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| ESRF | **Experiment title: Characterisation by X-ray imaging of the solidification of multi-crystalline silicon for photovoltaic applications.** | **Experiment number**: MA-1138 |
| **Beamline**: BM05 | **Date of experiment**: from: 02/02/2011 to: 08/02/2011 | **Date of report**: 01/03/2012 |
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## Scientific Objectives:

The general scientific objectives of our experiments are to provide benchmark data concerning the solidification of multi-crystalline Si (mc-Si) for photovoltaic (PV) applications. These original experiments consist in synchrotron X-ray imaging characterisation of the mc-Si during its solidification.

These experiments and proposal are part of a larger project entitled Si-X (Characterisation and understanding of the crystallisation of photovoltaic **Si**licon: **X**-ray synchrotron imaging) which is funded by the HABISOL (Habitat Intelligent et Solaire Photovoltaïque) program of the French National funding agency (ANR). ESRF is a full partner of this project.

MA-1138 campaign was the first one devoted to this project. Its primary objective was to perform X-ray radiography of the solid-liquid interface during the solidification of multicrystalline silicon.

## Experimental method:

The experiments were carried out on the beamline BM05. The samples are prepared at the IM2NP laboratory by cutting and mechanical polishing and have a surface exposed to X-rays of *40x 6 mm*$^2$ with a thickness of about *300µm*. Crucibles made of pyrolitic boron nitride are used and the sample/crucible set is introduced in a bridgmann furnace. This furnace specifically designed by IM2NP for these experiments consists of two graphite heaters and three pyrometers (two pointing on the heaters and one on the sample edge) and allows X-ray imaging. Our solidification experiments are achieved by decreasing the temperature of the two heaters and keeping the sample/crucible set motionless.

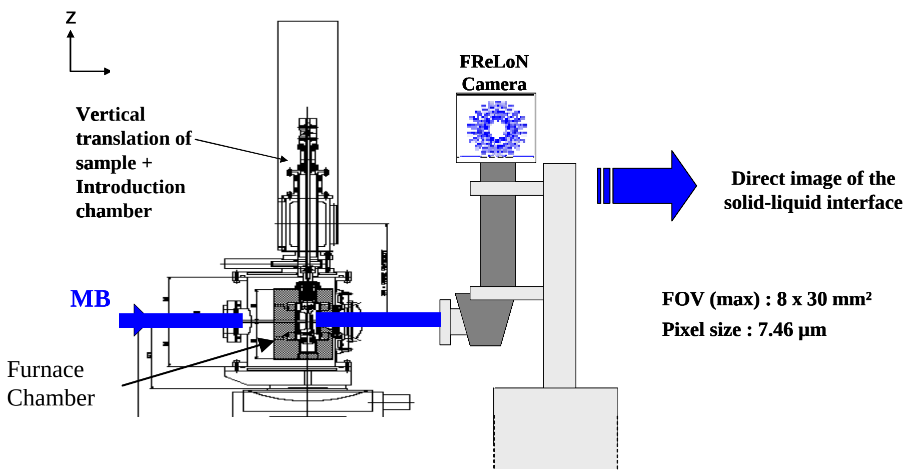


**Fig. 1**: Schematic drawing of the X-ray radiography set-up using monochromatic beam (MB).

In the reported experiments, the X-ray radiography characterization was achieved by illuminating the solidifying sample with a monochromatic beam (Energy = 13.5keV) and by collecting on a CCD camera the transmitted beam (Figure 1). Using this method, we can observe *in situ* and in real time the solidification of the sample and the evolution of the solid/liquid interface. Further analysis can give information on the interface velocity and its morphology.

## Preliminary results:

The X-ray radiography images obtained during our experiments are based on the absorption contrast. The absorption coefficients of the solid and liquid silicon depend on the mass density and are not much different (*2.31g/cm3* for the solid and *2.56g/cm$^3$* for the liquid) which means that the solid/liquid interface on the raw images is hardly distinguishable.

