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Benchmark experiments and numerical modelling of the columnar-equiaxed dendritic growth in the transparent alloy Neopentylglycol-(d)Camphor

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Abstract. Solidification benchmark experiments on columnar and equiaxed dendritic growth, as well as the columnar-equiaxed transition have been carried out under diffusion-dominated conditions for heat and mass transfer in a low-gravity environment. The system under investigation is the transparent organic alloy system Neopentylglycol-37.5wt.-%(d)Camphor, processed aboard a TEXUS sounding rocket flight. Solidifications was observed by standard optical methods in addition to measurements of the thermal fields within the sheet like experimental cells of 1 mm thickness. The dendrite tip kinetic, primary dendrite arm spacing, temporal and spatial temperature evolution, columnar tip velocity and the critical parameters at the CET have been analysed. Here we focus on a detailed comparison of the experiment “TRACE” with a 5-phase volume averaging model to validate the numerical model and to give insight into the corresponding physical mechanisms and parameters leading to CET. The results are discussed in terms of sensitivity versus numerical parameters.

1. Introduction

Support of advanced castings by process simulations is of interest to understand structure formation and to enhance product quality, as well as to minimize material and energy consumption. The as-cast structure (grain-structure, dendrite morphology, element segregation and other casting defects) of the product is linked to mechanical properties, although some features of the structure can be further improved by subsequent processes like heat-treatment or hot isostatic pressing. For a given alloy, the as-cast structure is a result of the competition between nucleation and growth of the different phases emerging subsequently during cooling towards ambient conditions. In technical multi-component alloys the primary solid phase formed from the melt often exhibits a dendritic columnar or equiaxed grain structure. The type of grain structure (columnar, equiaxed or mixed) is dependent on the alloy properties, experimental conditions like thermal gradient and solidification advancement, melt flow driven by uneven density field of the melt or by volume shrinkage during solidification, sedimentation



and buoyancy of solid or mushy particles in the melt and finally the probability to form and activate nuclei or other sources for subsequent equiaxed dendrite growth in the melt [1].

The complex interaction between time-dependent nucleation and solidification conditions on one hand and time-dependent convective melt flow can be reduced by low-gravity experiments, in which convective flow, sedimentation and buoyancy are minimized or completely suppressed. Accordingly, the investigation of dendrite growth, grain formation and the Columnar-to Equiaxed Transition (CET) under diffusive conditions for heat and mass transport can be simplified significantly. Comparative reference experiments on ground may then shed light on the gravity effects and help to properly validate casting simulation tools.

Columnar growth is mandatory in single crystal growth and also used for casting of some turbine blades, where anisotropic properties of the product improve for example creep resistance at elevated service temperature [2]. Polycrystalline equiaxed growth is intended for more isotropic properties or smaller grain sizes. The CET in grain structure is generally not desired in the final product, since it may induce a change of material properties, but its investigation helps identifying suitable processing conditions for columnar and equiaxed growth.

Here we recall the main experimental results from the low-gravity experiment “TRACE” (TRANSPARENT Alloys in COLUMNAR and EQUIAXED Solidification) [3-5], executed on the sounding rocket mission TEXUS-47 in 2009 with about 7 minutes of low-gravity time. The experiments were focusing on dendrite growth and the CET in the binary hypoeutectic transparent organic model system Neopentylglycol (NPG)-37.5 wt.-% (d)Camphor (DC). Allowing for in-situ and real-time observation of the solidification phenomena by standard optical methods, a detailed comparison of this benchmark experiment to numerical models was intended.

A 5-phase volume averaging model [6,7] was used to calculate the columnar tip front propagation, front undercooling, appearance of equiaxed dendrite crystals, CET, etc. The numerical results will be evaluated by comparison with the experiments. For the uncertainty of some physical properties or process parameters, e.g. the dendrite selection constant σ^* and the maximum nucleation density N_0 , a numerical parameter study is performed to analyze the influence of them on the solidification result. Actually an optimal agreement can be achieved by adjusting those parameters.

