In situ X-ray observations of gas porosity interactions with dendritic microstructures during solidification of Al-based alloys

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Abstract. In situ X-radiography solidification experiments were performed on Al-based alloys, using both synchrotron and laboratory-based X-ray sources, in conjunction with a gradient furnace and a newly developed isothermal furnace, respectively. The effect of gas porosity nucleation and growth within the semi-solid mush during both columnar and equiaxed solidification was thereby observed. In all experimental cases examined, gas porosity was observed to nucleate and grow within the field-of-view (FOV) causing various levels of distortion to the semi-solid mush, and thereafter disappearing from the sample leaving no permanent voids within the solidified microstructure. During columnar growth, a single bubble caused severe remelting and destruction of primary trunks leading to secondary fragmentation and evidence of blocking of the columnar front. Equiaxed solidification was performed under microgravity-like conditions with restricted grain motion in the FOV. The degree to which the nucleated gas bubbles affected the surrounding grain structure increased with increasing solid fraction. However, bubble sphericity remained unaffected by apparent solid fraction or grain coherency.

1. Introduction
Defects in cast components, such as micro and shrinkage porosity, can be detrimental to service-life performance, causing a reduction in ductility and fatigue life [1,2]. While shrinkage porosity is largely due to restricted liquid transport in the semi-solid mush during the late stages of solidification, thereby causing voids in the as-cast microstructure, gas porosity is due to the effervescence of dissolved gas within the liquid melt as a consequence of lower gas solubility in the solid than in the liquid, e.g. hydrogen in aluminium alloys [3]. Traditionally, the method used to characterise porosity distribution in castings has been through post-mortem metallurgical examination of the as-cast microstructure. These examinations, however, provide little information on porosity-microstructural interactions during the early stages of alloy solidification. Real-time in situ X-radiography has emerged as a powerful tool in solidification science, providing invaluable and unique insights into early and transient solidification dynamics [4], e.g. nucleation, growth kinetics, porosity formation [5,6], etc.

Some brief in situ X-ray observations of gas porosity nucleation, interaction with dendritic microstructure during solidification, and subsequent disappearance from the sample are presented and
discussed. This work is the result of both laboratory and synchrotron-based *in situ* X-radiography solidification experiments.

### 2. Equipment and experiment description

The results presented were obtained from two independent experimental campaigns. The first involved the use of a Bridgman-type solidification furnace and synchrotron-based X-radiography, and the second used a newly developed prototype isothermal solidification furnace and laboratory-based microfocus X-radiography. Figure 1 shows the general arrangement of the *in situ* X-ray diagnostics setup used in both campaigns. The incident X-ray beam (arrow, figure 1) intersects the alloy sample at the location of the adiabatic zone inside the solidification furnace. The transmitted beam then projects an image onto the scintillator/CCD (charge-coupled device), showing the contrasting solid and liquid phases as light and dark zones in the field of view (FOV), respectively. The differing intensities at the scintillator/CCD are as a result of differing X-ray attenuation coefficients of the independent solid and liquid phases within the FOV giving rise to absorption contrast X-radiography.

**Figure 1.** Schematic illustration of *in situ* X-ray diagnostic setup. The X-ray beam passes through the aluminium sample and projects an image onto the scintillator/CCD. Dimensions $x$ and $y$ indicate the size of the projected FOV.

The synchrotron experiments were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, on beamline ID15A, using a high energy white beam (30 → 500 keV). Mathiesen *et al.* [6] described in detail the relevant characteristics of the furnace, along with the method of sample manufacture and encapsulation used. The alloy material used was Al-12wt%Ge inoculated with 0.15wt%Al-Ti-B (5/1) master alloy, of dimensions $30 \times 15 \times 0.2$ mm ($L \times W \times D$, figure 2). Solidification was promoted by pulling the sample through a constant thermal gradient ($G_T$) applied across the adiabatic zone, at a constant velocity ($V_p$). Spatiotemporal resolutions of 2.54 μm/px (pixel) and 0.045 s were achieved, resulting in a FOV measuring $1.3 \times 1.3$ mm on a Sarnoff fast-readout 512 × 512 px CCD.

**Figure 2.** Sample orientation relative to the force of gravity. Dimensions $D$, $W$, and $L$ represent the sample thickness, width, and length, respectively. $F_g$ denotes the direction of buoyant grain motion, w.r.t. gravity ($g$) for the alloys used.

The laboratory-based *in situ* X-radiography equipment used incorporated a Hamamatsu microfocus X-ray tube with a 7 μm focus spot operating at 6 W, outputting a 39° conically divergent white beam. The sample material was an Al-20wt%Cu sample inoculated with 0.1wt%Al-Ti-B (5/1) master alloy. Optimal settings for the X-ray tube with the Al-20wt%Cu system used were 55 keV × 120 μA. The X-ray detector comprised a Vosskuhler 11000XR CCD visible light camera fitted with a Scint-X
structured scintillator [7]. The camera was operated in 2 × 2 binning mode providing a 2012 × 1340 px CCD, with a physical pixel dimension of 18 μm. As a consequence of the high beam divergence (green cone, figure 1) a magnification effect was produced at the CCD, which is the ratio of the source-detector to the source-sample distance. Magnification in this case was measured at ~ 5.8 times, giving a virtual pixel size of 3.1 μm. The actual spatial resolution, however, is a function of both source size and scintillator pitch, thus the real spatial resolution was closer to 10 μm [7]. Images were recorded every 1 second.

