Numerical modelling and in-situ radiographic study of the grain nucleation and growth of inoculated aluminum alloys

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Abstract. To precisely predict the grain size of inoculated aluminium alloy castings has been a big challenge for the researchers in the field of solidification and casting. Up to date, most of grain size prediction models are based on the Free Growth Model, in which the nucleation process is stopped by recalescence. In a previous work [1], we have proposed a new grain size prediction model applicable for solidification of castings without recalescence. In the present work, an in-situ X-ray radiographic study on the grain nucleation and grain growth of inoculated Al-Cu alloys during isothermal melt solidification has been carried out, where the effect of melt convection is minimized. The influences of inoculant particles and cooling rate on the nucleation rate, grain growth rate and final grain size have been quantitatively studied. A comparison between the prediction results and the experimental results is presented.

1. Introduction
Grain refinement by inoculation is an important technique to improve the soundness of aluminium castings. Much research effort has been made in the last decades to develop analytical and numerical models to simulate the heterogeneous nucleation and growth of equiaxed grains in aluminium alloys during solidification [2-11]. The first numerical grain size prediction model for isothermal melt solidification of aluminium alloys has been attributed to Hellawell and Maxwell [2]. In 2000, Greer et al. proposed the Free Growth model [5]. The main progress of this model, in comparison to the Hellawell & Maxwell (H&M) model and other earlier solidification models, is that the heterogeneous nucleation is treated as a deterministic, instead of stochastic, process, where the size of potent nucleant particles was directly related to the undercooling needed for the initiation of free-growing grains. This makes the evaluation of the grain refinement efficiency of grain refiners with different size distributions possible. Similar to the H&M model, the heterogeneous nucleation of grain stops due to the recalescence. The model has been used to simulate the isothermal melt solidification and the simulated grain size is in a good agreement with the measured grain size of standard grain refining test (TP-1) samples, which have an approximately isothermal melt condition during solidification. However, the solidification of most real castings is far away from isothermal melt solidification and recalescence can not always be observed, which makes the application of the Free Growth Model has been limited to the TP-1 type castings. To overcome this limit, in a previous work[1], we proposed a new grain size prediction model based on a new nucleation stopping mechanism, solute stifling mechanism. Such a solute stifling mechanism has been discussed by Shu et al.[10], and Quested et al.[7] but in a less critical way. Although based on isothermal melt solidification, our model has been
validated with a DC-casting 5182 alloy, showing a reasonable agreement with experimental results. With the aim to make a more precise evaluation of the model, an isothermal melt solidification experiment has been designed and carried out by using a novel in-situ X-ray radiographic approach.

2. Experimental
The experimental alloy used in this work is Al–10 wt.% Cu alloy prepared by using 5N purity aluminium and 4N purity Cu. After melting, the alloys grain refined with different amounts of Al5Ti1B grain refiners were cast in copper molds of 50mm in diameter and 150mm in height. The cast ingot were then machined and mechanically polished into the final sample dimensions of 50 x 5 x 0.2 ± 0.05 mm. No further heat treatments or coatings were applied to the samples prior to installation into the crucible assembly.

The solidification furnace used for this experiment was a bespoke Bridgman-type gradient furnace (GF), manufactured by the Swedish Space Corporation (SSC) in cooperation with the European Space Agency (ESA) Applications Microgravity Promotion (MAP). By a careful temperature control, desirable temperature gradient can be realized in the sample melt during solidification. The polished thin alloy samples capsuled by glassy carbon are positioned in a sample holder device, which is designed specifically to maximize heat flow through the sample. In this work, the thin plate samples are melted and solidified in a horizontal position, namely, the wide surface of the sample is perpendicular to the gravity direction, which could to a large extent reduce the influence of convection. Constant cooling rates in the range of 0.025–1 K/s were applied for the solidification process. For all the samples, the cooling process started after the samples were completely melted and heated up to 670 °C. An in-situ micro focus X-ray monitoring system was used to record the microstructure evolution during solidification of the castings. The frame capture rate throughout these experiments was 1.5 Hz. A more detailed description of the furnace and experimental procedure can be found elsewhere [12].

