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Imaging of Aluminium Alloy Solidification by Synchrotron X Radiation

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Abstract

High intensity synchrotron X-radiation has been used for video microscopic studies of solidification in Al-Cu alloys. A fast readout low noise detector has allowed imaging with a high spatial and temporal resolution of growing crystals during controlled solidification in a Bridgman furnace. Columnar and equiaxed aluminium dendrites and Al-Cu eutectic growth has been studied systematically as well as formation of defects such as hot tears and gas pores. Dedicated image processing has permitted the extraction of quantitative information on crystal morphology, local growth rate and liquid concentration variations. The results are expected to be valuable for a better understanding of aluminium crystal nucleation and growth and for the validation of crystal growth models. This paper will focus on description of the new technique and give examples of imaging of aluminium dendritic growth, dendrite fragmentation, Al-Cu eutectic growth and porosity formation.

1. Introduction

Imaging of solidification is important in order to understand how the microstructure develops as the solidification proceeds. A metallographic investigation of the solidified material shows a microstructure that has transformed and coarsened during the solidification and subsequent cooling, and it is often difficult to conclude how the growing crystals have evolved during the early stages of solidification.

Quenching of the microstructure during solidification results in a substantial refinement and this method has been used in order to “freeze” the microstructure. Subsequent metallographic investigation shows a coarse structure which is assumed to be representative of the solid at the moment of quenching and a fine microstructure that is assumed to represent the melt. Transient phenomena subsequent to the quenching, however, results in a gradual transformation from the coarse to the fine microstructure and this often leads to errors like an overestimation of the fraction solid [1].

Another method to get an impression of the microstructure during solidification is decanting or drainage of the melt during solidification and subsequent investigation of the solidified crystal network. Drainage sometimes happens naturally in castings due to insufficient feeding but can also be promoted. This method is, however limited to relatively coarse, coherent structures.

Transparent organic materials have successfully been used to simulate the solidification of metals [2]. These organic materials have low entropies of fusion and therefore solidify in a non-facetted way like metals. Since they are transparent, growing crystals can be studied under the microscope due to phase contrast at the solid-liquid interface. These so-called transparent analogs have been used to study the morphology of cellular, dendritic and eutectic solidification. Even though the information from such studies has been useful to verify theories on cellular and dendritic morphology, the information that can be achieved is limited. Firstly, only a limited number of organic substances can be used. These materials are, of course, not representative of all various metal alloy systems with their different constitutions, and crystal structures. Secondly, the phase contrast image is limited to the solid – liquid interface

Metals are transparent to X-rays, and provided that a sufficiently powerful X-ray source and a high-resolution fast detector can be employed, dynamic, in-situ imaging of alloy solidification can be made. If the X-rays are coherent, the images will show both phase contrast and absorption contrast, and they will show information of alloy distribution in the solid and liquid provided that the atomic absorption coefficient between the base metal and the alloying element is sufficiently different. At synchrotrons, sufficiently powerful and coherent X-ray sources can be found to allow for simultaneous absorption- and phase contrast imaging to be carried out on at least thin sections of melts. Absorption contrast radiography [3], and also synchrotron radiation topography [4,5] has, in fact, been used to image solidification previously, but only recently, experiments have been carried out that has taken full advantage of simultaneous spatial and time resolution [6,7]. These experiments were undertaken in order to explore the possibility of using X-rays for in-situ dynamic high resolution imaging of alloy solidification and to use the method to study the development of dendritic and eutectic microstructures as well as defects such as pores and hot tears and macrosegregation. The present paper reviews some of these results obtained from Al-Cu alloys.

2. Experimental

Al-Cu alloys with Cu-concentrations ranging between 10 and 30 wt% were produced from melting high purity aluminium in a graphite crucible and chill casting. Thin slices 25x10x0.2 mm were cut from the castings, ground parallel and placed in an envelope made from 0.1 mm thick quartz plates. Prior to inserting the samples they were pre-oxidized at 720K for 2 hours and coated with a thin layer of boron nitride to prevent subsequent reactions between the melt and the quartz. The sample container was then mounted in a Bridgman furnace system that provided control over experimental solidification conditions like sample pulling velocity and temperature gradients, Sample containers were also made with internal thermocouples that were used to calibrate the Bridgman apparatus and sample temperatures, and also to optimize the thermal contact between the sample and the furnaces. The Bridgman rig was mounted on an (xyz) micro-translation stage that allowed for accurate alignment and sample positioning flexibility in the X-ray beam.

The X-ray imaging experiments were performed at the ID22 undulator beamline at the European Synchrotron Radiation Facility (ESRF).

