ESRF	**In situ constrained sintering of ceramic powders to improve understanding of anisotropy formation**	**Experiment number**: MA2587
Beamline: ID16B-NA	**Date of experiment**: from: 25 June 2015 to: 30 June 2015	**Date of report**: 29.02.2016
Shifts: 12	**Local contact(s)**: Julie Villanova	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

* MARTIN Christophe: SIMaP Laboratory, Univ. Grenoble Alpes

* BERNARD Dominique: ICMCB laboratory, Univ. Bordeaux

* LHUISSIER Pierre: SIMaP Laboratory, Univ. Grenoble Alpes

* VILLANOVA Julie: ESRF

GUILLON Olivier : Forschungszentrum Jülich GmbH

Report:

The research aimed at understanding microstructural evolution during the **sintering of ceramic powders**. In particular, we focused on the effect of a geometrical constraint on the microstructure. When a powder is constrained by a rigid substrate, it develops anisotropy, which is generally considered as a defect.

In-situ and ex-situ experiments of sintering of powders were carried out at high temperature (700 to 1300°C). The challenge was to obtain good quality images at such high temperatures and with scan times that are short enough to ensure that the microstructure does not evolve too much within a scan. Fig. 1 shows the experimental set-up, with the high temperature furnace being the original and most sensitive part.

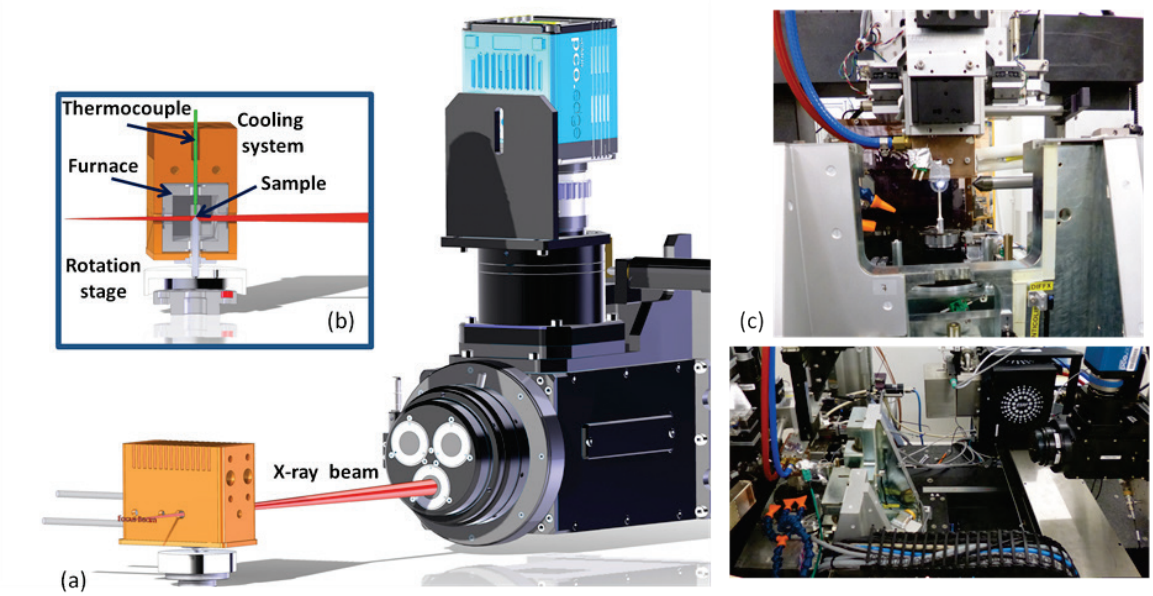

Fig. 1: (a) Schematic representation of the in-situ nano-tomographic set-up.(b) Cross-section of the furnace. (c) Photographies of the set-up. The Sample is mounted on an alumina or mullite sample holder and placed on the rotation stage at a fixed distance from the focal plane of the beam which acts as a secondary source. The furnace surrounded by a cooling system is motorized on the vertical axis. Images are recorded on a PCO edge camera coupled with an objective 10 x magnifications.

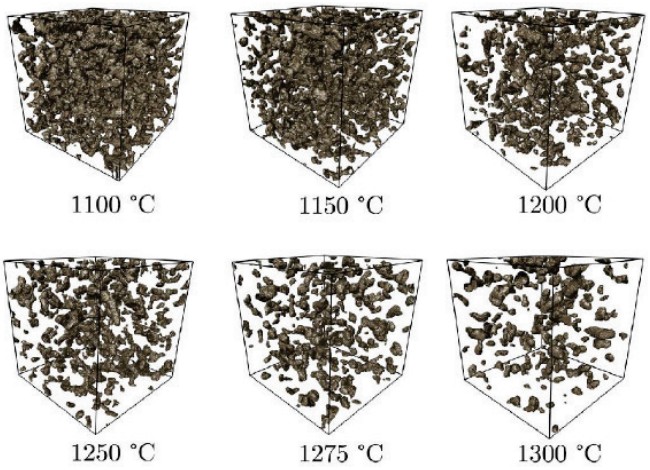

Fig. 2: Porosity evolution with increasing temperature in a sub-volume (15 µm × 15 µm × 15 µm) of a TiO2 microsphere.