3. Model description
The grain size prediction model used in this work is based on the as-cast grain size prediction model reported in [1]. The same free grain growth criterion and the same growth velocity equation of grains (assumed as spherical growth) solely controlled by solute diffusion are applied. The diffusion field surrounding each growing solid grain is at quasi-steady state, namely the concentration profile of the solute keeps the same shape during grain growth.

\[
C(r) = C_0 + \frac{R_g}{r} (C^* - C_0) \quad \text{(Eq. 1)}
\]

where \(C_0\) is the initial alloy composition, \(C(r)\) is the solute concentration in the liquid with a distance of \(r\) to the center of the growing grain, \(C^*\) is the liquid concentration at the surface of growing grain and \(R_g\) is the radius of the grain and \(R_0\) the radius of inoculant particle. The same size distribution function of inoculation particles adopted by Greer et al., [5] is used in the present model.

\[
PSD(d) = N_v \frac{\exp\left(-\frac{d}{d_0}\right)}{d_0} \quad \text{(Eq.2)}
\]

where \(d\) is the diameter of disc-shaped inoculant particle , \(d_0\) is the characteristic width of the distribution (\(d_0 = 0.72 \text{ \mu m}\) ) and \(N_v\) is the total number of TiB_2 particles per unit volume of aluminum melt when Al-5Ti-1B grain refiners are added. \(N_v\) was calculated according to the empirical rule [5]: for 1 Part Per Thousand (PPT) by weight Al-5Ti-1B addition, \(N_v\) is equal to 1.15\times10^{12} \text{ m}^{-3}.\) No tuning was carried out in the present work.

The nucleation process stops due to the solute stifling due to the of the solute segregation layer surrounding growing grains in the melt.

\[
\dot{T} - \dot{T}^i(\bar{X}_i) = 0 \quad \text{(Eq.3)}
\]

Where \(\dot{T}\) is the temperature changing rate, \(\bar{X}_i\) is the average concentration of element \(i\) in the residual liquid of the metal and \(\dot{T}^i(\bar{X}_i)\) is the change rate of the liquidus of the bulk melt.
4. Results and discussion

Fig. 1 shows the grain structure evolution of Al-10%Cu alloy grain refined by 0.5PPT (0.05wt %) Al5-Ti-1B grain refiners during the in-situ radiographic solidification at two different cooling rates.

As can be seen, in both cases, equiaxed grains form and grow in the samples and the number of grains increase with increasing cooling time (temperature decreasing). It has to be noted that the very early stage of the grain growth could not be observed due to the resolution limit. By a careful observation, it can be seen that equiaxed grains appear slightly earlier in the right side than in the left side of the samples, indicating that a small temperature gradient exists in the sample melt. The temperature gradient is measured as about 0.2 K/mm. It is interesting to see that the cooling rate has a strong influence on the heterogeneous nucleation of the grains in the alloy. The final number density of

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Fig. 1. In-situ X-ray radiographic images showing the grain structure development of Al-10Cu alloy inoculated by 0.5 PPT (part per thousand) Al5Ti-1B grain refiners, with two different cooling rates, 0.5K/s for images at the left side and 0.025 K/s for the images at the right side. The images were recorded when the temperature of the alloy melt reached 645 °C.
grains in the sample solidified with a cooling rate of 0.5 K/s (Fig. 1e) is much higher than that cooled with a cooling rate of 0.025 K/s (Fig. 1f).

