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# The Interdependence model of grain nucleation: A numerical analysis of the Nucleation-Free Zone

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**Abstract**

Predictions by a 1-D analytical model and a 3-D numerical model of the formation of the Nucleation-Free Zone (NFZ) surrounding each growing grain during the initial transient of equiaxed solidification are compared. The extent of NFZ formation was studied under different solidification conditions in 1-D, 2-D and 3-D for both single-grain and multiple-grain growth scenarios with different geometric grain arrangements. The previously hypothesised NFZ concept presented by the analytic Interdependence model has been clearly demonstrated to exist for a range of solidification conditions. While there is good agreement between the 1-D numerical and analytic models, the 2-D and 3-D simulations of NFZ formation demonstrate that for some conditions the analytic model should be rederived in spherical coordinates. Further, the strong influence of the overlap of the diffusion fields between neighbouring grains was clearly demonstrated, revealing that the effect of competition between the rate of solute accumulation and cooling rate determines whether or not additional nucleation events are able to occur. It is also shown that a judicious choice of the growth rate term is essential for the analytical model to provide an accurate prediction of NFZ. Application of the computationally intensive 3-D simulations has allowed an improved solution to be derived that can be run at very low computational cost.


*Keywords:* Grain refinement; Nucleation; Nucleation-free zone; Numerical solution; Undercooling

## 1. Introduction

It is well established that cast products with fine equiaxed grains possess superior mechanical properties. Grain refinement is typically achieved using inoculants added in the form of master alloys containing potent nucleants, e.g. $TiB_2$ in Al alloys. Although grain refinement has been practised for several decades the exact mechanism of grain refinement has been elusive.

A review by Easton and StJohn [1,2] of grain nucleation theories for Al alloys showed that in addition to potent nucleant particles, growth-restricting solute and the associated development of constitutional supercooling (CS) is a necessary condition for achieving good refinement. Since then several analytical models of increasing rigor have been developed [3–5], leading to the development of the Interdependence model in 2011 [6].

The Interdependence model is an analytical model that predicts the possibility of nucleation events occurring while the already nucleated grain grows. The nucleation event is governed by the degree of CS ahead of the growing grains. The concept of CS-driven nucleation was first proposed by Winegard and Chalmers in 1954 [7]. Maxwell and Hellawell [8] developed a predictive model wherein grain nucleation occurs in the bulk at a given undercooling, following which growth takes place in the form of a sphere which subsequently changes to dendritic growth. Several columnar-to-equiaxed transition (CET) models have been developed in the past [9–22] to study the phenomena of

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nucleation of equiaxed growth in the centre of the casting ahead of columnar grains growing from the mould wall. For instance, Hunt [13] developed an analytical model of equiaxed grain formation in the CS region ahead of the growing columnar front. On the other hand, the Interdependence model ignores columnar growth and concentrates solely on the equiaxed grain nucleation and growth mechanism after the CET has taken place. In terms of equiaxed grain growth, the model developed by Thevoz and Rappaz [23,24] and Thevoz et al. [25] solves for the equiaxed grain growth within a casting. However, nucleation density is predetermined and consequently the average grain size is fixed a priori [23]. In the other case [25], nucleation is made to occur beforehand based on an imposed cooling rate and a given inoculant size distribution. Thus these models differ from the Interdependence model which integrates nucleation and growth of equiaxed grains into a single model (Eq. (4)).

Dong and Lee [12] and then Badillo and Beckermann [9] used published numerical models to study the parameters that affect the CET, building on previous work by Wang and Beckermann [20]. In both studies, the nucleation of the seeds was found to be a function of the CS ahead of the columnar tips (as well as thermal gradient and the position of the seeds). These studies both found that "neglecting the solutal built-up at the interface leads to a significantly lower velocity being predicted for the onset of the CET" [12]. Without explicitly stating it, both these studies found it was important in columnar growth to include a Nucleation-Free Zone (NFZ) of the CS region ahead of the tip to produce accurate predictions.

Other recent work on CET using phase-field modelling was published by Montiel et al. [17]. The CET was being investigated as part of a study of the columnar and equiaxed microstructure formation in resistance spot welding of AZ31. In this case the seeds for nucleating equiaxed grains were placed randomly ahead of the growing columnar grains. The heterogeneous nucleation events on the seeds were based on local solutal, temperature and inoculant type conditions. The solute diffusion equations were solved for the subsequent equiaxed grain growth. Thus, although the role of solute undercooling in triggering nucleation was inherent in this model, this work does not discuss the role of CS on nucleation to any extent.

