

POSSIBLE ORIGIN OF PRIMITIVE AMORPHOUS SILICATES IN CARBONACEOUS CHONDRITES.

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Introduction: Less-altered carbonaceous chondrites (CCs) called primitive CCs have fine-grained matrix of amorphous silicate, silicate mineral grains, complex organic compounds, and relatively abundant presolar grains. The amorphous silicates more or less resemble glass with embedded metal and sulfides (GEMS) observed in chondritic porous interplanetary dust particles (CP-IDPs) (e.g., [1]). Such GEMS-like materials in primitive CCs are considered to represent the building blocks of the solar system [2-4] as well as GEMS and micrometeorites (e.g., [5]). Primitive CCs are present in various CC groups (CM, CR, CO, and C-ungrouped) (e.g., [2-4,6-8]). GEMS-like materials are more or less aqueously altered and show chemical and textural diversity, which is explained by different degrees of aqueous alteration of GEMS or related materials (e.g., [2]) In this paper, a possible process of the formation and alteration of GEMS-like materials in primitive CC matrix is proposed based on the observation of GEMS-like materials and condensation [9,10] and aqueous alteration experiments [11] conducted so far.

Diversity of GEMS-like materials: Some of GEMS-like materials are spherical objects like GEMS and observed in porous regions of Paris (CM) [2] and Acfer 094 (C-ungrouped) [4] and organics-rich regions of MIL 090657 (CR) [12] and LAP 02342 (CR) [3]. The majority of the GEMS-like materials seem to be aggregates of such spherical objects [4]. In contrast to GEMS, abundant Fe(-Ni) sulfide nanograins are observed in GEMS-like materials, and Fe-Ni metal is mostly absent. Some of them contain Fe oxide nanograins (unknown phase in Paris [2], magnetite and goethite in MIL 090657 [12]) and metal nanograins in LAP 02342 [3], DOM 08006 (CO). Amorphous silicates are relatively rich in Fe (Fe²⁺ and Fe³⁺) and more or less hydrated (e.g., [4,13]). The electron diffraction of amorphous silicate usually has rings, indicating poorly crystallized feature [2,4], which correspond to magnesium silicate hydrate (MSH) [4]. Layered hydrous silicates are also observed (e.g., [7]).

Condensation and aqueous alteration experiments: Condensation experiments were performed using gases of the GEMS mean composition in the system Mg-Fe-Si-O-S [9] and those of CI composition in the system Mg-Fe-Si-Al-Ca-Ni-O-S [10] under various redox conditions. In these experiments, fine particles of amorphous silicate with Fe-bearing nanograins, which resemble GEMS and GEMS-like materials, were produced. In oxidizing conditions, abundant FeS nanograins are in Fe-rich amorphous silicates and metal is absent or rare. Monosulfide solid solution (MSS: (Fe,Ni)S) also formed in the Ni-bearing system. In intermediate redox conditions, FeS and Fe or α -(Fe,Ni) nanograins are in Fe-poor amorphous silicates. In reducing conditions, Fe₃Si instead of metal, FeS and MgS nanograins are in almost Fe-free amorphous silicates. Only a small amount of S is included in amorphous silicates.

In in-situ aqueous alteration experiments, amorphous MgSi₂O₄ and MgSiO₃ immersed in pure water at room temperature were observed under TEM or in-situ measured by X-ray diffraction [11]. The amorphous silicates are hydrated first. Then, Mg is selectively dissolved into water and MSH forms from the Mg-rich solution. Layered hydrous silicates particularly mixed layer silicates may form from further alteration of MSH.

Formation and alteration of GEMS-like materials: The above features of the GEMS-like materials strongly suggest that they have experienced aqueous alteration in different degrees. However, aqueous alteration of GEMS ((Fe,Ni), FeS nanograins in amorphous silicate) cannot simply explain the diversity of the GEMS-like materials. For example, abundant Fe(-Ni) sulfide nanograins without metal in the GEMS-like materials requires sulfidation of metal (e.g., [5]), but there is no sufficient source of S for the sulfidation. In addition, Fe-rich amorphous silicate does not easily form via aqueous alteration because it requires dissolution of amorphous silicate and re-precipitation of Fe-bearing MSH. Fe-rich amorphous silicates with FeS (and probably MSS) formed in relatively oxidizing condition are most likely original materials of some GEMS-like materials. We should distinguish between original material and later aqueous alteration features. If this is the case, GEMS and GEMS-like materials were formed by condensation probably in different locations of the primitive solar nebula, and CP-IDPs and the primitive matrix in CCs may represent samples of the building blocks of the solar system in different locations.

References: [1] Keller L. P. and Messenger S. (2011) *Geochimica et Cosmochimica Acta* 75: 5336–5365. [2] Leroux H. et al. (2015) *Geochimica et Cosmochimica Acta* 170: 247–265. [3] Nittler L. R. (2019) *Nature Astronomy* <https://doi.org/10.1038/s41550-019-0737-8>. [4] Matsumoto M. et al. (2019) *This meeting*. [5] Noguchi T. (2017) *Geochimica et Cosmochimica Acta* 208: 119–144. [6] Greshake A. (1977) *Geochimica et Cosmochimica Acta* 61: 437–452. [7] Abreu N. M. and Brearley A. J. (2010) *Geochimica et Cosmochimica Acta* 74: 1146–1171. [8] Nittler L. R. (2018) *Geochimica et Cosmochimica Acta* 226: 107–131. [9] Enju S. et al. (2019) *This meeting*. [10] Imura Y. et al. (2019) *Japan Geoscience Union Meeting 2019* Abstract PCG23-P03. [11] Igami Y. (2019) *Goldschmidt2019* Abstract. [12] Tsuchiyama A. et al. (2018) *Japan Geoscience Union Meeting 2018* Abstract PPS06-01. [13] Le Guillou C et al. (2015) *Earth and Planetary Science Letters* 420: 162–173.