	Experiment title: In-situ hydration of hydraulic binders using fast diffraction-tomography	**Experiment number**: MA4498
Beamline: ID11	**Date of experiment**: from: 09/10/2020 to: 14/10/2020	**Date of report**: 11/09/2023
Shifts: 15	**Local contact(s)**: Carlotta Giacobbe	*Received at ESRF:*

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Report:

Summary:
The experiment was divided into two main parts. One of them was devoted to investigating the hydration process of gypsum plaster *in situ* with a combination of diffraction-tomography techniques (scanning 3DXRD and phase contrast tomography). In the other one, we acquired 3DPDF scans of hydrated alite (C_3S) to gain insights into the crystallographic structure of C-S-H which is the main binding phase of cement. The full experiment was done in 9 shifts. The first 6 shifts were used to align the beamline and to perform the *in situ* hydration of gypsum plaster in the 3DXRD station of EH3 (one of the experimental hutches of ID11). The last 3 shifts were used to collect the 3DPDF data on two samples of hydrated C_3S in the Nanoscope station of EH3.

In situ hydration of gypsum plaster:
The experiment was conducted alternating the setups for scanning 3DXRD and phase contrast tomography (PCT). Two samples were measured. The goal was to obtain both spatial and crystallographic information on the phase transformation from bassanite ($CaSO_4$ x 0.5 H_2O) to gypsum ($CaSO_4$ x 2 H_2O) and on the microstructure of the hydrated gypsum plaster. We acquired a total of 2 s-3DXRD measurements per sample, one before starting the hydration (bassanite) and one at the end of the reaction (gypsum), and several PCT scans while monitoring the hydration process (up to 36 h of hydration). The samples were prepared by filling quartz capillaries with 300 µm of diameter with bassanite grains. The hydration was performed *in situ* with a solution saturated with $CaSO_4$. The energy used was 44 KeV.

- **Scanning 3DXRD setup**

For the scanning 3DXRD setup the beam was focused to a size of 5 µm (see Figure 1). In both the measurements that have been taken on the two samples, a layer of the capillary was scanned horizontally (x-direction) with steps of 5 µm. For each Δx the sample was rotated of 180° with angular steps of 1°. The diffraction spots coming from the sample were collected with the FreLoN2k camera placed at 154 mm from the samples.

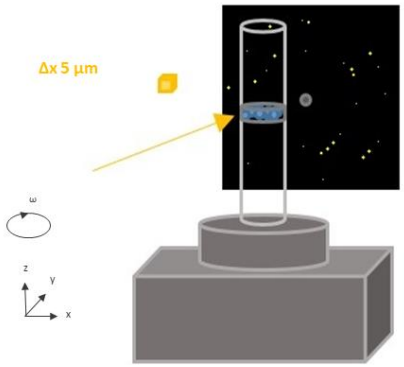

Figure 1. Experimental setup used to acquire the scanning 3DXRD data of the two samples measured. Layers of the capillary were scanned with a pencil beam of 5x5 µm size. The diffraction spots were collected on the FreLoN2k camera.

The data analysis was conducted using Jupiter notebooks based on the ImageD11 software developed in-house at ID11. At first, all the diffraction peaks were extracted from the 2D frames of the detector (see Figure 2a). Successively, the sinograms of the peaks and the correspondent iradon transformed intensity maps were reconstructed.

