

Gas-melt interaction experiments at high temperature and high SiO(g) partial pressure – implication of melt composition to chondrule formation. A. Kropf¹ and G. Libourel^{1,2}, ¹CRPG-CNRS, 15, Rue Notre-Dame des Pauvres, BP20, 54501 Vandoeuvre les Nancy, France, (akropf@crpg.cnrs-nancy.fr) ²ENSG-INPL, BP40, 54501 Vandoeuvre les Nancy, France.

Introduction: Chondrules are major parts of primitive meteorites. They are typically mm-sized and consist of olivine, pyroxene, sulfide, metal and glass. Mainly based on spherical shape and characteristic structure, chondrules were suggested to have formed by a brief but very intense heating event in a solar nebular environment [1,2]. Interactions between nebular gas and chondritic melt droplets could have played a major role in the early solar system [3]. But the extent to which chondrules reacted with the surrounding gas, is still under debate [3]. Silicon is a moderately volatile element [4] and the cation with the highest abundance in PO, POP and PP chondrules.

[3] present high temperature experiments where synthetic chondrule-like samples were exposed to elevated SiO(g) partial pressures (P_{SiO}). Silicon is suggested to get evaporated and to form elevated SiO(g) partial pressures. This could be responsible for recondensation into the melt. [5] report on Si-rich igneous rims (SIRs) in the outer portions of many type-1 chondrules and suggest that they are either formed by a gas-solid condensation of Si-normative materials onto the chondrule surfaces or by direct SiO(g) condensation into the chondrule melts. [6] explore changes in type-1 chondrule glass compositions and conclude that type-1 chondrules didn't obey closed system formation.

In order to explore this putative nebular gas-melt interaction, we conducted new silica evaporation/condensation experiments in the system CMS at high temperature and in reducing atmosphere.

Methods: Samples with three different starting compositions (CMS-1 (forsterite), CMS-2 (16 wt.% CaO) and CMS-3 (31 wt.% CaO), see **fig. 1**) were prepared by using the Pt-wire loop technique (e.g. [7]). All samples were pretreated for 24 hours at the experimental temperature ($T = 1450^\circ\text{C}$) in a muffle furnace, followed by quenching. This results in porphyritic olivine grains in a quench phase as starting texture. In contrast to [3], we used a powder of "amorphous" SiO (=Si+SiO₂, *Patinal*) to produce an atmosphere with a high SiO partial pressure. Using data of [8] and the equation $K = P_{\text{SiO}}^2/a_{\text{Si}} \cdot a_{\text{SiO}_2}$ we calculated $P_{\text{SiO}} = 0.1$ bar and $\log f_{\text{O}_2} = -6$. The SiO-source was placed on the bottom of a small alumina crucible. The sample was hanging a few millimeters above the source. The alumina crucible was covered by a larger one to close the system and both crucible were placed in a third crucible that was hanging in a vertical tube furnace.

Results: The product of the CMS-1 experiment (60 min) contains some forsterite, that is enclosed by enstatite. Finally, these minerals are surrounded by a

dominant glassy phase with > 90 wt.% SiO₂. CMS-2 and -3 experiments lasting 40 or 60 min resulted in the presence of large, euhedral low-Ca pyroxene (enstatite) crystals located in a quench phase. The quench phase consists of mainly pigeonite and minor parts of high-Ca pyroxene. Enstatite is also occurring in the center areas of the pigeonite (see **fig. 2**) whereas high-Ca pyroxene is observed as border phase to the glass. Over the entire range of time, the products of the experiments show elevated SiO₂ bulk concentrations. The longer an experiment lasted the higher became the SiO₂ bulk value and the lower became the ratio of olivine crystals in the melt (see **fig. 3**). Long-time products also show Si-rich rims with 90-100 wt.% SiO₂ (**fig.2**).

Discussion: The changes of the sample bulk compositions in the ternary diagram reflect interactions of Mg-Ca-silicates with added SiO₂. The CMS-2 starting matter produces forsterite as liquidus phase. Thus, the coexisting melt has a CaO content of about 30 wt.%. With increasing time (and additional SiO₂) the melt composition changes along the 1450°C-isotherm until the stability field of cristobalite is reached (**fig. 1**). For CMS-3, the crystallized phases at 1450°C are Ca-rich olivine (e.g. monticellite, CaO · MgO · SiO₂) that is located on the extrapolation of the bulk evolution line. Therefore the coexisting melt composition is analogue to the bulk evolution. The observed Si-rich rims reflect an equilibrium between melt and gas: $\text{SiO(g)} + \frac{1}{2}\text{O}_2 \rightleftharpoons \text{SiO}_2\downarrow$. The presence of Ca in the system inhibits the reactions between olivine, pyroxene and quartz.

The sample CMS-1 starts with 43 wt.% SiO₂ (bulk) and forsterite and melt as phases. At 1450°C, an addition of SiO₂ leads first to a common precipitation of forsterite and enstatite and then of enstatite and quartz as coexisting phases (see phase diagram). At 1450°C and in contrast to CMS-2 and -3, all phases are below the subsolidus. Thus, a comparison of CMS-1 data with CMS-2 and -3 experiments is difficult and more investigation is necessary.

The sample data confirm theoretical calculations, based on the phase diagram. SiO₂ has entered the samples and changed significantly the mineralogy within 40 minutes. Mineral paragenesis and texture show strong parallels to chondrules (formation sequences, Si-rich rims with nearly pure SiO₂ phases [5]).

We support a model of chondrule formation in a nebular region with high P_{SiO} (due to increased dust/gas ratio?), This should cause a direct condensation of SiO₂ into the melt and finally result in SiO₂ enrichments parallel to the chondrule rim [3,5,6].

