Synthesis of GEMS analogue particles with condensation experiments in the system Fe-Mg-Si-O-S

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Amorphous silicate particles called GEMS (glass with embedded metal and sulfides) are included in cometary dust particles [1.2]. GEMS are submicron-sized (100-500 nm) particle composed of amorphous silicate and nanometer-sized (10-20 nm) Fe-Ni metal and Fe sulfides. Metals are typically present inside of an amorphous silicate grain while sulfides on its surface [1,3]. GEMS are considered to be one of the most primitive materials in solar system and understanding its origin is important to clarify the origin of the solar system materials. The origin of GEMS is still in controversy although some origins are proposed; GEMS were formed with non-equilibrium condensation from protoplanetary disk gas [1] and GEMS were formed by irradiation of charged particles to interstellar crystalline grains [2]. Some condensation experiments based on [1] have been performed so far to reproduce GEMS analogue particles [3,4]. However, experiments using sulfur-bearing system have not been performed. In this study, condensation experiments using sulfur-bearing system were performed systematically by changing redox conditions to understand the origin of GEMS.

In the present experiments, induction thermal plasma (ITP) system(JEOL, TP-40020NPS) was used to reproduce GEMS-like materials by condensation. ITP system can provide high temperature (~10^4K) for vaporizing starting materials and high cooling rate for condensation of nanomaterials from gas. Powder of starting materials (SiO_2, Si, MgO, Fe, FeS_2) with GEMS mean composition (Mg:Fe:Si:S=0.7:0.6:1:0.3 mol ratio) were used as starting materials. The mixing ratio of Si and SiO_2 was changed to obtain different redox conditions; $Si/(Si+SiO_2)$ = 0 (Run1), 0.25 (Run3) and 0.5 (Run2). By using the same starting material as used in Run3, an additional experiment was carried out (Run4) with direct current plasma (DCP) system (Technoserve, YC-500TSPT5), which can generate higher temperature (~1.5×10^5K) and higher cooling rate than ITP system. Run products were characterized using powder X-ray diffraction (XRD, SmartLab, Rigaku), Fourier transform infrared spectrometry (FT-IR, MFT- 680, JASCO), transmission electron microscopy (TEM, JEM-2100F, JEOL) and electron-beam tomography with TEM (TEM-CT) for the 3D structures.

In Run1 with oxidized condition, sulfur-bearing amorphous silicate was mainly synthesized. In Run3 with moderate redox condition, amorphous silicate, iron and a small amount of iron sulfide (troilite) were confirmed by XRD pattern. FT-IR spectrum has a broad peak at 9.8 μm, which can be attributed to amorphous silicate. Submicron-sized particles (~100 nm) composed of amorphous silicate, which contains a small amount of sulfur, and nanoparticles of iron and iron sulfide (10-20nm) were confirmed by TEM/EDS. In Run2 with reduced condition, amorphous silicate particles containing Fe_3Si (gupeiite) and MgS (niningerite) were formed. In Run4 with DCP system, nanoparticles containing a larger amount of iron sulfide than Run3 was formed. TEM-CT result showed that iron sulfides are present on the surfaces of amorphous silicate grains, consisting with the distribution of iron sulfides in GEMS [1,3], although irons were also present on the surfaces.

The run products similar to GEMS were obtained in Run3 and Run4, suggesting that GEMS were formed in conditions close to these experiments. Meanwhile, nanoparticles somewhat different from GEMS were

produced in Run1 and Run2 under different redox conditions with slightly different oxygen contents. These results indicate that condition for GEMS formation is limited to a narrow range.

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