIMENT USING GEMS ANALOG PARTICLES. S. Enju, A. Tsuchiyama, Y. Imura and A. Takigawa, Research Organization of Science and Technology, Ritsumeikan University (satomi.enju@gmail.com). 2 Guangzhou Institute of Geochemistry, Chinese Academy of Science. 3 Department of Earth and Planetary Sciences, Graduate School of Science, Kyoto University. 4 The Hakubi Center for Advanced Research, Kyoto University.

Introduction: Glass with embedded metal and sulfides (GEMS) is one of the major components of CPIDPs (Chondritic Porous Interplanetary dust particles), considered as one of the most primitive materials in the solar system [1]. GEMS has the chemical compositions similar to the CI chondrite composition and shows characteristic textures composed of amorphous silicates with nanoparticles of Fe, Ni metal and Fe, Ni sulfides [2, 3]. Also, GEMS-like materials similar to GEMS are found in several primitive carbonaceous chondrites (pCCs) [4, 5, 6]. The difference from GEMS is that amorphous silicate is relatively rich in Fe, more abundant in sulfides than metals, and some Ni-rich sulfides are observed.

Amorphous silicate in GEMS is highly sensitive to aqueous alteration compared with coexisting crystalline silicate minerals. In the aqueous alteration experiment by [7], GEMS were altered to serpentine (and saponite) in all conditions at high water-rock ratio (W/R >1000). GEMS-like materials were thought to be made by weak aqueous alteration of GEMS, considering the close coexistence with the layered silicates [4]. However, particles similar to GEMS and GEMS-like materials were produced in our previous condensation experiments [8]. To reveal the relationship between GEMS and GEMS-like materials, we need to consider both the effect of the condensation process and the aqueous alteration process.

In this study, we performed a condensation experiment to produce GEMS analog particles and aqueous alteration experiment using the GEMS analog particles to reveal the alteration process of GEMS.

Experiments: Induction thermal plasma (ITP) system was used for the condensation experiment. This system creates a high-temperature plasma (around 10^4K) which evaporates injected starting materials and a high temperature gradient of the gases where fine particles condense. The starting material is a mixture of powder regents (SiO₂, Si, MgO, Fe, FeS₂, Ni, Al₂O₃, CaO) with GEMS mean composition [3]. We adopted the same experimental conditions as the intermediate redox condition in [8], where condensed particles are most similar to GEMS. Run product was used as a starting material of the aqueous alteration experiment.

The procedure of the aqueous alteration experiment method is basically the same as [9]. Powder starting material was mounted on a closed XRD holder and deionized pure water (W/R = 5) was gently dropped on the surface. In order to prevent the formation of carbonates, the water was boiled in the N₂ gas flow and the dissolved CO₂ gases were reduced. In-situ XRD measurement was performed during aqueous alteration at room temperature for 66 hrs. The sample was dried after the experiment for the following analysis. All the procedures were done under nitrogen atmosphere.

The starting material and run products were analyzed using X-ray diffraction (XRD), scanning electron microscope (SEM), and transmission electron microscope (TEM).

Results: The run product of the condensation experiment, used as the starting material of the aqueous alteration experiment, was mostly composed of amorphous silicate nanoparticles and smaller particles of Fe, Ni metal, and Fe, Ni sulfides were often present on their surface and rarely inside (Fig. 1). Relatively large amorphous silicate grains (> 50 nm) were spherical, while smaller ones showed irregular shapes. The chemical composition of amorphous silicate particles was not homogeneous. The Mg/Si ratio of the grains varies between 0.08 and 2.26.

During the in-situ aqueous alteration experiment, the intensity of the broad XRD peak of amorphous silicate decreased. However, any characteristics of hydrous layered silicates, even of poorly-crystalline phase (MSH: Mg silicate hydrate) that were formed using pure Mg amorphous silicate [9] were not observed. After drying, the sample showed a layered structure with a black layer on the surface and a greyish green layer beneath. In the XRD pattern, the broad peak of clay mineral was observed in the greyish green part, while the black part was similar to the starting material.

In TEM analysis, the alteration products could be roughly divided into 3 groups (Fig. 2): spherical amorphous silicate with nano-particles of Fe, Ni metal and Fe, Ni sulfides (group 1; Fig. 2-a, d), which is mostly the same as the starting material, aggregate of extremely thin sheets (group 3; Fig. 2c, f) in which nanoparticles of Fe, Ni metal and Fe, Ni sulfides were scarcely seen, and a mixture of (1) and (3) with clear borders between them (group 2; Fig. 2b, e). The alteration degree of group 3 is larger than group 1. The Fe content of amorphous silicate in group 3 was larger than group 1, while the bulk analysis of the entire grain showed a similar composition regardless of the group. In SEM observation, the black part was mainly composed of
group 1 grains, while the greyish green part was mainly composed of group 3 grains.

**Discussion:** The aqueous alteration was very heterogeneous in both megascopic and microscopic scales, indicating the heterogeneity of water-rock ratio in the sample during alteration. Meanwhile, the bulk chemical composition was mostly homogeneous regardless of the alteration degree, preserving the bulk composition of the starting material. This heterogeneity of the alteration degree well resembles the occurrence of GEMS-like material coexisting with phyllosilicates [4]. However, preferred disappearance of Fe, Ni metal and formation of Ni sulfides were not observed in this experiment. This may be due to the difference in starting (condensed) material or the alteration environments, which is going to be examined in future experiments.

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Fig. 1. (a) TEM image (bright field) and (b) X-ray mapping image of representative grains formed by condensation experiment.

Fig. 2. TEM images (bright field) and X-ray mapping image of run products of aqueous experiment. TEM image of a representative grain of (a) group1, (b) group 2, and (c) group 3. (d) and (f) Enlarged TEM images of (a) and (c) respectively. (e) Overlaid X-ray mapping image of (b) group 2 grain. The right part on (e) is similar to group 1 including abundant nanoparticles, while the left part similar to group3 rarely including nanoparticles.