Impact of gravity-related phenomena on the grain structure formation: comparative study between horizontal and vertical solidification of a refined Al-20wt.%Cu alloy

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Abstract. On earth, gravity-related phenomena are unavoidable, such as thermo-solutal convection caused by density gradients in the melt and buoyancy when the liquid phase is denser than the solid phase. Such phenomena can drastically affect both the grain density and their morphology during equiaxed solidification processes. For these reasons, fundamental studies comparing the influence of solidification parameters with and without gravity effects are important to obtain benchmark data, which are useful to understand and then control the final structure of materials in industrial processes. In the present work, the impact of the solidification parameters on the dendritic grain structure formation and on the final grain size and shape was investigated in situ by using X-radiography for different growth orientations with respect to gravity. In a first step, experiments were carried out with various solidification parameters and with the furnace in horizontal position, with the main surface of the sample being perpendicular to gravity to limit gravity-related phenomena. In a second step, experiments were carried out with identical solidification parameters but with the furnace in a vertical position, and for two solidification directions (upward and downward). A comparative study between horizontal and vertical experiments was carried out. Phenomena related to gravity have been highlighted and their respective impact on the solidification front propagation was analysed.

1. Introduction

The mechanical properties of metal alloys strongly depend on the grain structure forming during the solidification step, so a precise control of the growth process is crucial in engineering. Two types of grain structures are commonly obtained during metal alloy solidification: a columnar grain structure with anisotropic properties, or an equiaxed grain structure with more uniform and isotropic properties. Equiaxed grain structure is required in most applications of aluminum-based alloys. The occurrence of equiaxed grains structures is often promoted by adding refining particles acting as preferential sites for heterogeneous nucleation. These particles are activated in liquid regions that are undercooled [1] and their efficiency depends on several characteristics: chemistry, amount, size distribution.

The control of the grain refinement involves the understanding of additional complex mechanisms, from nucleation to dendritic growth and solutal interactions. Grain structure prediction is made even more difficult when phenomena such as convection and buoyancy must be considered. Any density
gradient in the melt that is not suitably oriented with respect to the gravity vector can cause fluid flow. Even when solidification is performed in both thermal and solutal stabilizing configuration (i.e. upward solidification and increased liquid density with solute segregation), horizontal residual thermal gradients can initiate strong convective flow [2]. Convection in the melt can yield to significant inhomogeneity in solute distribution and thus liquid undercooling variations, and ultimately micro- and macro-segregation. Furthermore, newly formed solid grains can be carried away by fluid flow [3]. Upward motion of grains can also occur due to buoyancy forces when the liquid phase is denser than the solid phase [4], and downward motion (sedimentation) when the solid phase is denser than the liquid phase [5]. Experimental investigations of gravity effects in controlled environments are therefore of great interest for the validation of theoretical and numerical models.

In situ and time-resolved X-ray imaging is a method of choice for unveiling the dynamical evolution of dendritic microstructures and grain structure formation during the solidification of metal alloys [6]. Investigations taking advantage of the high brilliance of synchrotron sources began with the application of radiography [7], and a combination of radiography and diffraction imaging [8]. Then, the application of tomography enabled three-dimensional observations to be carried out [9]. Although radiography only provides a two-dimensional projected image, it remains an attractive imaging technique due to its ease of implementation. Recently, new opportunities arose with the improvement of compact micro-focus sources and X-ray sensitive detectors that enable in situ and time-resolved radiography to be carried out in laboratories, with a sufficient spatial resolution to distinguish the microstructure features [10], enabling different orientations of the sample with respect to gravity [11] and compatible with microgravity platforms [12].

The present investigation reports on the solidification of refined Al-20wt.%Cu samples observed in situ using the SFINX (Solidification Furnace with IN situ X-radiography) laboratory device. Experiments were carried out with identical applied parameters but with different orientations of the furnace with respect to the gravity vector. A comparative study between horizontal and vertical experiments was thus carried out to analyze and highlight phenomena related to gravity and their impact on the solidification front propagation.

