

Synthesis of refractory minerals by high-temperature condensation of a gas of solar composition. A. Toppani^{1,2}, G. Libourel^{2,3}, F. Robert¹, J. Ghanbaja⁴ and L. Zimmermann², ¹Muséum National d'Histoire Naturelle, France (toppani@crpg.cnrs-nancy.fr), ²CRPG-CNRS, France, ³Ecole Nationale Supérieure de Géologie, France, ⁴Université Henri Poincaré (UHP), Nancy, France.

Introduction: Condensation processes are often proposed to explain the formation of the first objects of our solar nebula, i.e., Ca-Al-rich inclusions (CAIs) [1]. Indeed, CAIs show (i) refractory minerals whose formation and compositions have been predicted by theoretical calculations of condensation from a cooling solar gas [2] and (ii) peculiar fluffy textures, which have no terrestrial equivalent [3]. Even if volatilization of chondritic objects or crystallization from a melt could form some of the refractory minerals of CAIs [4,5], these processes do not explain, for example, these fluffy textures or the mineralogical rimming of the fine-grained CAIs.

However, since the first descriptions of CAIs, only few set of condensation experiments [6, 7] were performed mostly because of experimental complexity such as the production of a gas of solar composition or the control of the composition of the gas to be condensed. We have thus set up a new experimental design which allows to perform condensation from a gas of controlled composition up to 1600 K in a pressure range from 10^{-8} to 1 bar, under controlled oxygen fugacity for any run times. Here, we report high-temperature condensation experiments that address questions about nature, crystallinity, size or kinetics of condensation of the condensates and their implications to the CAIs minerals formation.

Experimental: The design consists of a reaction chamber connected to a high-vacuum metal line in which oxygen fugacity is controlled by flowing gas mixtures (Ar, CO₂, CO, He...). In the reaction chamber (fig. 1), the gas to be condensed is produced by laser ablation (Nd-Yag laser - 1064 nm) of a glass target of known composition. Thanks to the use of this laser, the produced gas is not fractionated relative to the target. Condensation of the gas occurs at a controlled temperature inside a platinum furnace located above the target (see fig. 1). Condensates are deposited directly on a Pt-grid welded to the end of a Pt-Rh10 thermocouple held by an alumina rod entering the furnace. In order to control location of condensation, the Pt-furnace was designed to show a positive temperature gradient from its bottom to its top. Condensation temperature are known with an accuracy of $\pm 20^\circ\text{C}$. High-temperature experiments last about 1 hour to optimize the quantity of deposited matter on the Pt-grid versus the amount of laser-ablation droplets. Each experiment was performed in static vacuum mode.

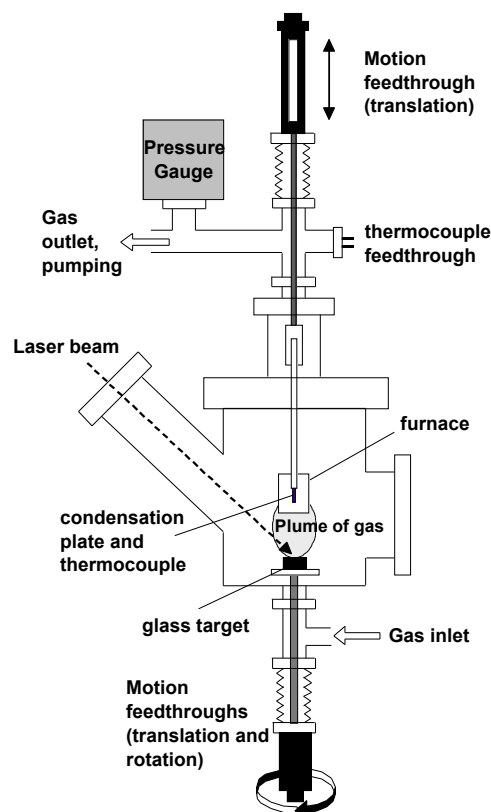


Figure 1: Schematic drawing of the reaction chamber.

Pt-grids were studied by transmission electron microscopy at UHP (Nancy) to determine the texture, the composition and the crystallinity of the condensates.

Results and Discussions: One-hour condensation experiments were performed in CO₂ or Ar background gas at a total pressure of 10^{-3} bar. The gas to be condensed was of solar composition restricted to 4 elements: Ca, Mg, Al and Si. In order to simulate predicted CAIs formation conditions, temperatures of condensation were varied from about 1300 K to 1600 K.

