CONDENSATION DURING CHONDRULE FORMATION: ELEMENTAL AND MG ISOTOPIC EVIDENCE.

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Introduction: Recent petrochemical studies have revealed that chondrules were formed in an open system [e.g. 1,2], and models that explain the compositional change by evaporation have been proposed [3,4]. Although volatile loss has been one of the most concerns [5,6], the models include the major elements such as MgO and SiO₂. On the contrary, clear evidence for open system behavior of MgO and SiO₂ has not yet been found. We have studied highly refractory chondrules in Semarkona and found evidence for condensation of MgO, SiO₂, FeO, Na₂O, and K₂O during chondrule formation under the condition of decreasing temperature.

Texture: A polished thin section of Semarkona (USMN 1805-9) was studied with a JEOL 8900L EPMA. Two chondrules were selected for the study, which are characterized by highly refractory compositions, anomalous porphyritic texture, and perfectly rounded shape. Chondrule #5 (CH5) is about 800µm in diameter, and consists of forsterite and glass. Forsterite crystals with ten to several tens of µm size and euhedral to partly hopper shape are rimming the inner surface, which occupy 70-80% of the entire surface. There is only one tabular forsterite crystal with 80x200 µm size in the apparent center. Modal concentration of forsterite is 10-20 vol%. Another portion of the chondrule is clear glass.

Chondrule #13 (CH13) is about 500µm in diameter, and consists of forsterite and glass. Rectangular to tabular forsterite crystals grow from the surface, and one of it grows inward to form a 250µm large hopper crystal. About 70% of the chondrule surface is occupied by forsterite, and 30-40 vol% of the entire chondrule is occupied by forsterite. Glass is clear under an optical microscope. CH13 is texturally similar to CH5 except for a little higher forsterite modal concentration.

Chemical Composition: Both CH5 and CH13 are highly refractory; forsterite is poor in FeO (<1.0 wt%), and glass is rich in CaO and Al₂O₃. Forsterite in CH5 shows Al₂O₃ and CaO decrease and FeO increase from the core to rim. The concentration of those trace elements ranges from 0.05 to 0.46 wt% (Al₂O₃), 0.26 to 0.4 (CaO), and 0.44 to 1.0 (FeO), which are similar to forsterite in other type IA chondrules [7] or group A1 chondrules [8], but differs in that the CaO content is almost constant. Forsterite in CH13 is zoned with Al₂O₃- and CaO-depletion and FeO-enrichment from the core to rim, and those near the margin show growth zoning in terms of Al₂O₃. Contrary to fairly constant CaO concentration in CH5 forsterite (0.3-0.4 wt%), those in CH13 (0.4 to 0.65 wt%) is linearly correlated with Al₂O₃ (0.1 to 0.4 wt).

Groundmass glass in CH5 and CH13 is strongly zoned. Al₂O₃, CaO, MgO and TiO₂ decrease from the center to surface (Fig. 1). Local heterogeneity of strong Al₂O₃ enrichment, weak CaO enrichment, and MgO depletion around forsterite crystals are found, which is attributable to forsterite crystallization. SiO₂ decreases monotonously from the surface to the center, whereas Na₂O and K₂O are enriched only near the surface, of which distribution is scarcely affected by forsterite distribution. Na₂O is specifically enriched along curved veins which resembles perlitic texture in rhyolite glass formed by cooling contraction or expansion due to hydration.

If we ignore the effect of crystallization of forsterite, the glass composition of CH5 shows positive correlation between Al₂O₃ or TiO₂, and MgO (Fig. 1) and negative correlation between FeO, Na₂O, or SiO₂ and MgO (Fig. 2). CaO is weakly negatively correlated with MgO. CaO in CH13 is positively correlated among Al₂O₃ and TiO₂. The glass before or in the early stage of crystallization of forsterite has composition of 50 wt% SiO₂, 21 Al₂O₃, 17 CaO, 12 MgO, and 1 TiO₂ for CH5, and 48, 21, 20, 9, and 1, respectively for CH13.