Fig. 3: Slice of (Mn1.0)(Co0.9,Fe0.1)O4 material at 850°C.

A first set of in-situ experiments was concerned with the sintering of SiC/TiO2 powders as microspheres in between 1100°C and 1300°C. Fig. 2 shows the type of results that are still being analysed [Lesseur, 2015]. The in-situ sintering experience by nanotomography X has confirmed the sudden changes in the microstructure of a microsphere TiO2 in the initial calcination stages (1100 - 1300 ° C). The porosity decreased very rapidly between 1100 and 1200 ° C, while between 1200 and 1300 ° C, it decreased only by 1.6%. Material transport mechanisms are faster in the early stages of sintering because of the large number of grain boundaries. In advanced stages, the number of grain boundaries decreases and it becomes more difficult to eliminate pores. A second set of experiments was concerned with the sintering of (Mn1.0)(Co0.9,Fe0.1)O4. The samples were obtained by simply breaking some material as tips from a layered material. Two morphologic modifications were investigated. The first is a sintering of the cracks that can appear between 400 and 500°C. The second is the slow sintering at 850°C. Both phenomena are still under analysis.

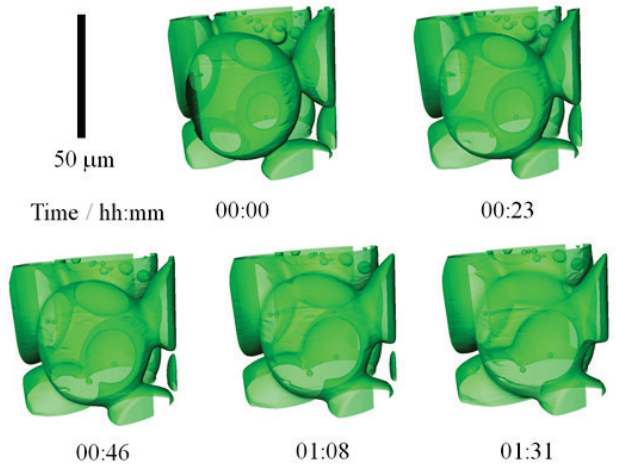

Fig. 4: 3D volumes of glass beads during sintering at 670°C. Glass particles of 50 µm diameter are enclosed in a quartz capillary.

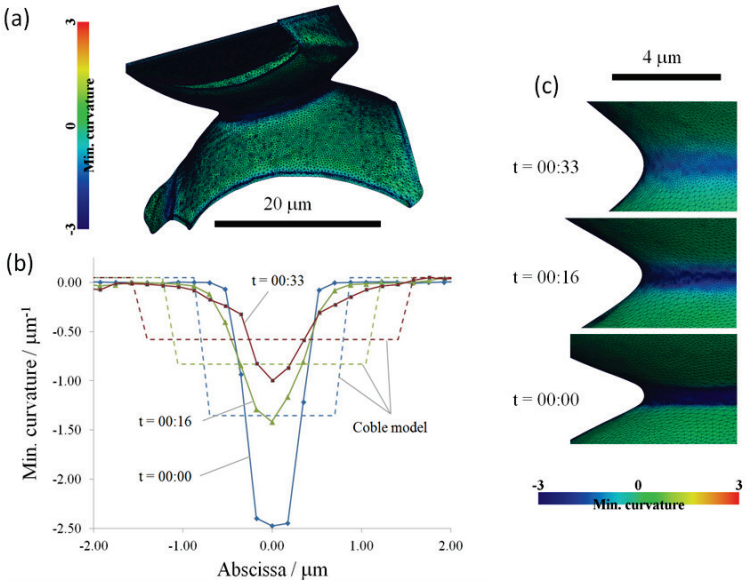

Fig. 5: (a) neck with computed local curvature. (b) Comparison between experimental minimum curvature and coble model as a function of the distance from the neck. (c) Evolution of the neck geometry and curvature with time.

A third set of experiments relates to the observation and quantification of the 3D microstructure evolution during the isothermal sintering of soda-lime glass particles at 670°C. The particle diameter was around 50 µm, which allowed for a precise observation of contact evolution between glass beads (Fig. 4 and 5). Data

on the neck curvatures were extracted. The procedure to assess local curvatures involves the computation of an isocontour on the grayscale reconstruction followed by an adaptative remeshing. The experimental local curvature is compared to the Coble geometrical model of the neck in figure 5. This model suggests that the neck geometry can be approximated by an inverted tore tangent to the spherical particles and obeying volume conservation. The 3D experimental observation confirms that the ideal neck geometry proposed by Coble, even though being useful to construct densification models, is only a rough representation of reality.

The three experiments described in this report demonstrate the feasibility of high temperature in-situ X-ray nano-tomography. In both cases, the 3D observation at high resolution of the very early stages of the investigated phenomena gives relevant information to confirm or develop realistic simulation of these physical processes. In conclusion, in-situ X-ray nano-tomography has been successfully performed during thermal treatment between 500°C and 1300°C on different materials with a pixel size as low as 100nm.