In these conditions, we succeed to condense refractory minerals such as corundum, hibonite, spinel, anorthite, melilite, Al-diopside, forsterite and

enstatite. For all these phases, observed condensates were crystals. Depending on their mineralogy, some were predominantly euhedral, sub-euhedral (e.g. Al-diopside, spinel) or anhedral (e.g. anorthite). Furthermore, condensed crystals show different textures. They can be single or associated with crystals of similar composition, sometimes showing chain-like structures (fig.2). All of these characteristics are in favor of condensation sensu-stricto (i.e. nucleation and growth from the vapor phase), excluding any processes of sintering of amorphous matter. The size of the crystals varying from about 30 nm to about 300 nm seems to depend on (i) the temperature of the condensation and (ii) the mineralogy.

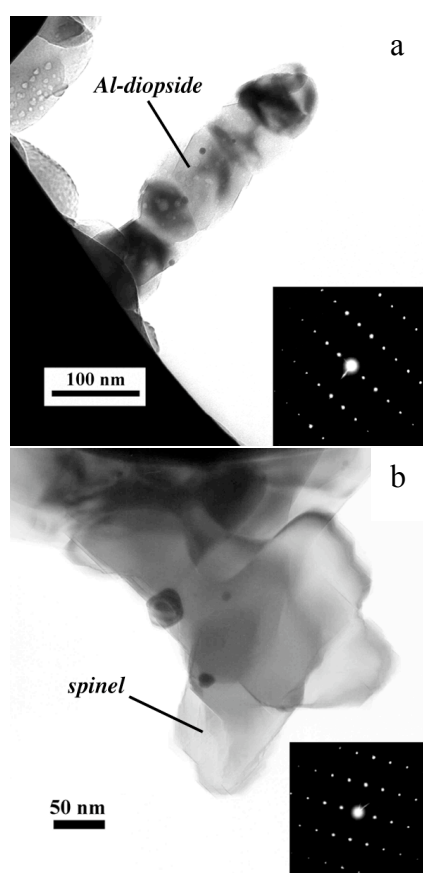


Figure 2: TEM image of condensates showing different textures. (a) Crystals of Al-diopside showing a chain-like structure. Image inserted in the bottom-right corner is the SAED pattern along the $[-112]$ zone axis of one of the crystals. (b) Crystals of spinel on the Pt-grid with a more complex texture. Image inserted in the bottom-right corner is the SAED pattern along the $[114]$ zone axis of one of the crystals.

Interestingly, the mineralogical paragenesis of the condensates seems to vary according to the temperature of condensation. A preliminary study suggests that the higher the temperature, the more refractory the minerals are. For example, crystals of spinel and corundum are the major phases around 1600K, whereas at about 1350K, forsterite, spinel and Al-diopside are the main phases to occur.

Although uncertainties remain about attainment of equilibrium and gas partial pressures during the experiments, the temperature of occurrence of the condensed minerals are qualitatively in agreement with those theoretically predicted at equilibrium. Further comparisons with theoretical calculations will bring information about kinetics of condensation of phases and kinetics of gas-solid reactions. For example, spinel seems to be present at all condensation temperature in large quantity, which suggests a fast kinetics of condensation (including nucleation and growth of the phase) or/and a very low reactivity with gases.

Conclusions: For the first time, CAIs minerals were condensed at high temperature ($\sim 1300\text{K}$ to 1600K) from a gas of solar composition in laboratory conditions. This work brings the first experimental support to the idea that CAIs were formed by condensation, which was mainly based on theoretical calculations since 30 years. It will also bring new information about kinetics of condensation of CAIs minerals and thus, about their conditions of formation.

References: [1] Christophe M.I. (1968) *Bull. Soc. Fr. Minéral. Cristallogr.*, 91, 212-214. [2] Grossman L. (1969) *Geochim. Cosmochim. Acta*, 36, 597-619. [3] Macpherson G.J. et Grossman L. (1984) *Geochim. Cosmochim. Acta*, 48, 29-46. [4] Hashimoto A. et al. (1979) *Earth Planet. Sci. Lett.*, 43, 13-21. [5] Stolper E. (1982) *Geochim. Cosmochim. Acta*, 46, 2159-2180. [6] Nagahara et al. (1988) *Nature*, 331, 516-518. [7] Mysen B.O. et al. (1985) *Earth Planet. Sci. Lett.*, 75, 139-146.