As a consequence, two image processings which are bringing complementary information are used to reveal the solid-liquid interface. One consists in dividing the current image by the first image before solidification is triggered (Figure 2). This image processing shows a grey zone which is the liquid not yet solidified and a white zone which is the solidified part of the sample. The solid appears in white because it is less dense and thus less absorbing than the liquid phase. An interesting feature is that we observe a light grey part between the two zones (Figure 2.b). This zone reveals the solid protruding in the liquid and indicates that the interface is not flat.

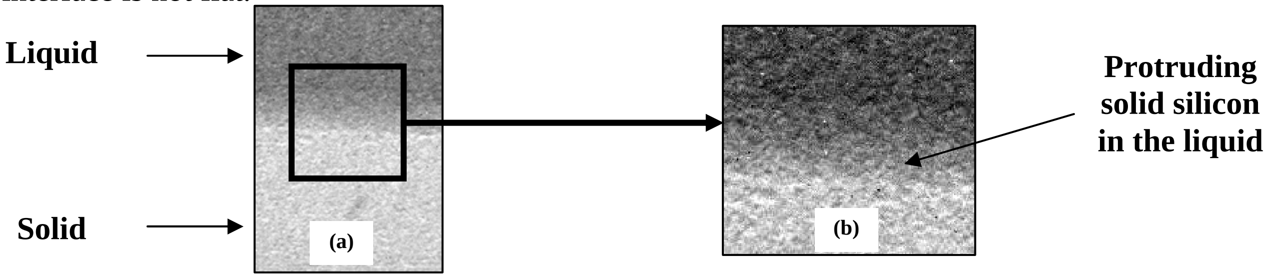


**Fig. 2.** (a) Results of the division of a raw image obtained during solidification by the first image before solidification in a Czochralski silicon sample under a temperature gradient G = 10 K/cm & a cooling rate R = 0.5 K/min, (b) zoom showing the protruding solid in the liquid phase.

The other image processing consists in dividing successive images i.e. dividing the current image by the previous image. Then, we observe the shape of the interface more accurately (rough or facetted for instance) and, reveal the evolution of the interface between two images (Figure 3).

Figure 3 shows subsequent images obtained during the solidification of an Upgraded metallurgical (UMG) -Si sample after prior complete melting at a temperature gradient of 16K/cm and for a cooling rate of 0.4K/min. In this rather low growth rate experiment, we were able to observe the formation of facets and their time evolution at the side of the silicon droplet. Moreover, other facets were observed at the interface during the solidification and growth rates were measured.

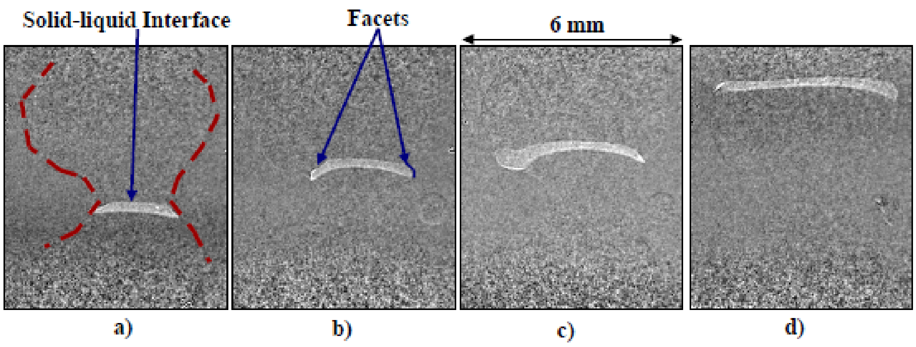


**Fig. 3**: Subsequent images of UMG Si solidification at G=16K/cm and R=0.4K/min. Division of the current image by the previous one to reveal the solid-liquid interface. a) $t_0$, b) $t_0$+11min, c) $t_0$+12min, d) $t_0$+21min.

## Future work

This first campaign allowed us to validate the experimental devices: furnace and X-ray imaging environment. The next experiments will consist in more scientific objectives (identified phenomena like twins and grain competition, different silicon grades..) associating both X-ray radiography and topography methods.