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A method to determine the active particle nucleation undercooling distribution in a refined alloy

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Abstract. We propose a method to determine the active particle distribution of nucleation undercooling in a refined alloy. The experimental data used in this work are inferred from solidification experiments on a refined Al-3.5 wt% Ni alloy performed with X-ray radiography at the European Synchrotron Radiation Facility. These in situ and real time observations allow the accurate and direct determination of the grain origin (heterogeneous nucleation on particles or fragmentation), of the density and of the equiaxed front growth rate. The LGK classical dendrite growth model is used to evaluate the front undercooling (ΔTc) corresponding to the measured equiaxed front growth rate. Then, the corresponding cumulative distribution of active refining particles is determined. From this cumulative distribution, we derive the corresponding Gaussian and log-normal laws to obtain the nucleation undercooling distribution of active particles. Results are discussed and compared to available measurements in the literature. The standard particle distribution parameters (density of nuclei, mean nucleation undercooling and standard deviation) are determined. We plan to use the determined nucleation undercooling particle distribution in a stochastic CAFE model for the grain structure without preliminary adjustment of the nucleation undercooling.

1. Introduction
Al-based alloys are used primarily because of their mechanical properties for structural applications (e.g. automotive industry). During industrial casting, different types of grains structures form in Al alloys, which vary from dendritic columnar to globular/dendritic equiaxed. The grain structure is called columnar if the grain growth is oriented in one direction, whereas equiaxed grains are growing evenly in all directions. To control the final grain structure, it is thus essential to understand the physical mechanisms, which govern the transition from columnar to equiaxed growth (CET). When a fine grain structure is desired, adding particles to the molten metal is a convenient way to provide a high number of substrates for heterogeneous nucleation of equiaxed grains. Indeed, grain refining is widely used in the aluminium shaped casting industry to foster equiaxed growth and obtain a fine grain structure. However, grain refinement is still a pending key issue for the foundry industry [1] as, aside the processing parameters (temperature gradient and growth rate), the active particle distribution plays a major role on the final grain size and morphology. In particular, the distribution of nucleation...
undercooling corresponding to the active particles is required in all existing models predicting CET and its determination remains a major challenge.

Few authors studied CET or grain refinement taking into account the nucleation mechanisms on particles. Schumacher et al. [2] made an important contribution on the nucleation mechanisms on refining particles using TEM characterisation. Greer and co-authors [3] developed an average model for the prediction of as-cast grain size from inoculated melts. According to Greer et al. [3] the grain size depends on the cooling rate of the melt, its solute content and the amount of added inoculant. Free growth of a crystal starts on a given particle at an undercooling inversely proportional to the particle diameter [3] so that the distribution of particle size is equivalent to the particle nucleation undercooling distribution. However, the particle efficiency can, among others, be restricted by the poisoning of particles [2, 4] and interaction with latent heat and solute release by the already growing equiaxed grains. Easton et al. [5] proposed a nucleation model driven by constitutional supercooling to take the effect of the solute diffusion field on heterogeneous nucleation into account. Recently, Shu et al. [6] studied in particular the nucleation potency in isothermal conditions, and showed the significant effect of the solute field built around a growing equiaxed grain. In stochastic models, the columnar-to-equiaxed transition (CET) or the gradual change of grain structure morphology occurs naturally without definition of any blocking criterion [7].

Due to the implementation difficulty, few grain structure models [7] can take into account a complex and non uniform distribution of nucleation undercooling for refining particles. However, the distribution of active particles and/or associated nucleation parameters is needed in all models. If they are not available, adjustment of the standard distribution parameters is performed to find a good agreement between the calculated and the experimentally determined grain structure [8]. Despite this established necessity, there is a lack of studies on the determination of these missing parameters, which are of prime importance for models and numerical predictions. By making use of differential scanning calorimetry, Jung et al. [9] previously determined the mean nucleation undercooling of active refining particles.

The objective of the work reported in the present paper is to determine the nucleation undercooling distribution of active refining particles from experimental results obtained by in situ X-ray synchrotron imaging [10]. The present work demonstrates that we can determine the particle nucleation undercooling probability density function for a refined Al-3.5 wt% Ni alloy. Moreover, the method could be extended to other refined alloys.