Mg Isotopes: Mg isotopic compositions were obtained with Cameca IMS-1270 SIMS with reference of ²⁶Mg/²⁵Mg of 0.12663 [9] and ²⁶Mg/²⁴Mg of 0.139354, the mean value of mass fractionation corrected from the terrestrial standards. The measured Mg isotopic ratios of forsterite and glass are between -3 and -4 permil/amu and +0.5 and +2 permil/amu, respectively, which are in the similar ranges for the instrumental mass fractionation of the terrestrial
olivine and glass standards. Therefore, Mg isotopes of forsterite and glass is assumed to be unfractonated. Mg isotopic compositions of glass in CH5 and CH13 is homogeneous regardless of spatial location within the chondrules and difference in chemical compositions. Mg isotopic composition of forsterite is related to the Al2O3 concentration, which is heavier in the Al2O3-rich portion and lighter in the poor region. (Fig. 2).

Fig. 2 Mg isotopic composition of forsterite.

Discussion: The texture, chemical composition of forsterite and glass, and elemental distribution suggest the following formation scenario for CH5 and CH13. They were completely molten droplets of which chemical composition is rich in Al2O3, CaO, SiO2 with lesser amount of MgO. It is not clear whether the melt droplets were formed by direct condensation or as evaporation residue of pre-existing condensed phases. We can not read the records before forsterite crystallization. With decreasing temperature, SiO2 condensed and diffused to the interior. The droplets were homogeneous at least before crystallization of forsterite, which is shown by the presence of homogeneous highly refractory region in the glass. When the melt composition reached the olivine saturation due to SiO2 condensation, forsterite started to crystallize mostly from the chondrule surface. The high Al2O3 content in the forsterite core is due to higher Al2O3 content in the melt at the earlier (higher temperature) stage of forsterite crystallization. During forsterite crystallization, MgO and small amount of FeO also condensed, because Mg isotopic composition and FeO content in the melt changed during crystallization. Crystallization of forsterite resulted in the enrichment of Al2O3 and CaO and depletion of MgO around the crystals. SiO2 condensation continued after forsterite crystallization, because SiO2 is coming into the chondrules from the surface where glass is directly exposed to the surface. Further temperature decrease caused Na2O and K2O condensation. The penetration distance of Na2O from the surface to interior in about 100µm and that of K2O in several tens of µm. The difference may be due to difference in condensation temperature or diffusion rate. After consolidation of the chondrules, the chondrules suffered Na2O condensation only on the surface and preferentially along the cracks formed by contraction of the sphere. This condensation did not affect the elemental distribution in the interior. The concentration of Na2O in the outermost portion of the chondrules is higher than the chondrule rim. Thus, this later-stage Na condensation should be predated to accretion of chondrule rim and matrix materials.

The texture of forsterite crystals rimming the surface is similar to that synthesized in crystallization under significant evaporation (our unpub. data), where crystallization took place due to rapid compositional change during incongruent evaporation. The elemental and mineralogical evidence in CH5 and CH13, however, is not indicative of evaporation process, but is suggesting condensation-induced crystallization during temperature decrease. The heavier Mg-isotopic composition of Al2O3-rich portion of the forsterite than the poor portion can be two possible origin. One is that the precursor materials were heated to become isotopically heavier, and subsequent condensation of lighter Mg resulted in crystallization of forsterite with isotopically lighter composition at the later stage of crystallization. Alternatively, Mg isotopic fractionation took place during condensation of Mg from heavier to lighter composition if the melt droplet initiated as a condensation product.

CH5 and CH13 are the first evidence for direct condensation of SiO2 and MgO which is directly responsible for chondrule formation. To reveal the earlier history of the droplets and quantitative temperature and time sequence of chondrule formation, isotopic data for other elements, particularly for Si and O, will be needed. Quantitative modeling of evaporation, condensation, crystallization, and diffusion will tell us the entire story.