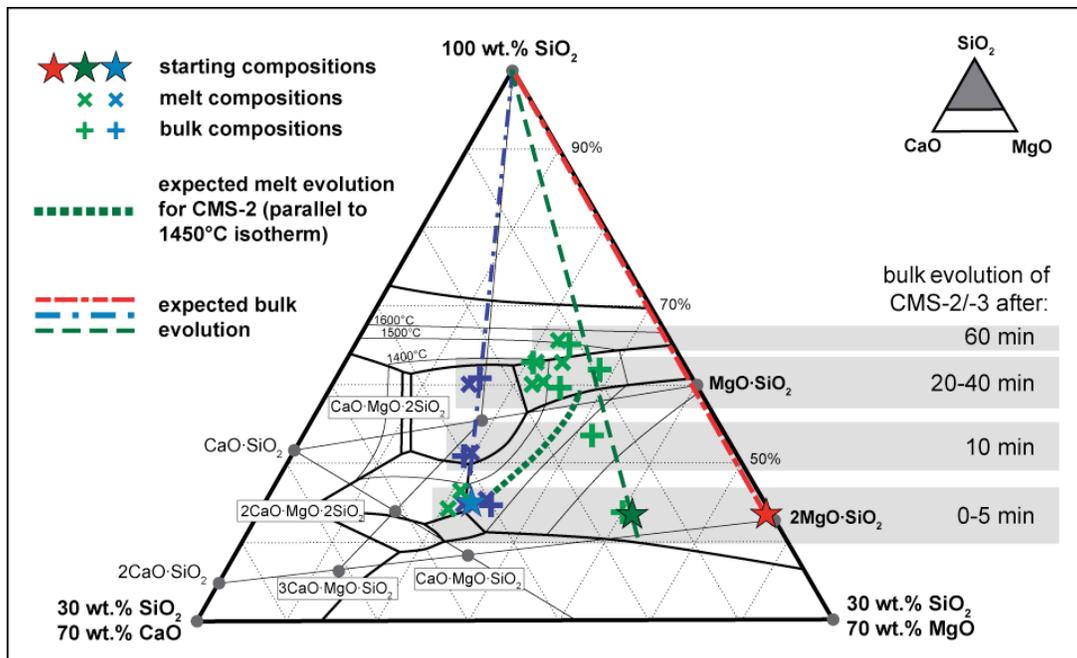


Fig.1: Ternary diagram of the system CaO-MgO-SiO₂ with the starting compositions and the bulk and melt compositions of the 1450°C experimental products (red = CMS-1, green = CMS-2, blue = CMS-3).

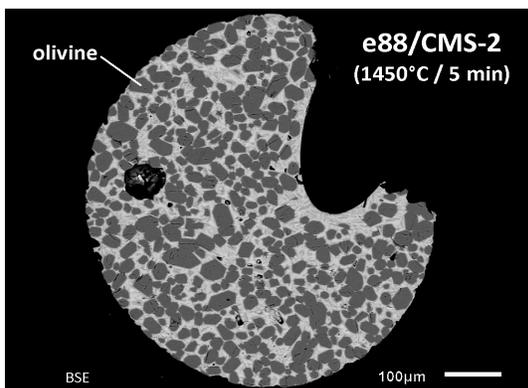


Fig. 2a: Sample e88/CMS-2 (after 5 min). Olivine and coexisting melt

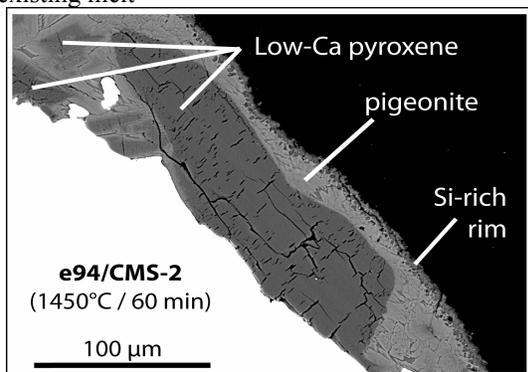


Fig. 2b: Sample CMS-2 (60 min). Low-Ca pyroxene as euhedral crystal and also poikilitically enclosed by pigeonite and high-Ca pyroxene. Note the Si-rich rim of the sample.

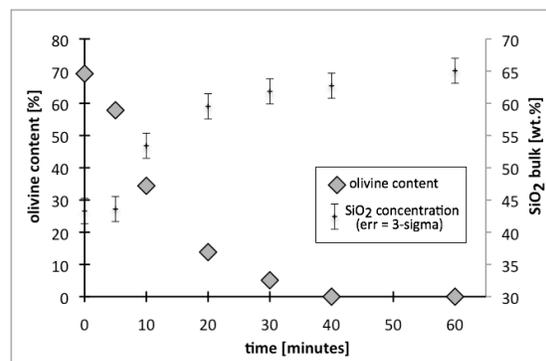


Fig. 3: change of SiO₂ (bulk) and olivine content with the time in CMS-2 experiments.

References: [1] Hewins R. et al. (2005) in *Chondrites and the Proto-planetary Disk*, Krot A. et al. (eds.), Astron. Soc. of the Pacific, 286-316. [2] Scott E. R. D. (2007) *Ann. Rev. EPSL*, 35, 577-620. [3] Tissandier, L. et al. (2002) *MAPS*, 37, 1377-1389. [4] Lodders, K. and Fegley, B. (1998) *The Planetary Scientists Companion*. Oxford Press. [5] Krot, A.N. et al. (2004) *MAPS*, 39, 1931-1955. [6] Libourel, G. et al. (2006) *EPSL*, 251, 232-240. [7] Borisov, A. et al. (2006) *Am. Min.*, 91, 762-771. [8] Lou, V.L.K et al. (1985) *JAmCerSoc*, 68, 49-58.