2. Experimental section

The experiment has been described in detail in [3-5]. Figure 1 shows a schematic drawing of the main parts of the experimental set-up. The evacuated experimental volume of 20 mm width, 16 mm height and 1 mm thickness was filled with the molten alloy NPG-37.5 wt.-% DC. The phase-diagram of the alloy system is given in [8]. In contrast to earlier publications [3, 4], the liquidus-temperature T_L of the alloy is directly taken from the CALPHAD-description of the calorimetric data [8] and given as 67.1°C. This seemingly minor increase by 3.1°C (when compared to the former value of 64.0°C) has impact on the calculation of growth and nucleation undercooling related to T_L . The experiment is controlled by a peltier cooler and a resistance heater, both were close to but not directly in contact with the bottom and the top of the experimental cell, respectively. Optical observation is carried out perpendicular to the height and width-axes of the cell. Macro-images of cell-size are acquired, as well as detail images with roughly ten times smaller field-of-view with two different CCD-cameras and corresponding optical resolutions. Polycrystalline columnar dendritic growth was started on ground well before lift-off of the sounding rocket with a cooling-rate of $-1/300 \text{ Ks}^{-1}$ applied to cooler and heater. After steady-state columnar growth, the CET was achieved in transient regime by setting the cooling-rates simultaneously to $-1/30 \text{ Ks}^{-1}$ at the beginning of the low-gravity period; 60 s after lift-off of the sounding rocket. A gravity level lower than $\pm 4 \text{ mg}$ in all directions was obtained within the low-gravity period of 410 s from $t=60 \text{ s}$ to $t=470 \text{ s}$. The experimental sequence for the cooler and heater temperatures is indicated in figure 2 with the corresponding gravity levels. The sequence is simplified in the graph and in the simulations for the first 200 s, where in contrast in the experiment an inverted temperature field is active for thermal mixing (figure 6). Times are normalized to the numerical simulation configuration (sec. 3), where the end of the microgravity period is located at time $t=6000 \text{ s}$.

The effect of change of gravity levels in figure 2 is ignored in the simulations here, zero-g is used throughout the complete simulation time. This is justified by the relatively strong viscous damping of fluid flow in the thin sheet of 1mm thickness.

Temperatures were measured within the bulk material by means of five thermocouples type K of diameter 0.25 mm, being inserted 2.5 mm through one side of the cell. From image-processing of the macro-images and the thermocouple data the position, velocity and temperature of the columnar and later equiaxed dendritic interface were measured. After heterogeneous nucleation of equiaxed dendrites on unknown substrates their density and nucleation temperatures were determined as a function of time. Volume fractions of liquid, equiaxed and columnar phases were determined at the interface. Main results are published elsewhere [2, 3], here we focus on the determination of nucleation parameters and the dendrite selection constant σ^* , as input or comparison to the numerical modelling. Detail images could not resolve the dendrite tip radii with sufficient accuracy, but were used to estimate the morphological factors for columnar and equiaxed dendrites in numerical simulations (Tab. 2).

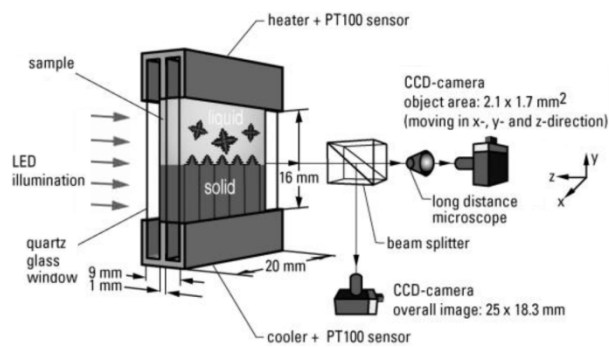


Figure 1: Schematic drawing of the main parts of the experimental setup.