2. Experimental details

2.1. SFINX facility

The SFINX facility is a duplicate of the device used during the MASER-12 sounding rocket mission [12] and parabolic flight campaigns [13]. These facilities were developed within the framework of the ESA MAP named XRMON (In-situ X-ray monitoring of advanced metallurgical processes under micro gravity and terrestrial conditions), devoted to the application of X-radiography during microgravity conditions [14].

The X-radiography system figures a micro-focus X-ray source with a molybdenum target (3 μm focal spot), which provides enough photon flux with two peaks of energy 17.4 keV and 19.6 keV that ensure a good image contrast to study Al–Cu based alloys. The camera system is made of a scintillator plate and a digital camera with a CCD-sensor. Due to the X-ray beam divergence, a geometric magnification of the object is observed at the detector, which is the ratio of the source-detector by source-sample distances (figure 1). In this work a magnification of approximately 5 was used, for a Field-of-View (FoV) of about 5 × 5 mm² leading to an effective pixel size of ~4 μm. The acquisition rate was set to 2 frames per second. The solidification furnace consists of two heaters, separated by a gap, that applies a longitudinal temperature gradient $G_{app}$. The gradient furnace enables directional solidification with applied temperature gradient within the range of 5–15 K/mm and cooling rates $R$ within the range of 0.01–1.5 K/s.

In the present work, the studied alloy was Al-20wt.%Cu inoculated with 0.1wt.% AlTiB. Sheet-like rectangular samples with dimension of 5 mm × 50 mm in area and about 250 μm in thickness were used. The sample was placed in the middle of stainless-steel spacers sandwiched between two flexible
glassy carbon sheets sewn together with a silica thread. The crucible was enclosed inside the furnace and meets both sides to achieve the expected thermal profile.

The furnace was set in two positions allowing either horizontal solidification of the sample with its main surface perpendicular to the gravity vector $g$ (figure 1a), or vertical solidification of the sample with its main surface parallel to the gravity vector $g$ (figure 1b and 1c). The horizontal configuration is chosen to minimize gravity-related phenomena such as buoyancy and thermo-solutal convection [15]. The vertical configuration is chosen to highlight gravity related-phenomena for two directions of the solidification: bottom-up ($G_{app}$ anti-parallel to $g$, figure 1b) and top-down ($G_{app}$ parallel to $g$, figure 1c) as previously termed by R. H. Mathiesen et al. [16].

![Diagram](image)

**Figure 1.** Schematic layout of the three configurations used to study the impact of the growth direction with respect to the gravity direction.

### 2.2. Image processing

During the sample solidification, the X-radiography system recorded a stack of raw images. The grey level variations in the acquired images are related to the attenuation coefficient of each different parts of the sample. Image legibility is enhanced by applying a “flat-field” correction, which consists in dividing each frame by a reference image recorded just before solidification. This procedure reduces the noise and removes defects related to the detector and crucible [17]. After applying the image processing, the bright regions in the processed radiograph are solid Al-grains whereas the dark regions are the Cu-rich liquid regions.

### 3. Results and discussion

In the present study, we will consider experiments with two different cooling rates, $R_1 = 0.1$ K/s and $R_2 = 0.9$ K/s, with the same applied temperature gradient $G_{app} = 7.5$ K/mm. These parameters were chosen to compare the evolution of the microstructure formation for relatively low and rapid solidification front velocities with respect to the achievable parameters of the furnace, and thus investigate the impact of gravity-related phenomena at different growth velocities.

#### 3.1. Impact of gravity related-phenomena on the solidification front at low growth rate

For each experiment, the directional solidification started with fully melted samples and the solidification front was observed passing through the field of view after the cooling rates were applied (figure 2a, 2b and 2c). During the horizontal solidification experiment, the equiaxed front propagated...
in a similar way to the observations reported by H. Nguyen-Thi et al. [18], A. Prasad et al. [19] or E. Liotti et al. [20] using synchrotron X-radiography, and recently by Xu et al. [21] by using a laboratory device similar to the present one. Several dendritic grains grew concomitantly towards the top of the field of view. This grain network formed an effective front that progressively created a constitutionally undercooled zone ahead. Then, a new series of grains nucleated and developed in the undercooled zone. The new grains eventually blocked the previous ones by solutal interactions and were incorporated to the effective front.