In this study, we will compare 2-D and 3-D numerical simulations to validate the formation of the NFZ as defined in the analytical Interdependence model.

## 2. The analytical Interdependence model

The analytical Interdependence model predicts the average grain size $d_{gs}$ as

$$d_{gs} = x_{cs} + x'_{dl} + x_{sd} \tag{1}$$

$d_{gs}$ is the predicted grain size and is defined by the sum of three distances as illustrated schematically in Fig. 1a. $x_{cs}$ is the amount of grain growth that results in sufficient CS,

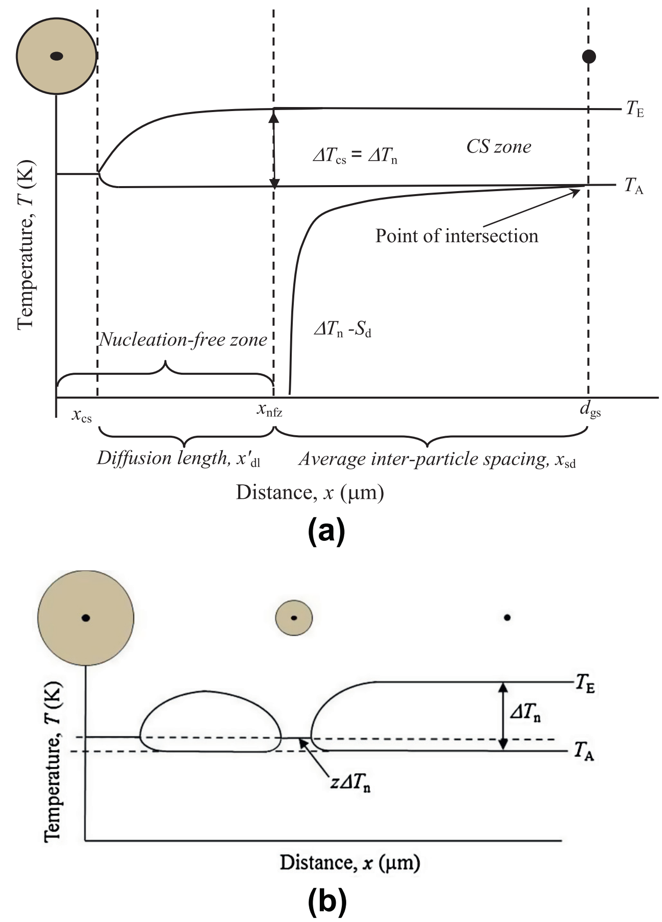


Fig. 1. (a) The Interdependence theory [6]: the presence of constitutional supercooling, CS, ahead of a single growing grain results in a nucleation free zone (NFZ), given as the sum of grain growth $x_{cs}$, and the length of the solute diffusion field $x'_{dl}$. The maximum value of $\Delta T_{cs}$ is achieved at $x_{nfz}$. The $\Delta T_n$–$S_d$ curve is superimposed on the plot which intersects the actual temperature, $T_A$, defining the location of the nucleation event (i.e. when $\Delta T_{cs} = \Delta T_n$). $\Delta T_n$ is the nucleation undercooling of the largest inoculant particle. (b) Extension of (a) to the case of NFZ formation between two grains. The accumulation of solute due to overlapping diffusion fields between two growing grains would result in a lower $\Delta T_{cs}$ value compared to the CS achieved ahead of a free growing grain (right-hand interface of the second grain). The lower CS suggests that the probability of nucleation occurring between the two grains would be low or negligible.

$\Delta T_{cs}$, to trigger a nucleation event on an adjacent potent nucleant with a nucleation undercooling of $\Delta T_n$. $x'_{dl}$ is the length of the solute diffusion zone that results in this critical value of $\Delta T_{cs}$. $x_{cs} + x'_{dl}$ constitutes the NFZ because the amount of CS in this zone is less than the maximum amount, $\Delta T_{cs}$, at the end of this zone. The distance from the end of NFZ to the next available potent nucleant is given by $x_{sd}$. $x_{sd}$ is the average distance between particles of the same size. It can be considered as the probability of finding a particle of a given size once $\Delta T_{cs}$ is equal to or greater than the particle's nucleation potency described by $\Delta T_n$. Fig. 1a shows the relationship between the developing CS zone and the distribution of $x_{sd}$ vs. particle size converted to nucleation undercooling (i.e. the $\Delta T_n$–$x_{sd}$ curve).