Unlike the synchrotron experiments discussed previously, the solidification furnace used in this work was a newly developed prototype isothermal design. While a fully detailed description is beyond the scope of this article, the furnace comprised a circular alloy sample, measuring diameter, Ø 23 × 0.2 mm, surrounded by eight independently controlled heater elements arranged concentrically about the sample axis. The X-ray FOV was coincident with the centre of the sample. Solidification was initiated by reducing all heater temperatures at a constant cooling rate. The sample manufacture and preparation techniques used for the circular sample were largely similar to those reported for similar rectangular samples, previously [8].

The modular and compact nature of laboratory-based in situ X-ray equipment allowed the entire setup to be oriented in any direction w.r.t. the force of gravity. Figure 2 demonstrates this aspect from the perspective of the sample geometry. Figure 2 (a) shows the sample in the horizontal orientation, with the X-ray beam projection anti-parallel to gravity, g. In this configuration gravity acts through the thin dimension of the sample, D, resulting in significantly reduced equiaxed grain buoyancy in the FOV. However, in the vertical orientation (figure 2 (b)), which was the case for the synchrotron-based experiments, significant grain buoyancy was unavoidable, thereby adding significant complexity to characterisation of equiaxed nucleation and growth. Thus, to provide an environment to minimise equiaxed grain movement during solidification the X-ray diagnostics in this work were oriented such that the sample was horizontal, (figure 2 (a)).

![Figure 3](image)

**Figure 3.** Directionally solidified Al-12wt%Ge alloy showing porosity nucleation within the interdendritic mush, causing remelting and destruction of the dendritic network. $V_p = 85 \, \mu m/s$; $G_f = 79 \, K/mm$; $g \downarrow$. 

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3. Results and discussion

3.1. Solidification experiments using synchrotron X-radiography

Figure 3 demonstrates the effect of porosity nucleation within the interdendritic mush on the evolving microstructure. Columnar growth was initiated by pulling the sample, at a constant speed $V_p = 85 \, \mu m/s$, through a relatively high thermal gradient, $G_T = 79 \, K/mm$, similarly described in Ref. [6]. The indicated image time, $t = 0 \, s$, represents a datum time prior to the observed bubble nucleation. At $t \approx 1.53 \, s$, a bubble was observed nucleating deep within the interdendritic mush and, thereafter, relocated to a position coincident with a primary dendrite trunk. As the bubble continued to grow, remelting of the microstructure was observed at the bubble location, causing the top portion of the primary columnar dendrite to buoyantly rise out of the mushy zone, consequently melting as the fragment approached the super-heated liquid. Concurrently the surrounding columnar dendrites also experienced remelting, developing a large interdendritic pocket, as a consequence of the bubble development. The pore achieved a maximum diameter $\approx 200 \, \mu m$, i.e. of the order of the sample thickness, and, thereafter, disappeared from the FOV. The cause of the microstructure remelting was likely the bubble acting as a thermal baffle within the mushy zone. Heat flow from the top of the FOV to the bottom, as a consequence of the imposed thermal gradient, was, therefore, restricted, causing a build-up of heat within the mushy zone ahead of the pore, thus leading to the remelting observed.

3.2. Solidification experiments using laboratory microfocus X-radiography

Figure 4 shows the equiaxed dendritic mush apparent in the FOV starting from ~ 76 seconds after the first $\alpha$-solid nucleation event was observed. Note, image time, $t$, was set to zero at ~ 8 seconds prior to the onset of equiaxed nucleation, while the sample was fully liquid. The temperature indicated on each of the images in figure 4 was defined based on the assumption that the sample temperature at the instance of first equiaxed nucleation was equal to the liquidus temperature of the alloy, i.e. $T_L = 602.6 \, ^\circ C$ at $t = 8 \, s$. Solidification was initiated by application of a constant cooling rate of $T = 0.084 \, K/s$ near-isothermally across the entire sample. The total FOV dimension shown in figure 4 was limited to $3.17 \times 3.17 \, mm$ to save space as well as to remove some CCD edge effects. Analysis of the real-time image data showed the temperature difference between nucleation commencement and nucleation cessation within the FOV as $\Delta T \approx 1.5 \, K$, with no significant aberrant temperature gradient evident.