The final grain size of Al-10wt%Cu alloy inoculated by different amount of grain refiners under different cooling rate has been measured. The measured grain size as a function of cooling rate is plotted in Fig. 2. As can be seen, the grain size decreases with increasing cooling rate and increasing addition level of grain refiners. As a comparison, the calculated grain size by the grain size prediction model is also presented in the figure. It is interesting to see that the predicted grain sizes have a good agreement with the experimental results in trend, although the absolute quantity of the predicted grain size is slightly lower. If considering that no tuning was done with the size distribution of TiB₂ particles in the master alloy and it is assumed 100% yield of the added inoculant particles, the prediction results of the model is satisfying. It confirms that our grain size prediction model based on the nucleation stopping by solute stifling mechanism is feasible.

Fig. 2 The measured grain size (based on the final number density of grains) and the predicted grain size of Al-10Cu alloy inoculated by two different level of Al-5T-1B master alloy, 0.1 and 0.5 ppt (part per thousand) under different cooling rates.

By using the in-situ radiographic images, the grain size evolution of the grains during solidification has also been investigated. The equivalent grain radius of the equiaxed grains were obtained by measuring the area of the 2-D projection image of the grains. It has to be noted that the grain size of the grains obtained by such a treatment is larger than the real 3-D equivalent grain size. Fig. 3 shows the evolution of the measured grain size of an equiaxed grain shown in Fig. 1(b) in comparison with the predicted grain size evolution of a grain (nucleated on a inoculant particle of 4µm in radius) during solidification with a cooling rate of 0.025 K/s. As can be seen, the predicted grain size is close to the measured size in the early stage of grain growth, but smaller than the measured grain size in the late stage, which is partly attributed to the over estimation of the grain radius by measuring the 2-D projection image. It has been determined that the grain nucleation stopped within 30 seconds since the first equiaxed grain was observed, namely the maximum nucleation undercooling is less than 0.7K. Within such a short time interval, the predicted grain radius is very close to the real grain size evolution. It suggests that the spherical grain growth is a reasonable assumption for the grain size prediction purpose.
Fig. 3 The evolution of the measured equivalent radius of a single growing grain shown in Fig. 1(b), in comparison to the predicted grain radius evolution of a grain nucleated on an inoculant particle with radius of 4 \( \mu \text{m} \). The measured grain radius is calculated from the measured projection area of the grain.

The thickness of the solute diffusion layer around the growing grains has a strong influence, in terms of solute stifling[1,7], SSN[10] or nucleation free zone (NFZ)[11], on the nucleation of new grains in the melt surrounding the growing grains. Fig. 4 shows the evolution of the predicted ratio between the thickness of solute diffusion layer and the radius of growing grains with different initial grain radius during solidification with cooling rates of 0.025 K/s (Fig.4a) and 0.5 K/s (Fig. 4b). It shows that, until the maximum nucleation undercooling is reached, the ratio \( \frac{\delta_c}{R_g} \) is less than 10 for the slow cooling rate case, while less than 7 in the fast cooling case. These calculated \( \frac{\delta_c}{R_g} \) values are much less than the value obtained by the approach by Shu et al[10], but is consistent with our previous analysis in [1,13] on \( \frac{\delta_c}{R_g} \), which gives a range between 2.5 and 25.

Fig. 4 Evolution of the predicted ratio between the thickness of solute diffusion layer \( \delta_c \) and the radius of growing grains \( R_g \) with different initial grain radius \( (R_i) \) during solidification with cooling rates of (a) 0.025 and (b) 0.5 K/s.

Fig. 5
5. Conclusions
An in-situ X-ray radiographic study on the solidification of inoculated Al-10wt%Cu alloy under nearly isothermal melt solidification with constant cooling rate is realized. The evolution of the size, morphology and number density of equiaxed grains during solidification has been quantitatively studied. The influence of cooling rate and addition level of grain refiners has been revealed.

A new grain size prediction model based on a new nucleation stopping mechanism, solute stifling mechanism, has been applied to model the heterogeneous nucleation of the of the experimental alloys during isothermal melt solidification. It shows that the grain size prediction model could well address the influence of cooling rate and addition level of grain refiners and, most importantly the predicted grain size is in a good agreement with the measured grain size.

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