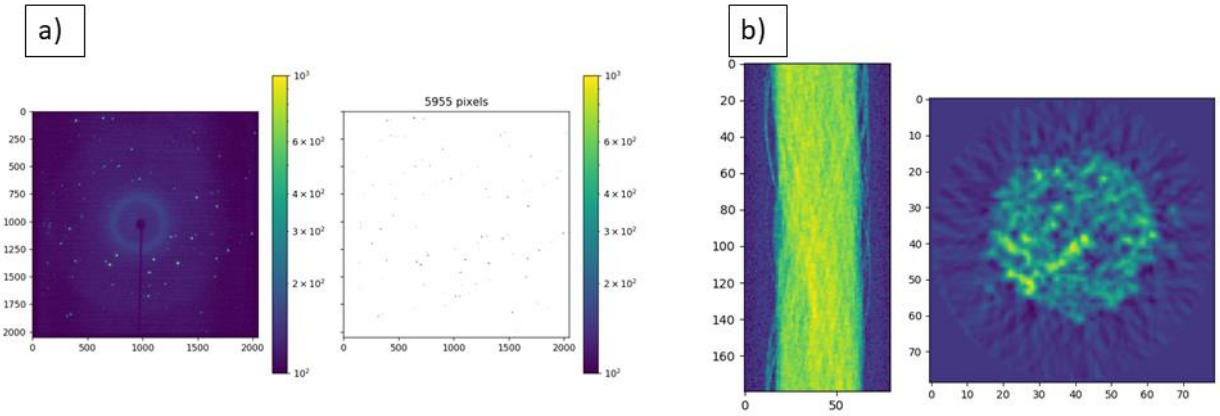

Figure 2. a) Extraction of the diffraction peaks from the 2D frames of the detector. b) Reconstruction of the sinograms of the peaks and relative iradon intensity map.
The diffraction peaks of all the scanning 3DXRD measurements were then indexed to retrieve the phase, cell parameters and crystallographic orientation of the crystals.

Phase contrast tomography setup
For the PCT measurements, the setup presented few differences compared to the previous one. Here, no focusing device was used and the projections were collected with a tomography detector (FreLoN4M) (see Figure 3). The detector was placed at 300 mm from the sample to enhance the contrast between the different phases of the sample. We used a 10x magnification lens, giving a pixel size of 1.56 µm. A total of 500 projections were recorded per each scan during a rotation of 360°. The length of each scan was 10 min and they were recorded each 30 min up to the end of the hydration reaction.

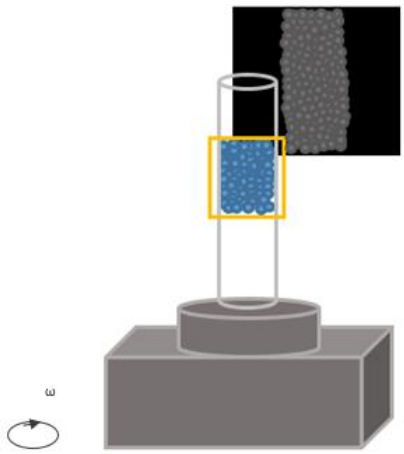

Figure 3. Experimental setup used to acquire the PCT data *in situ*. The full beam was illuminating a portion of the capillary of 1 mm height.

Transversal slices of the samples were reconstructed using the in-house software nabu and tomwer canvas applying the Paganin algorithm to retrieve the phases (see Figure 4 for the full data reconstruction strategy).

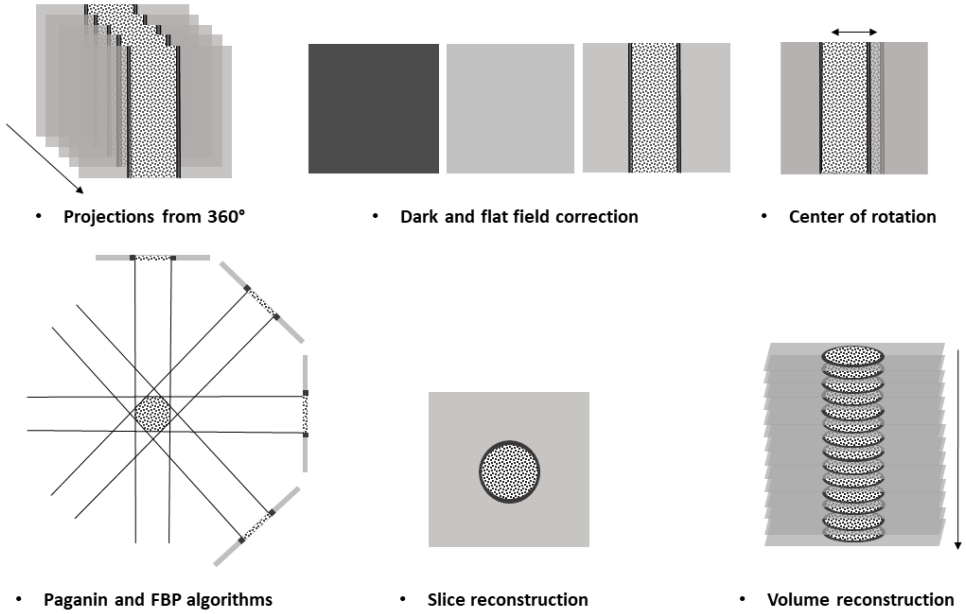

Figure 4. Complete workflow used to reconstruct the transversal slices (volumes) of the PCT scans. This task was performed using nabu and tomwer canvas.