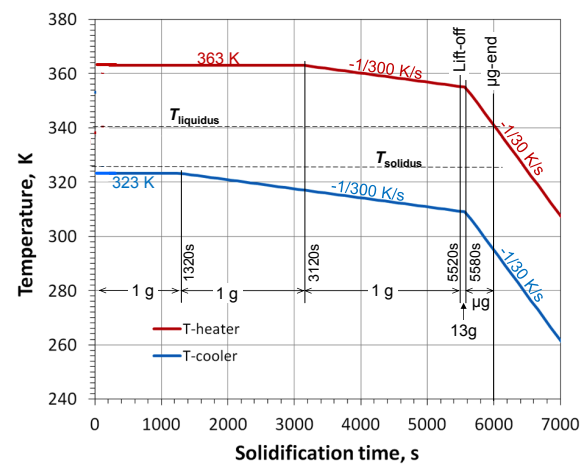


Figure 2: Imposed temperatures at the heater and cooler in simulations, similar to exp.

As shown for example by Mangelinck-Noël et al. [9] by x-ray in-situ observations of equiaxed dendrite growth in Al-Ni alloys, a series of experiments at different cooling rates can be used to estimate their nucleation parameters, assuming specific probability functions for the nucleation temperatures. Here, we obtain the nucleation temperature distribution experimentally from a single transient experiment by increasing the cooling rates and measuring nucleation positions and interpolating thermocouple temperatures. The resulting distribution is given in [3], relative to the previously used value of $T_L=64.0^\circ\text{C}$. The nucleation densities of active particles are calculated from the number of nucleated equiaxed dendrites within the given temperature interval and the average volume (width x depth x height from columnar front to equiaxed dendrite at highest position) with nucleation events. For comparison and input to numerical simulations, we assume a Gaussian probability function f for the nucleation undercooling ΔT :

$$f(\Delta T) = \frac{N_0}{\Delta T_\sigma \sqrt{2\pi}} e^{-\frac{1}{2} \left(\frac{\Delta T - \Delta T_N}{\Delta T_\sigma} \right)^2}, \quad (1)$$

with the maximum nucleation density N_0 , the mean nucleation temperature ΔT_N and the standard deviation of the distribution ΔT_σ . By referencing the nucleation temperatures to $T_L=67.1^\circ\text{C}$, the Gaussian fit (1) of the nucleation density can be plotted, see figure 3.

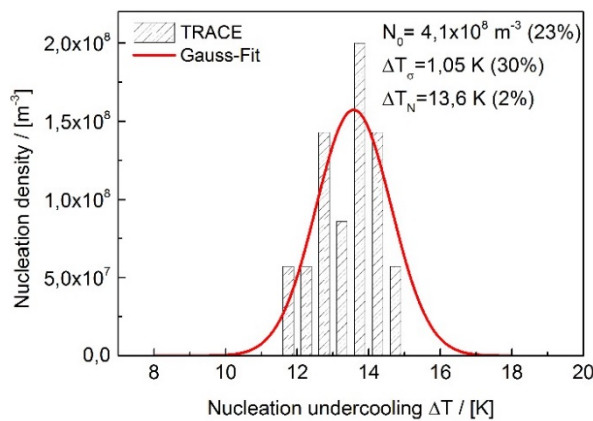


Figure 3: Nucleation density distribution and fitted Gaussian probability function. Number in brackets give the relative fit-errors.

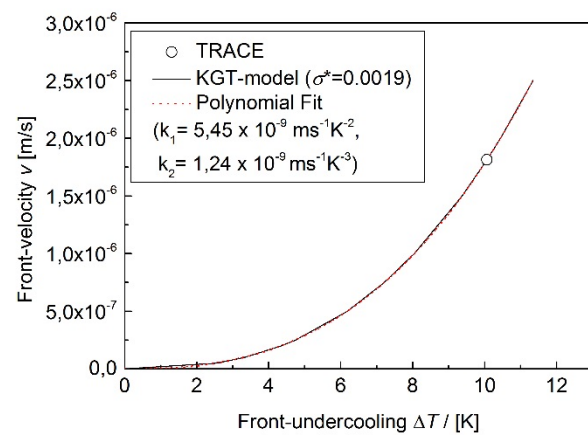


Figure 4: Kinetic law for columnar growth from experiment (single data point), the KGT-model [10] fit and a polynomial fit according to (2).