Monochromatic incident beam energies between 12-15 KeV were employed. Slits were used to produce a (1.35 x 1.35) mm^2 quadratic incident beam cross section on the sample.

A Fast Read-out Low-Noise (FReLoN) high resolution CCD detector system [7] with 4.2 MPixel array and 14 bits dynamical range was used.

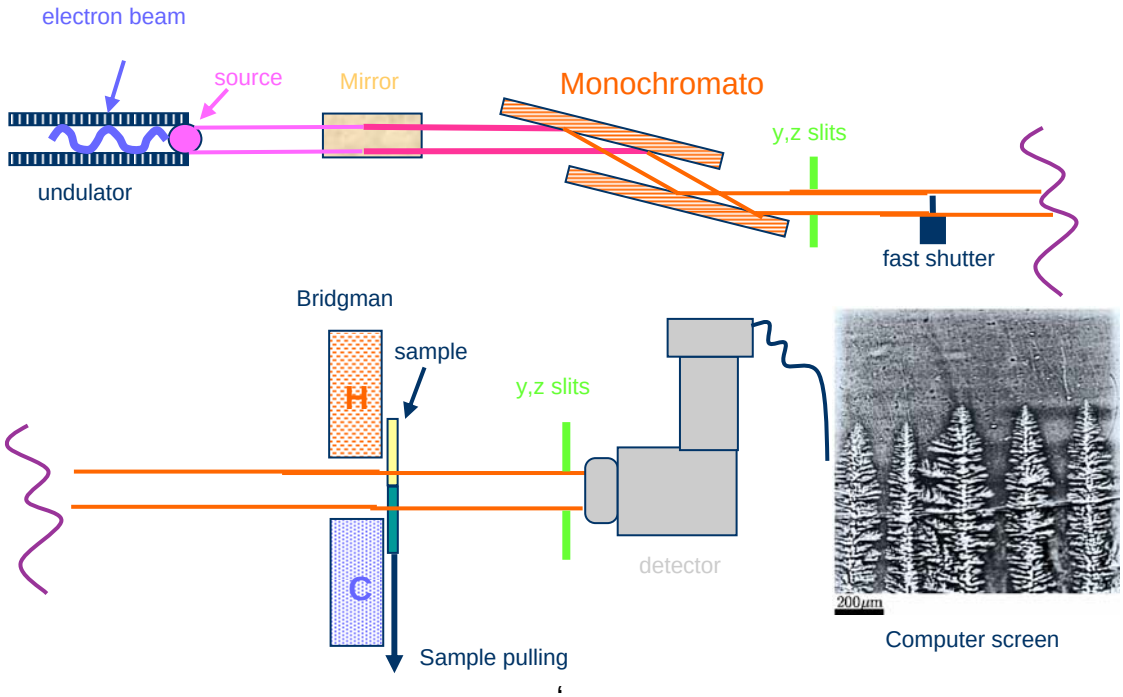

Figure 1: Experimental arrangement for synchrotron X-ray imaging of alloy solidification.

The detector system offers different read-out modes, but the one employed for the results presented here gave nominal spatiotemporal resolutions of 2.0 µm/100 ms, all system aberrations included. Figure 1 shows, schematically the experimental set-up for X-ray imaging experiments.

3. Results and Discussion

Figure 2a shows columnar solidification of an Al-30wt%Cu alloy. The temperature gradient is 27K/mm which means that growth of both Al dendrites and Al-Cu eutectic can be seen in the same field of view. Figure 2b shows the same image where the contrast has been increased to reveal the concentration variations in the liquid boundary layer. Figure 2c shows a contour of the solid-liquid boundary that has been extracted from the image. Figure 2 demonstrates how quantitative information can be extracted from the images by advanced image processing techniques.

Figure 3 shows a sequence of images in the same alloy as Figure 2, Al-30wt%Cu. The figure illustrates the ranges of microstructures that can be obtained by variations in the growth parameters.

At moderate temperature gradients, a moderate growth rate gives columnar dendritic growth, as seen in Figure 2, while an increase in growth rate results in equiaxed growth, Figure 3a. At higher temperature gradients and low growth rate, 7µm/s, dendritic growth is suppressed, Figure 3b; the limiting growth rate for dendrites, given by the constitutional undercooling criterion for this alloy is 10 µm/s. An increase in growth rate at this gradient results in eutectic cellular growth, Figure 3c.