The image sequence in figure 4 reveals the effects of hydrogen porosity nucleation and growth on equiaxed grains during solidification, with the additional complexity that bubbles, after reaching a diameter approximately equal to the sample thickness (200 $\mu m$), then disappeared from the FOV. The arrows in figure 4 (a) show the position in the FOV where bubble nucleation was first observed for a solid fraction of $g_s = 0.13$. In both cases highlighted, bubbles were observed to nucleate in approximately the same location as an equiaxed grain. Consequently, the bubbles, remaining stationary, increased in diameter and pushed the equiaxed grains from their original nucleation positions. Comparing the highlighted grains in figure 4 (a-iii) with the original grain positions (figure 4 (a-i)) it is clear that prior to mechanical grain coherency the effect of bubble nucleation was localised to grains within the immediate bubble vicinity. Finally, however, as shown in figure 4 (a-iv), both bubbles eventually disappear from the FOV leaving no permanent visible trace in the semi-solid microstructure. Maximum diameters measured were $d \approx 210 \, \mu m$ and $d \approx 225 \, \mu m$ for the top and bottom bubbles in figure 4 (a-iii), respectively. Figure 4 (b) shows the equiaxed dendritic microstructure at an apparent solid fraction of $g_s \approx 0.47$, approximately 384 s after figure 4 (a-iv). At this stage during solidification the equiaxed microstructure was more densely packed, with mechanical grain impingement clearly apparent, figure 4 (b-i). The arrows in 4 (b-i) indicate the position of a single pore nucleation event within the semi-solid mush. With such a high solid fraction, however, the distortion of the dendritic mush was more far-reaching, as illustrated by the highlighted regions, figure 4 (b-i) - (b-iii)). Interestingly, the maximum diameter achieved by the bubble was measured at $d \approx 198 \, \mu m$, which suggests the coherent microstructure was insufficiently rigid to cause any distortion to the
pore shape. Further, as observed earlier during solidification, once the nucleated bubble reached a maximum diameter, it disappeared from the FOV this time leaving a relatively large intergranular spacing in the microstructure, (arrow in figure 4 (b-iv)).

![Figure 4](image-url)  

**Figure 4.** Near-isothermally solidified grain refined Al-20wt%Cu sample. Cooling rate, $\dot{T} = 0.084$ K/s. Rows (a), (b), and (c) show the apparent solid fractions ($g_S$) 0.13, 0.47, and 0.52, respectively. Columns (i), (ii), (iii), and (iv) show time increasing from left to right. Arrows indicate the location of porosity nucleation. Colourisation shows which grains were affected by the nucleation and growth of pores; blue (column (i)) represent pre-pore nucleation position, red (column (iii)) represents the maximum mush distortion observed.

Figure 4 (c) shows a similar case to figure 4 (b) with a further increase in the apparent solid fraction, $g_S \approx 0.52$. The remaining liquid was close to the eutectic composition and thus the sample was almost completely solidified. Once again a gas bubble nucleated in the intergranular liquid, pushing and distorting the microstructure further than previously observed (highlighted regions, figure 4 (c)). As with the previous case, however, no distortion was observed in bubble morphology, maintaining relatively uniform sphericity at a high apparent solid fraction, and reaching a maximum diameter, $d \approx 218$ μm. As before, when the bubble disappeared from the FOV a large liquid void remained in its place. Approximately 29 seconds after figure 4 (c-iv) was captured, the eutectic transformation was observed crossing the FOV from the bottom-left corner towards the top-right corner. Figure 4 (c-iv), however, represents the final as-cast microstructure observed, showing no detectable signs of porosity in contrast to the real-time imaging which demonstrated bubble nucleation continuously during solidification.
4. **Summary, conclusion, and outlook**

This article presents a brief analysis of gas porosity nucleation and growth observed at various stages during both columnar (directional) and equiaxed (isothermal) *in situ* X-ray solidification experiments. In all cases examined the maximum pore diameter appeared to be limited by the sample thickness, i.e. 200 μm, suggesting that once the pore came into contact with the free surface oxide layers the gas was expelled from the sample. After the pore disappeared from the FOV no significant trace remained within the semisolid mush, giving a further demonstration of the importance of real-time *in situ* studies. During columnar growth the nucleation and growth of a gas bubble deep within the dendritic mush caused rapid remelting of the dendritic network and significant fragmentation of the remaining primary and secondary dendrite arms. These fragments continued to grow, both ahead and within the dendritic mush, causing a CET-like (Columnar-to-Equiaxed Transition) phenomenon. While the experimental evidence is somewhat limited, it would seem that gas bubbles nucleating in the dendritic mush and causing remelting/fragmentation may be possible as a further mechanism of CET formation. Pore growth velocity measurements for the near-isothermal equiaxed (microgravity-like) solidification experiments showed no significant dependence on temperature. However, a slight decrease in pore growth velocity was observed as the solid fraction increased. This was related to the reduction in the physical space available for the pore to grow, thereby requiring the gas volume to push against larger and larger regions of interconnected equiaxed dendrites. Most significant was the lack of any observed distortion in sphericity of the gas bubble as a consequence of interaction with the dendritic structure suggesting relatively high internal gas pressure.

Finally, the near-isothermal solidification experiments and analysis presented in this work are currently in their infancy. Development and testing of the prototype isothermal furnace for *in situ* X-radiography solidification studies of Al-based alloys is in the final stages. The furnace is scheduled to fly on board the MASER 13 sounding rocket in spring 2015, investigating near-isothermal equiaxed solidification under microgravity conditions.

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