ESRF	**Experiment title:** **"In situ" and real time study of metallic alloy solidification by synchrotron X-Ray imaging**	**Experiment number**: ME-691
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Report:

It is well known that the properties of grown materials are strongly determined by the solidification step, so that a precise mastering of growth processing is essential to reproducibly tailor products of specified quality. In particular, during the growth segregation of chemical species may be induced by defects (grain boundaries, dislocations, ..), grains structure or the solidification microstructure itself, which could appear at the solid-liquid interface due to the Mullins-Sekerka instability [1]. A complete understanding of growth processing, from the microscopic to the macroscopic scale is a key point both from technological and fundamental science points of view.

Our objective is to achieve in situ and real time investigations of the solidification of metallic alloys using most advanced X-ray imaging techniques (radiography and diffraction topography) with the increased capabilities of 3rd generation synchrotrons. More precisely, the project addresses two different issues:

- Columnar and equiaxed growth of refined and non-refined binary alloys (Al –3.5 wt% Ni) [1, 2]
- Analysis of growth mechanism in quasicrystal (Al-Pd-Mn system) [3]

[1] Mullins, W.W. & Sekerka R.F., J. Appl. Phys. 35 (1964) 444.
[2] Martorano, M.A., Beckermann, C. & Gandin, Ch.-A., Metall. and Mater. Trans. A, 34A (2003) 1657.
[3] Janot, C. & Dubois, J-M., *Les Quasicristaux : Matière à paradoxes* (EDP Sciences, Les Ulis, France, 1998) Ch. 3.

For both issues, directional solidification is a key technique which allows one to analyze independently the effect of solute concentration, temperature gradient and pulling rate on the solid-liquid interface. For this project, an experimental set-up has been developed in collaboration with ID19 beamline team members. It is composed of an Ultra-High-Vacuum furnace for "in situ" and real time observations of solidification by X-

ray imaging. Its specificity lies in that it allows both to control the two main parameters of unidirectional solidification (pulling velocity and temperature gradient) and to record X-ray radiographs and X-ray topographs, either independently or simultaneously. During previous allocated beam time (6 months project), we performed a series of tests and the preliminary results showed that **[4]** it is necessary to use a furnace ensuring a vertical growth direction instead of a horizontal one, in order to avoid disturbances of the solidification process induced by natural convection. During these preliminary investigations, X-ray imaging techniques have also been tested in close relation to the scientific objectives.

Therefore, during the 1st part of the 1st year of the project, in collaboration with the ID19 engineering staff (Pascal Bernard and René Chagnon), we have mainly performed the experimental development of the UHV-furnace and X-Ray imaging techniques (absorption/phase contrast radiography and topography). The vertical axis furnace was successfully tested.

Then, in the 2nd part of the 1st year of the project, several outstanding observations were obtained, often for the first time. These results were presented in international conferences:

- Columnar dendritic growth:
  - Initial transient **[5,7,9]**
  - Bending of secondary arms during dendritic growth **[7]**
- Columnar to equiaxed transition and equiaxed growth:
  - Dynamical arrangement of equiaxed dendrites in the equiaxed regime **[5, 6]**
  - Grain structure maps and columnar to equiaxed transition **[6]**
- Quasicrystal growth:
  - Facetted growth **[5,8,9]**
  - Nucleation and free growth of grains above the interface at high growth rate **[5,8,9]**.

These appealing results opened new fields of investigations with X-ray techniques applied to these phenomena.

Articles and abstract are listed below.

[4] *H. Nguyen Thi.,H. Jamgotchian, J. Gastaldi, J. Härtwig,T. Schenk, H. Klein, B. Billia, J. Baruchel, Y. Dabo*
Preliminary in situ and real time study of directional solidification of metallic alloys by X-ray imaging techniques.
Journal of Physics D: Applied Physics, vol. 36, p.83-86, 2003
Abstract: *During directional solidification of a binary alloy, the solid–liquid interface exhibits a variety of patterns that are due to the Mullins-Sekerka instability and governed by the growth conditions. It is well known that properties of the grown material are largely controlled by the microstructures left in the solid during processing. Thus, a precise mastering of the solidification is essential to tailor products in a reproducible fashion to a specified quality. One major difficulty for this study is the real time and in situ observation of the interface, especially for metallic alloys. A possibility is to use an intense and coherent 3rd generation X-Ray beam. By combining different X-ray imaging techniques (absorption/phase contrast radiography and diffraction topography), we have studied the directional melting and solidification of aluminium-based alloys. The preliminary results show the great potential of these techniques for the study of the coupling between stress effects and microstructure formation in solidification processing.*