Figure 4 shows hydrogen porosity formation in an Al-30wt%Cu alloy. It can be seen that two pores are formed close to the eutectic front and are incorporated into the solid material. In most cases, porosity in this alloy formed on the eutectic front, but occasionally porosity was also found to form in the eutectic region, closer to the dendrite tips. Note that the solidification direction is as shown in the images. Growth upwards, as seen in Figures 2 and 3 normally resulted in the immediate escape of hydrogen bubbles due to buoyancy effects whereas growth in the direction of the gravity vector resulted in gas entrapment as porosity in the microstructure. Hydrogen porosity was observed to occur quite frequently in the samples despite hydrogen removal by inert gas purging of the melts prior to casting of the samples. It appears that some hydrogen pick-up might have occurred on remelting during the imaging experiments.

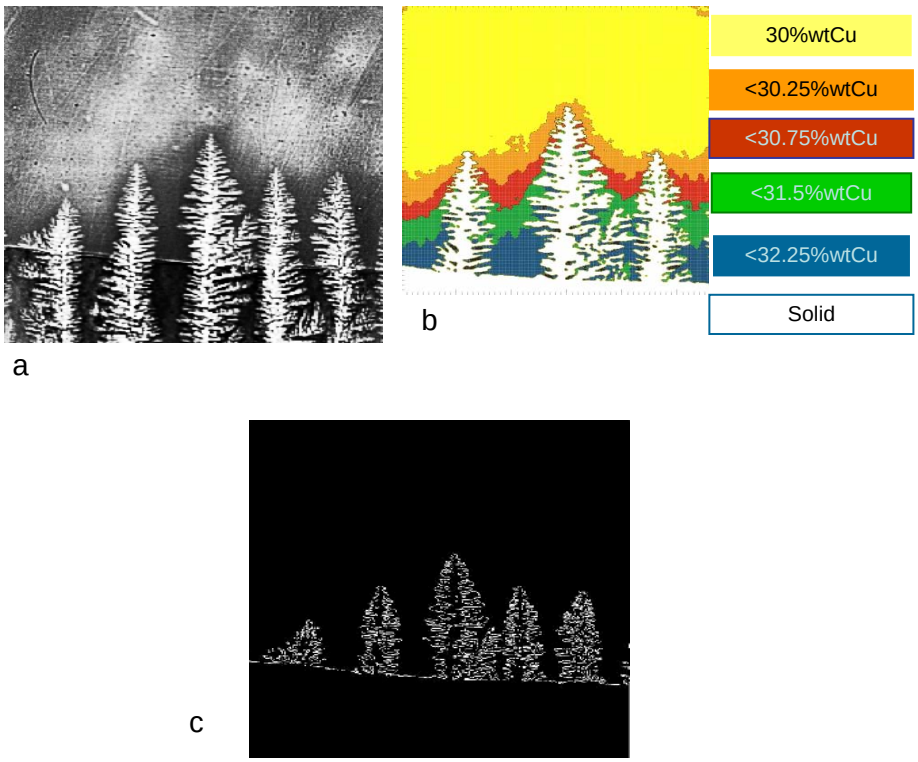

Figure 2: Images of an Al-30wt%Cu alloy grown at 22 µm/s in a temperature gradient of 27K/mm. a) shows the image after cropping, flat field correction and some contrast stretching. b) shows the image after passing through a binary mask to separate areas of different liquid concentrations and c) shows the image after a filtering and thinning operation to reveal the solid-liquid boundary.

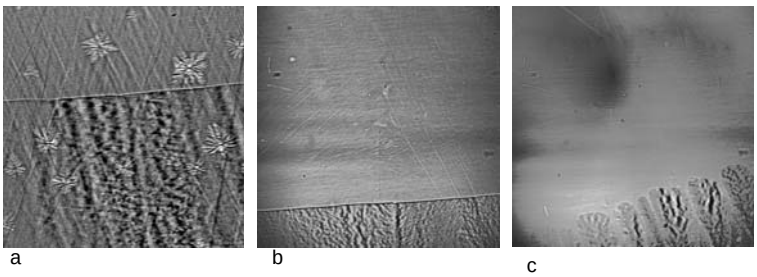

Figure 3: Images of an Al-30wt%Cu alloy showing variations in growth morphology. a) shows equiaxed dendritic growth at a temperature gradient, G, of 30 K/mm and a growth rate, V, of 50 µm/s. b) shows planar eutectic growth at G=46 K/mm and V=7 µm/s. c) shows cellular eutectic growth at G=46 K/mm and V=32 µm/s.