Furthermore, in numerical simulations a polynomial fit for the kinetic law in columnar growth

$$v_{tip,columnar}(\Delta T) = k_1 \Delta T^2 + k_2 \Delta T^3 \quad (2)$$

to the KGT [10] model is used. Figure 4 shows the steady-state columnar growth velocity and undercooling, experimentally determined before lift-off at the end of the low cooling-rate phase with $-1/300 \text{ Ks}^{-1}$, as well as the fit to the KGT-model (with the selection constant σ^* as fit-parameter and other parameters given in Tab. 1) and the polynomial fit, according to (2). Due to these estimates from experimental results given above all simulations are carried out using $\Delta T_N = 13.6 \text{ K}$, $\Delta T_\sigma = 1.05 \text{ K}$, $k_1 = 5.45 \times 10^{-9} \text{ ms}^{-1} \text{ K}^{-2}$ and $k_2 = 1.24 \times 10^{-9} \text{ ms}^{-1} \text{ K}^{-3}$.

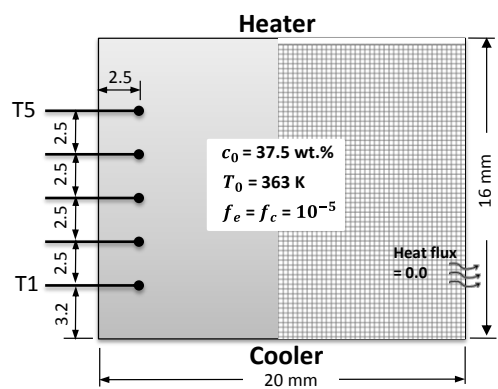
Three comparative simulations have been carried out; as reference case-1 the maximum nucleation density $N_0 = 4.1 \times 10^8 \text{ m}^{-3}$ obtained from the fit and a significantly lowered value of $\sigma^* = 0.0076$ (when compared to the standard value used in the LGK-model [11] $\sigma^* = 1/4/\pi^2 = 0.0253$) are used. The value of $\sigma^* = 0.0019$ fitted to experimental data (figure 4) could not be adapted numerically yet for the kinetic law of equiaxed dendrites (LGK), nevertheless the effect of increased dendrite selection constant (to $\sigma^* = 0.0253$) is investigated in case-2. Finally, the effect of an increased $N_0 = 10.0 \times 10^8 \text{ m}^{-3}$ is investigated in case-3. This is due to the fact, that the fitted value for N_0 is related to an increase in the cooling rate to $-1/30 \text{ Ks}^{-1}$ and a probably higher value for N_0 could be expected, when higher cooling rates would be applied. Moreover the average volume was used in the calculation of the nucleation density, leading to some errors and a value of $N_0 = 5.0 (\pm 1.0) \times 10^8 \text{ m}^{-3}$ was found experimentally as the maximum nucleation density in time in the available low-gravity period [3]. A summary of the three simulation cases with their nucleation and kinetic, as well as morphological parameters and thermo-physical properties of the alloy are given in Tab.1 and Tab.2.

Model features

A 5-phase mixed columnar-equiaxed solidification model with dendritic morphology was described previously [7]. The growth of the crystals is presented by the expansion of the grain envelope, whose profile connects the primary, secondary or tertiary dendrite tips to form a ‘natural’ enclosure of the equiaxed grains or columnar trunks. The growth of the grain envelopes and the solidification of the interdendritic melt, i.e. the liquid-to-solid phase transition, are treated differently. The growth of the envelopes is determined by dendrite growth kinetics. The semi-empirical Kurz-Giovanola-Trivedi (KGT, [10]) model is fitted by the polynomial given in equation (2) and is used for the growth of columnar primary dendrite tips. The Lipton-Glicksman-Kurz (LGK, [11]) model is applied for the growth of columnar secondary dendrite tips and equiaxed primary dendrite tips, based on different