Successively the volumes were treated with ImageJ and Dragonfly to visualize the morphology of crystals and evolution of microstructure (see Figure 5).

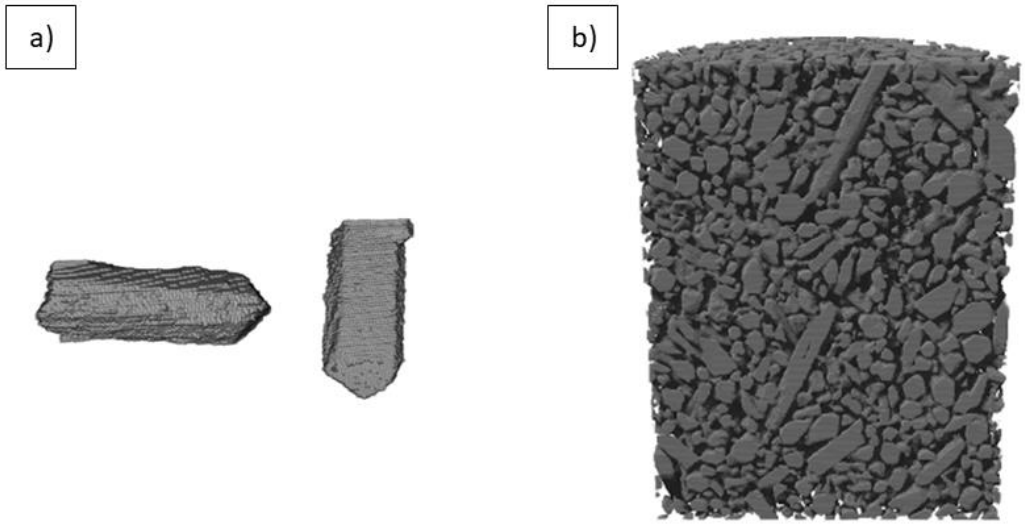

Figure 5. a) 3D volumes of two bassanite grains before the hydration. b) 3D volume of the full capillary scanned before the hydration.

All the results and the full detailed description of this combined scanning 3DXRD and PCT experiment can be found in [1].

1. **Characterization of C-S-H crystal structure in hydrated C_3S:**

This experiment aimed to characterize as much as possible the nanometric structure of C-S-H precipitated in hydrated C_3S samples. Even though it has been demonstrated that the nucleation of C-S-H occurs through a multi-step process, the mechanism of growing of C-S-H on the surface of C_3S particles is still an open question [2]. The use of 3DPDF offered the possibility to couple the information about the long-range order of the structure of C-S-H with its spatial distribution on the surface of C_3S. Two samples were measured. One in which the hydration reaction was stopped after 10 min and one that was hydrated for up to 24 h. The samples were prepared by gluing the hydrated C_3S powder on Kapton loops (see Figure 6).

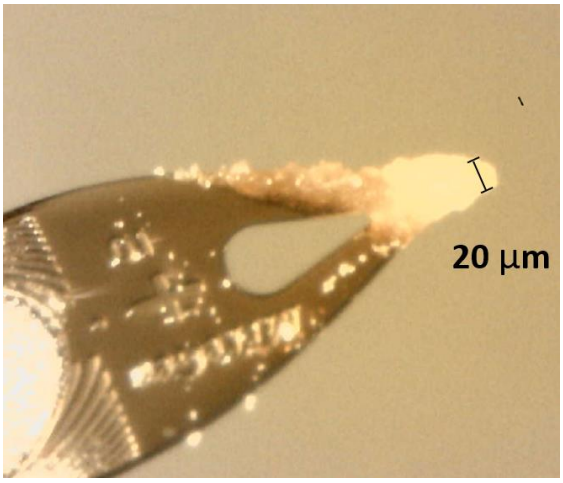

Figure 6. Picture of one of the samples measured with 3DPDF.

The energy used was 75KeV. The beam was focused to a size of 500 nm using Silicon Compound Refractive Lenses.

References

[1] La Bella M. et al., *J. Appl. Cryst.*, **56**, 660-672 (2023).
[2] Krautwurst N. et al., *Chem. Mater.*, **30**, 2895-2904 (2018).