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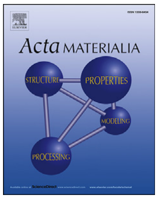

Full length article

A Nucleation Progenitor Function approach to polycrystalline equiaxed solidification modelling with application to a microgravity transparent alloy experiment observed *in-situ*

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ABSTRACT

A Nucleation Progenitor Function (NPF) approach that accounts for the interdependence between nucleation and growth during equiaxed solidification is proposed. An athermal nucleation density distribution, based on undercooling, is identified as a progenitor function. A Kolmogorov statistical approach is applied assuming continuous nucleation and growth conditions. The derived progeny functions describe the (supressed) distribution of actual nucleation events. The approach offers the significant advantage of generating progeny functions for volumetric (3D) data and projected image (2D) data. The main difference between 3D and 2D data in transparent alloy experiments is due to a stereological correction for over-projection. Progeny functions can be analysed to obtain statistical output information, e.g., nucleation counts, average nucleation undercooling and standard deviation. The statistical output data may be calculated in a formative (running) or a summative (final) mode. The NPF kinetics have been incorporated into a transient thermal model of equiaxed solidification. The model has been applied to characterise a microgravity solidification experiment with the transparent alloy system Neopentylgycol-30 wt%(d)Camphor. The model predicted thermal and observed nucleation and growth data with a good level of agreement.

1. Introduction

Polycrystalline equiaxed solidification, leading to grain refinement in microstructure, is a significant topic for the metal processing industry. The advantages of fine-grained equiaxed structures in casting processes are well-known and include a reduction in defects and improvements in component strength. In wrought alloy processing, initial fine grain structures can improve recrystallization kinetics. A relevant industry standard [1], which outlines test procedures for grain-refined microstructure characterisation, exists. However, the standard does not attempt to predict cause-and-effect relationships in a detailed way. Nevertheless, much is known about the heterogeneous nucleation mechanisms that promote polycrystalline equiaxed grain structures. Such methods include inoculation of aluminium castings with TiB_2 particles.

1.1. *Athermal nucleation distribution based on undercooling*

Recently, Greer [2] and Easton et al. [3] reviewed grain refinement in castings. Indeed, Greer and co-workers [4] provided the much-needed physical explanation of how free-growth conditions are established on the inoculating grain refiner particles. The attainment of free-growth conditions on the particles was identified as the controlling factor in the grain refinement processes. The undercooling for free-growth conditions, ΔT_{fg}, on any particle is related to the particle dimension, d:

$$\Delta T_{fg} = \frac{4\gamma}{\Delta S_V d}, \tag{1}$$

Where γ is the solid-liquid interfacial energy and ΔS_V is the volumetric entropy of fusion. Applications of athermal nucleation

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distributions based on undercooling, originally proposed by Oldfield [5], have been commonplace in microstructure modelling. As reviewed in Rappaz [6] and more recently in Rappaz and Dantzig [7], nucleation distributions are often assumed to follow standard statistical probability density functions, such as Gaussian or log-normal. Quested and Greer [8] showed that the particle size distributions could be described using a standard statistical distribution (e.g., log-normal); therefore, after application of eq. (1), the suitability of athermal nucleation distributions based on undercooling has been clarified within the literature.

1.2. *Predictive modelling of equiaxed solidification*

A significant number of equiaxed solidification models have been developed to predict microstructure information at the macro scale subject to definitive processing conditions. Maxwell and Hellawell [9] were the first to provide a simplified model. They used Arrhenius-type nucleation kinetics and globular-to-dendritic growth kinetics to predict the suppression of nucleation rate due to latent heat release. They ignored any correction for impingement, which they justified because of low solid fraction.

Several modelling approaches have assumed free-growth nucleation conditions and athermal nucleation distributions [10–12]. Quested and Greer [10] developed an equiaxed solidification model based on an athermal nucleation distribution with both spherical and dendritic morphologies included in the analysis for comparison purposes. They used a standard Johnson-Mehl-Avrami-Kolmogorov correction for the impingement of grains, which caused the deactivation of seed particles in the latter stages of solidification due to the process of engulfment by previously nucleated grains.

Shu et al. [11] developed an equiaxed model that included the nucleation-suppressing effects of solute rejection into the untransformed liquid phase. This model introduced a Solute Suppressed Nucleation (SSN) zone around each spherical envelope of dendritic mush. The effect of including the SSN zone was analysed and comparisons were made to experimental case studies. Plots of average grain size versus growth restriction factor were provided.