values of σ^* , as given above. The solidification occurs inside the grain envelope, which is driven by the supersaturation of the interdendritic melt and governed by the diffusion in the interdendritic melt region. The model comprises actually only 3 ‘hydrodynamic’ phases: the liquid melt, the equiaxed crystals, and the columnar grains, denoted as l -, e - and c -phases and have corresponding volume fractions; f_l , f_e , and f_c . However, two additional phase regions exist within each of the equiaxed and the columnar crystal envelopes: the solid dendrites and interdendritic melt inside crystal envelopes. Consequently, the system encompasses 5 ‘thermodynamic’ phases: the solid equiaxed dendrite and interdendritic melt within the equiaxed grain envelope, the solid columnar dendrite and interdendritic melt within the columnar crystal envelope, and the extradendritic melt. Interaction, i.e. competition, between columnar and equiaxed growths, and both hard blocking [12] and soft blocking [13] mechanisms for the columnar-to-equiaxed transition (CET) are considered. The origin of the equiaxed crystals is modelled by a heterogeneous nucleation law, equation (1), with three fit parameters taken from experiment. The maximum equiaxed grain nucleation density is either taken from the fitted experimental data (case-1&case-2) or from an assumed data (case-3) to investigate its effect.

The solidification cell filled with NPG-37.5wt.-% DC is simplified to a 2D grid with volume element size of $0.25 \times 0.25 \text{ mm}^2$ as shown in figure 5. The grid is provided by 5 measuring points (thermocouples) to record the cooling curves analogous to experiment. Similar to experiment, a temperature gradient is applied to the simulation grid by superimposing the experimental temperature control at the cooler and heater (figure 2) on the corresponding grid boundaries, with a heat transfer coefficient of $50 \text{ W m}^{-1} \text{ K}^{-1}$. The conservation equations of mass, enthalpy, species, and number density are solved sequentially at each time using CFD software package, ANSYS-Fluent version 14.5.0 based on the control volume method. A time step size of 0.2 s was used to achieve solution convergence over a time of $\sim 7000 \text{ s}$.



Heat transfer to cooler/heater: $50 \text{ W m}^{-1} \text{ K}^{-1}$

Figure 5: 2D grid of the solidification cell with boundary and initial conditions.

Table 1. Thermo-physical alloy properties.

Alloy conc. [wt.-% DC]:	$c_0 = 37.5$
Eutectic conc. [wt.-% DC]:	$c_E = 45.3$ [8]
Liquidus-temp. [$^{\circ}\text{C}$]:	$T_{\text{liq.}} = 67.1^{\circ}\text{C}$ (340.25K) [8]
Solidus-temp. [$^{\circ}\text{C}$]:	$T_{\text{sol.}} = 52.8^{\circ}\text{C}$ (325.95K) [8]
Liquidus slope [K/wt.-%]:	$m_{\text{liq.}} = -1.84$ [8]
Partition coef. [-]:	$k = 0.085$ [8]
Diffusion coef. [m^2/s]:	$D_L = 9.7 \times 10^{-11}$ [14] $D_S = 8.0 \times 10^{-20}$ [*]
Thermal capacity [J/kg·K]:	$c_{p,L} = 2400$, $c_{p,S} = 2650$ [8]
Thermal cond. [W/m·K]:	$\lambda_L = 0.125$, $\lambda_S = 0.265$ [16]
Density [kg/m^3]:	$\rho = 1035$ [5]
Latent heat fusion [kJ/kg]:	$L = 33.4$ [8]
Gibbs-Thomson coef. [Km]:	$\Gamma = 7.8 \times 10^{-8}$, adapted from [15]
Selection constant [-]:	$\sigma^* = 0.0019$ (figure 4)
(*) assumed	

Table 2. Nucleation, kinetic and morphological parameters for modeling.