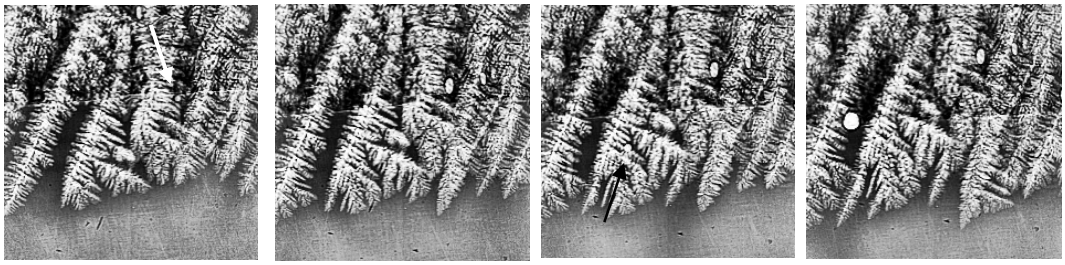

Figure 4: Porosity formation in an Al-30wt%Cu alloy grown at a temperature gradient of 27K/mm and a growth rate of 22 µm/s. a) shows a bubble forming on the eutectic front and becomes incorporated into the solid, b) as a pore. c) shows a bubble forming in the dendrite network and forms a pore in the solid, c). The images are taken at 2 s intervals.

An advantage with the present video-microscopy method is that the dynamics of solidification can be studied in detail. One feature that has been very obvious is the high degree of mobility that free, equiaxed crystals show, even in the confined space of the quartz glass container. Figure 5 shows the solidification of an Al-15wt%Cu alloy. Since the crystals contain a lower copper concentration than the melt, they are lighter and float, but crystal motion also in the horizontal direction can be seen in the sequence. Thermosolutal convection and crystal motion with horizontal components ahead of the solidification front has been observed in transparent analog experiments and has also been modelled [8].

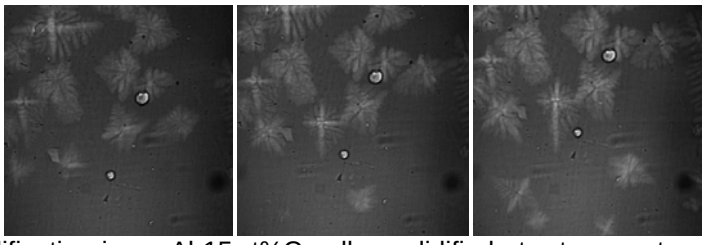

Figure 5: Equiaxed solidification in an Al-15wt%Cu alloy solidified at a temperature gradient of 25K/s and a growth rate of 10 µm/s showing moving crystals in the lower section of the image and a semi-stationary coherent crystal network forming in the upper section. The images are obtained at intervals of 1.5 s starting from the left. The two bright circular features are artefacts.

A phenomenon that was often observed both during columnar and equiaxed dendritic growth was the formation of new crystals by dendrite fragmentation. This often occurred by melting of secondary dendrite arms by the root. Figure 6 shows an example of fragmentation during growth of large equiaxed crystals in an Al-20wt%Cu alloy. At high temperature gradients, the newly formed crystals float into regions of hotter liquid and actually start to melt again, which can also be seen in Figure 6.

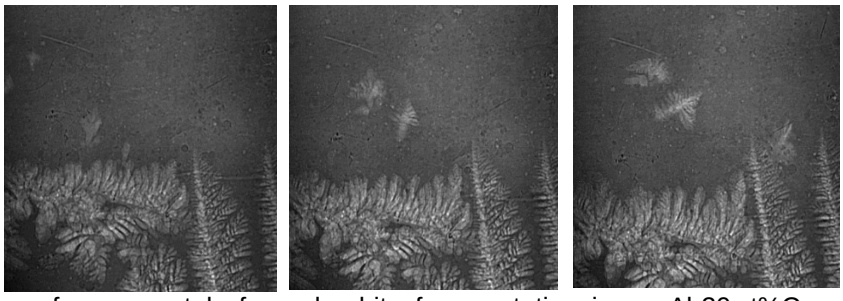

Figure 6: Formation of new crystals from dendrite fragmentation in an Al-20wt%Cu alloy solidified at a temperature gradient of 25K/s and a growth rate of 10 µm/s. The fragments grow into dendrites and float up into a hotter region at the top of the images where they start to melt again. The mages were obtained at intervals of 2 sec.

Conclusions

This review has intended to demonstrate the merits of a recent experimental technique for monitoring solidification processes in metal alloys where spatiotemporal data can be extracted for the phase front with resolutions that are comparable to those available in light microscopy with transparent analogs. Examples have been given from studies of dendritic and eutectic growth as well as dendrite fragmentation and porosity formation in Al-Cu alloys. The next step in developing and evaluating the merits of this method should be to compare the experimental data with results from modelling.

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