StJohn et al. [12] described the interdependence theory in which they examined the relationship between nucleation and growth. Their model included a nucleation free zone where nucleation is prohibited. The average radii of the spherical mushy envelopes plus the length of the solute affected zone around each envelope defines the nucleation free zone. The interdependence theory was used to predict average grain size and plots showed grain size as a function of the reciprocal of growth restriction factor.

Du and Li [13] extended an established precipitation model, the Kampmann-Wagner model, to include an SSN zone and multi-component growth kinetics by incorporating a CALPHAD approach. The model was applied to both uniform temperature-fixed cooling and direct casting scenarios.

1.3. *In-situ experimental monitoring of equiaxed nucleation and growth*

Traditionally, in the case of metal alloys, experimental characterisation is performed *ex-situ*, also known as post-mortem analysis. In *ex-situ* analysis, metal samples are prepared by cutting, polishing, and etching along recommended guidelines so that grain structures are revealed.

Alternatively, *in-situ* experimental observation provides information on solidification processes in real time. Metals are opaque at visible wavelengths, hence *in-situ* analysis of metals has relied on the application of real-time x-ray radiography techniques. Several dedicated *in-situ* experimental apparatuses have been developed to investigate equiaxed solidification [14–17].

Recently, Xu et al. [18] applied their model of equiaxed solidification from Ref. [13] to an *in-situ* x-ray experiment involving Al-10 wt%Cu alloy inoculated with Al-5wt.%Ti-1wt.%B grain refiner master alloy. They investigated nucleation and growth conditions at different cooling rates. They showed predictions of final grain size versus cooling rate. Their results compared favourably with experimental observations in the case of well-refined alloy systems.

The analysis of transparent alloy systems using optical methods pre-dates real-time x-ray radiography by several decades. Jackson and Hunt [19] were the first to propose transparent alloy systems as being analogous model systems of metallic solidification. Indeed, *in-situ* optical analysis of transparent alloys has been used extensively to investigate time-dependent solidification behaviour in cellular [20], dendritic [21], and eutectic [22] systems. Transparent alloy *in-situ* observation has provided insights previously unobtainable through *ex-situ* characterisation. For example, dendrite arm fragmentation was first observed in a transparent alloy system [23] and later confirmed in metallic alloys by application of *in-situ* x-ray radiography [24]. However, there are several distinctions between transparent alloy and metal alloy *in-situ* processing worth pointing out. Firstly, transparent alloy systems typically have much lower melting temperatures than most metal alloys. Secondly, suitable metal alloy systems for x-ray observation require sufficient density difference between solute and solvent to provide improved x-ray absorption and image contrast between the phases. Thirdly, *in-situ* metal samples are prepared in thin foil format (typically some 100 μm in thickness), whereas, transparent alloy samples are typically thick samples to avoid boundary effects. This final point raises the issue of stereology effects on the image data from bulky samples. These stereology effects will be investigated in this manuscript.

1.4. *The multiple equiaxed dendrite interaction (MEDI) experiment*

In 2015, an *in-situ* transparent alloy experiment into polycrystalline equiaxed solidification was conducted under microgravity conditions (reported in Ref. [25]). The MEDI experiment was launched on-board the MASER-13 sounding rocket. The sample contained the transparent material, Neopentylgycol-30 wt%(d) Camphor, which is a hypoeutectic alloy with face-centred cubic lattice structure in the primary dendritic phase. The sequence of events during the launch led to in-situ observation of polycrystalline equiaxed nucleation and growth under high-quality microgravity conditions. A controller allowed fixed cooling conditions to be set under a low temperature gradient. Three thermocouples recorded temperatures within the sample. Observations of equiaxed dendritic solidification (nucleation, growth, and interaction) were made *in-situ* and in real-time using optical magnification methods at both the macro and micro length scales. This MEDI campaign was conducted as part of the European space agency CETSOL (Columnar-to-Equiaxed Transition in SOLidification Processing) programme [25].

1.5. *Aims and objectives*

This manuscript proposes a Nucleation Progenitor Function (NPF) modelling approach for polycrystalline equiaxed solidification. The starting position for this approach is to define an athermal nucleation distribution as a progenitor function. Through application of the approach, different instances of continuous nucleation outputs (progeny functions) arise. Hence, an aim of this study is to define progenitor-progeny relationships. These progeny functions are then processed to give detailed information, e.g., average nucleation undercooling, standard deviation of nucleation