2. Experimental procedure
The experiments are carried out at ID19 beamline at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France), where in situ and real time observation of the solidification process is possible using X-ray synchrotron radiography. A thin sample of Al-3.5wt%Ni (40 mm long, 6 mm wide and 200 µm thick) is solidified in a Bridgman furnace. The global solidification and imaging device was previously described in details [10]. The grain refiner used is Al-5wt%Ti-1wt%B master alloy 0.5wt% of which is added to the alloy. The temperature gradient (G) is kept constant and is about 28 K/cm in all experiments analysed here. The samples are initially in the hot zone of the furnace where they are liquid. Then, they are pulled down in the lower and colder zone where they start to solidify. In all these experiments, the first applied pulling rate for which a columnar front is obtained is 1.5 µm/s. A higher pulling rate is then applied to provoke a macrostructural transition in the sample from columnar to equiaxed growth [10]. In order to quantify the grain size, the equivalent diameter is calculated based on the measurement of the real surfaces of the grains, which are later converted into circular areas. The key advantage of the X-ray characterization method is to provide dynamical observation, which allows to measure the real front growth rate and to check the origin of equiaxed grains (heterogeneous nucleation on particles or fragmentation). The growth, sedimentation and interaction of equiaxed grains can also be followed.
3. Results and discussion

3.1 Grain structure evolution as a function of the equiaxed front growth rate

Figure 1 shows the final morphology induced by the different pulling rates applied during the Bridgman solidification experiments at a constant temperature gradient. The equiaxed front growth rate ($V_f$) is measured in the region shown in the images of figure 1 using the time sequence of X-ray radiography images available for each experiment.

![Figure 1](image)

**Figure 1.** Equiaxed microstructures of a refined Al-3.5wt%Ni alloy, $G \approx 28$ K/cm and the measured equiaxed front growth rate $V_f$: (a) 6.5 $\mu$m/s; (b) 10 $\mu$m/s; (c) 16 $\mu$m/s; (d) 38 $\mu$m/s.

A strong variation in the mean grain size can be seen in the range of rates shown figure 1. The grain size decreases with increasing growth rate as more particles are activated. In contrast, a weak decrease of the grain size is observed for higher velocities. This tendency is evident on figure 2 showing the measured equivalent diameter (diameter of the circle having an area equivalent to the grain area) as a function of the measured front growth rate.

![Figure 2](image)

**Figure 2.** Measured equivalent diameter as a function of the measured front growth rate for refined Al-3.5wt%Ni alloy at $G \approx 28$ K/cm. The horizontal dashed curve represents the saturation limit giving a limit diameter: $d_0 \approx 200 \mu$m.

In these experiments, the constitutional undercooling above the dendritic front allows the nucleation of equiaxed grains on particles, which can become active (i.e. particles with the nucleation undercooling, $\Delta T_N$ lower than or equal to the front undercooling, $\Delta T_C$). While the undercooling increases, the growth rate increases as well. This allows a larger number of particles to become active. For higher front growth rate, the grain size is almost constant and saturation is achieved in these
conditions. Saturation is due to several mechanisms including the high (and increasing when the
growth rate increases) amount of latent heat and solute release during the solidification of equiaxed
grains, the particle trapping inside the already growing grain mush, and also the fact that all active
particles in the distribution are consumed.

From figure 2, we extrapolate the curve using a fit to determine the limit equivalent diameter
(corresponding to a surface density). We then calculate the volume density of active particles \( N_0 \) by
multiplying the surface density by the sample thickness and we obtain \( N_0 = 1.3 \times 10^{11} \text{m}^{-3} \). The volume
density measured in thin samples is of the same order of the one measured in 3D cylindrical samples
of the same alloy solidified in comparable conditions [11].

3.2 Method for the determination of the particle distribution
To determine the probability density function of the active particles, it is necessary to establish a curve
of the active particle density as a function of the nucleation undercooling i.e. the probability
 cumulative distribution function.

\[
\begin{align*}
\text{Figure 3. Schematic drawing of the active particle density function as a function of the nucleation}
\quad \text{undercooling: (a) for a low growth rate case } V_{f1} \text{ and, (b) for a medium growth rate } V_{f2} > V_{f1}.
\end{align*}
\]