[5] *T. Schenk, H. Nguyen Thi, J. Gastaldi, G. Reinhart, V. Cristiglio, N. Mangelinck-Noël, H. Klein, J. Härtwig, B. Grushko, B. Billia, J. Baruchel*
Application of Synchrotron X-ray imaging to the Study of Directional Solidification of Aluminium – based Alloys
Journal of Crystal Growth, in press
Abstract: *3rd Generation Synchrotron X-ray sources like ESRF (European Synchrotron Radiation Facility) have enhanced the capabilities of in situ and real-time X-ray imaging so that it is possible to follow the solid – liquid interface dynamics on metallic alloys with improved spatial and time*

resolution. In this paper, we focus on the solidification patterns of Al-based alloys, visualised by absorption and phase contrast radiography at ESRF. Two metallic systems were selected for this study: - i) Al – 3.5 wt% Ni (with and without refiners) for the analysis of columnar growth and equiaxed growth, and the influence of the refiners on the columnar to equiaxed transition (CET), which is a key point in solidification processing, - ii) $Al_{72.4}Pd_{20.5}Mn_{7.1}$ for the analysis of interface shape of growing quasicrystals (QCs). For both alloys, striking features were observed and a preliminary discussion is presented. Those results demonstrate the high potential of ESRF Synchrotron source for the characterisation of the dynamical formation of the solid microstructure in materials processing.

[6] *N. Mangelinck-Noël, H. Nguyen-Thi, G. Reinhart, T. Schenk, V. Cristiglio, M.-D. Dupouy, J. Gastaldi, B. Billia, J. Härtwig, J. Baruchel*
In situ analysis of equiaxed growth of aluminium-nickel alloys by x-ray radiography at ESRF
Journal of Physics D: Applied Physics, accepted
Abstract: *We present results obtained, at the ID19 beamline of the ESRF, by Synchrotron X-Ray Radiography during the solidification of Al-Ni alloys. We focus on the dendrite regime and more particularly on columnar dendritic solidification and equiaxed solidification, and the transition between both regimes. The columnar to equiaxed transition is a critical and still pending issue in metallurgy. Making use of the high potential of synchrotron experimental tools for in situ and real-time characterisation of the solid-liquid interface during directional solidification, we were able to provide insight on key physical phenomena, in particular: sedimentation, interaction and arrangement of equiaxed grains.*

[7] *G. Reinhart, H. Nguyen-Thi, J. Gastaldi, B. Billia, N. Mangelinck-Noël, T. Schenk, J. Härtwig, J. Baruchel.*
In situ and real time investigation of directional solidification of Al - Ni alloys by synchrotron imaging
Material Science Forum, accepted
Abstract. *Solidification is a dynamic phenomena and, as a consequence, it is of major interest to be able to investigate this process by in situ and real time observation. With synchrotron sources, this can be achieved by applying X-ray Imaging techniques (Radiography and Topography). Hence it is possible to follow the dynamical selection of solidification pattern on metallic alloys and to observe strain effects during growth process. In this paper, we present results obtained by using separately the two imaging techniques for the study of the microstructure formation during Al – Ni alloys solidification.*

[8] *T. Schenk, G. Reinhart, V. Cristiglio, G. Gastaldi, H. Klein, B. Grushko, H. Nguyen-Thi, N. Mangelinck-Noël, J. Härtwig, B. Billia and J. Baruchel*
Live Observation of the Growth of Quasicrystal Grains
ESRF Spotlight, 04/10/2004

[9] *T. Schenk, H. Nguyen-Thi, J. Gastaldi, G. Reinhart, V. Cristiglio, N. Mangelinck-Noël, H. Klein, J. Härtwig, B. Grushko, B. Billia, J. Baruchel*
Application of Synchrotron X-ray Imaging to the Study of Directional Solidification of Aluminium-based Alloys
ESRF Highlights, accepted