A significantly different solidification front was observed for the vertical experiment during upward solidification (figure 2b). Aluminum grains are buoyant for the present alloy composition [22]. Consequently, after nucleation they floated from the bottom to the top due to buoyancy force. The moving grains appear blurred on the images during their upward motion due to the too long acquisition time with respect to their floatation velocity (≈ 100 µm/s). The dendritic grains rapidly melted in the hot region of the sample until their complete disappearance. A similar behavior was reported by L. Abou-Khalil et al. [13] and G. Zimmermann et al. [23] for dendrite fragments detached from columnar structures. Only a few grains remained stuck in the thickness of the sample when their size was too large compared to the sample thickness. The solidification front was then composed of much larger grains than during horizontal solidification, with large liquid zones that ultimately lead to highly segregated areas.

![Figure 2](image_url). Radiographs of the solidification front for the low cooling rate ($R_l = 0.1$ K/s and $G_{app} = 7.5$ K/mm) during (a) horizontal solidification, (b) upward solidification and (c) downward solidification. The grey rounded shape surrounded by a white and black line in the bottom of the field of view in (c) corresponds to an artefact of the image processing following the formation of a porosity in this region of the sample (cf. figure 3c).

Downward solidification also led to the observation of a very different solidification front compared to both horizontal and upward solidification. Regarding the development of convective flow this configuration is both thermally unstable with the hot zone in the bottom part of the sample, and solutally unstable due to the rejection of heavy solute. No obvious effect of convective flow of thermal origin was detected, but large solute plumes made of Cu-rich liquid were seen developing toward the bottom part of the sample (figure 2c). It is worth noting that no segregated liquid channel was obtained for the presently investigated parameters, when this was the case in Ga-In alloys solidified upward (thermally stable and solutally unstable configuration) as reported by N. Shevchenko et al. [24]. In addition to the development of solute plumes, a striking difference with the previous configurations is the growth of very thin and elongated grains, even columnar grains in some areas. This observation suggests that many refining particles were inhibited by the propagation of the plumes that locally bring
solute-rich liquid. As consequence, the intensity of the undercooling in the liquid is significantly decreased in these regions, which prevents the nucleation of new grains on the refiners.

3.2. Solidification fronts at high growth rate

It has been shown that if the velocity of the solidification front propagation is higher than the characteristic velocity of gravity related-phenomena such as convective flow, then the microstructure formation is close to a solidification in a diffusive regime [25]. Experiments have been carried out with a high cooling rate to investigate such an effect.

The solidification front during horizontal solidification at high growth velocity (figure 3a) is analogous to the solidification front at low growth velocity (figure 2a) but composed of smaller and more compact equiaxed grains. It is worth noting that it is also comparable to the solidification front observed during reduced gravity periods aboard parabolic flights [13].

By comparing figure 3a for horizontal solidification and figure 3b for upward solidification, it is visible that both growth front are very similar. This can be attributed to the fact that the nucleation front in the upward experiment has been propagating at a faster rate (≈ 250 µm/s) than the average grain floatation velocity (≈ 100 µm/s). After nucleating, grains only have a short time to move upward before being stop by a layer of new grains that nucleated just above. In a similar way, the development of solute plumes was not observed at high growth rate during downward solidification, when the solidification front velocity is faster than solute sedimentation.

![Figure 3](image-url)

**Figure 3.** Radiographs of the solidification front for the high cooling rate ($R = 0.9$ K/s and $G_{app} = 7.5$ K/mm) during (a) horizontal solidification, (b) upward solidification and (c) downward solidification.

4. Conclusion

A study of the dendritic grain growth during directional solidification of a refined Al-20wt.%Cu alloy has been carried out for various solidification directions with respect to the gravity vector orientation. The *in situ* observation of the development of the solidification front was made possible at low and high growth rate by using the SFINX facility. Gravity-related phenomena strongly disturb the solidification front development at low growth rate, such as grain buoyancy during upward solidification and solute plumes during downward solidification. The impact of these effects becomes significantly reduced when the velocity of the solidification front is higher than both the characteristic grain floatation velocity and solute sedimentation. More detailed analyses of these phenomena are in progress to be compared quantitatively with analytical and numerical models in the future.
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