For low growth rates, small front undercooling is reached so that only refining particles with small
nucleation undercooling will become active. This case is schematically represented on figure 3(a)
where we use a hypothetical Gaussian probability function for the particle distribution. Only particles
with \( \Delta T_N \) lower than or equal to the columnar front undercooling \( \Delta T_{c2} \) corresponding to the growth
rate \( V_{f1} \) have a chance to become effectively active. Figure 3(b) represents a medium case for a higher
growth rate \( V_{f2} > V_{f1} \) where potent particles with a \( \Delta T_N \) lower than or equal to the columnar front
undercooling \( \Delta T_{c2} \) for the growth rate \( V_{f2} \) become active, including the ones already active in the case
of growth rate \( V_{f1} \). When all particles from the distribution of potent particles (i.e. excluding trapped
or inactive by latent heat or solute released) have been consumed, the limit case is reached, as no more
particles are available. Increasing the front undercooling from this state would not induce any new
grain nucleation. Thus, we construct the cumulative distribution function by noting that for each
growth rate step, the maximum nucleation undercooling of active particles is equal to \( \Delta T_{c2} \), i.e. the
nucleation undercooling of particles is between 0K and \( \Delta T_{c2} \).

We calculate the front undercooling from the measured equiaxed front growth rate using the LGK
model [12]. Figure 4(a) shows the measured growth rate as a function of the corresponding calculated
columnar front undercooling. Additionally, for each growth rate, we can measure experimentally an
equivalent diameter of the equiaxed grains, diameter that can be converted in a grain density
equivalent to the active particle density. We can then draw the grain density as a function of the front
undercooling equivalent to the maximum nucleation undercooling for each step (figure 4(b)).
3.3 Determination of the particle distribution law

According to the discussion in the previous paragraph, figure 4(b) is in fact the cumulative distribution function \( R(x) \) of the active particles as a function of the nucleation undercooling. From this cumulative function, we can calculate by integration the probability particle density function \( f(x) \).

The relationship between the cumulative distribution function \( R(x) \) and the probability density function \( f(x) \) is given by equation (1):

\[
R(x) = N_0 \int_{-\infty}^{x} f(t)dt
\]  

(1).

To obtain the probability function, we need to postulate the shape of the function \( f(x) \). As a first step, we calculate the corresponding Gaussian distribution which is used for instance by Guillemot et al. in a stochastic model [13]. For a Gaussian distribution, the function \( f(x) \) is given by:

\[
f(x) = \frac{N_0}{\sigma \sqrt{2\pi}} e^{-\frac{(x-\mu)^2}{2\sigma^2}}
\]  

(2)

with \( N_0 \), the volume particle density, \( \mu \) the mean nucleation undercooling and \( \sigma \) the standard deviation of the density function. With this assumption, we calculate the cumulative distribution and the
The corresponding Gaussian distribution shown respectively in figures 5(a) and (b) (dashed curves). For the Gaussian law, the mean nucleation undercooling ($\mu$) is 2.5K and the standard deviation ($\sigma$) is 0.8K.

Nevertheless, Greer et al. [3] measured such a particle distribution on commercial alloys and found that f(x) is actually closer to the log-normal probability density function. Consequently, we also calculate the log-normal probability particle density function for which the f(x) is given by:

$$f(x) = \frac{N_0}{x\sigma\sqrt{2\pi}} e^{-\frac{(\ln x - \mu)^2}{2\sigma^2}}$$  (3)

With this assumption, we calculate the cumulative distribution, and the corresponding log-normal distribution is shown respectively in figures 5(a) and (b) (plain curve). For the log-normal distribution, the mean nucleation undercooling ($\mu$) is 0.85K and the standard deviation ($\sigma$) is 0.3K. Using differential scanning calorimetry, a mean value of 0.9K was found for the critical nucleation undercooling in a refined Al-3.5wt%Ni [9]. This value is very close to the average value obtained in the present study for the log-normal distribution. Moreover, a value of nucleation undercooling lower than 1K was taken for this alloy in other studies to adjust numerical modelling and experiments [8].

4. Conclusion

A method to determine the particle nucleation undercooling distribution of active particles is proposed. According to this method, we derive for Al-3.5 wt%Ni alloy refined with a classical refiner (0.5wt % of Al-5wt%Ti-1wt%B) a Gaussian law with a mean nucleation undercooling of 2.5K and a standard deviation of 0.8K, and a log-normal law with a mean nucleation undercooling of 0.85K and a standard deviation of 0.3K. For both cases, the volume density is given by $N_0 = 1.3 \times 10^{11}$ m$^{-3}$. Considering previous experimental determinations and comparison to values adjusted in previous works, the log-normal distribution seems to give the best representation of the particle distribution. This distribution is now available and can be used, for instance, as input data in predictive models of grain structure in order to compare to experimental results. We plan to use CAFE model of the grain structure in this alloy using the parameters determined in the present work to compare the grain structure, the grain size, the grain elongation without preliminary adjustment of the particle distribution.

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