Nucleation parameters:	$\Delta T_N = 13.6 \text{ K}$, $\Delta T_{\sigma} = 1.05 \text{ K}$ (figure 3), N_0 (see below)
Kinetic parameters columnar:	$k_1 = 5.45 \times 10^{-9} \text{ ms}^{-1} \text{ K}^{-2}$, $k_2 = 1.24 \times 10^{-9} \text{ ms}^{-1} \text{ K}^{-3}$ (figure 4)
Shape factor / circularity col.:	0.798 / 0.50 (estimated from experiment)
PDAS / SDAS columnar:	400 μm / 50 μm (estimated from experiment)
Shape factor / sphericity equiaxed.:	0.58 / 0.84 (estimated from experiment)
Case-1:	$\sigma^* = 0.0076$, $N_0 = 4.1 \times 10^8 \text{ m}^{-3}$
Case-2:	$\sigma^* = 0.0253$, $N_0 = 4.1 \times 10^8 \text{ m}^{-3}$
Case-3:	$\sigma^* = 0.0076$, $N_0 = 10.0 \times 10^8 \text{ m}^{-3}$

3. Results and discussion

Figure 6 shows the control temperatures T_{heater} and T_{cooler} , the temperature evolution at the thermocouples T1-T5 as measured experimentally and the corresponding temperature data from simulation in case-1. As discussed before, the first 200 s in simulations are simplified and the initial isothermal conditions of 363K are applied to the bulk material. The cooling-rates in columnar growth ($t < \mu\text{g-start}$) are comparable to the experimental data, although a shift in temperatures from T1 to T5 is observable. Furthermore the application of the higher cooling-rate ($t > \mu\text{g-start}$) shows a slower reaction in simulations. Both effects can be explained by non-optimized heat-transfer coefficients at cooler and heater. Figure 7 shows the front position (columnar for $t < \mu\text{g-start}$, transient to equiaxed for $t > \mu\text{g-start}$). As all simulations use identical kinetic parameters k_1 and k_2 for columnar growth, the results are identical in that regime. The corresponding constant shift and delayed response is due to the effects explained in the cooling-curves (figure 6).

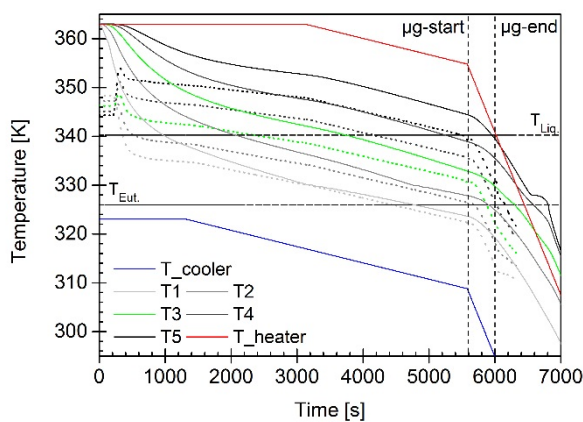


Figure 6. Temperature evolution for case-1. Solid lines (sim.), dotted lines (exp.).

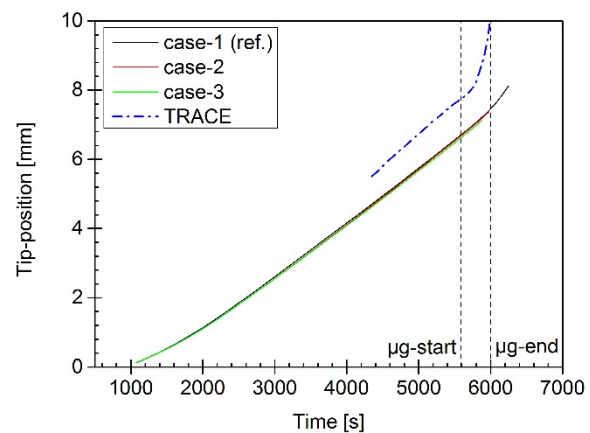


Figure 7. Front-position for cases 1-3 in comparison to "TRACE".

As can be seen in figure 8, the cases-2&3 show an increasing and considerable amount of equiaxed volume fraction towards the CET at positions 7.4 mm (case-2) and 7.6 mm (case-3). Accordingly the front undercooling is lowered in these cases in figure 9. Experimentally a very small amount of equiaxed volume fractions is found before the increase of the cooling-rate (~ 5580 s), similar to case-1. The agreement between the case-1 and the experiment is satisfactory.

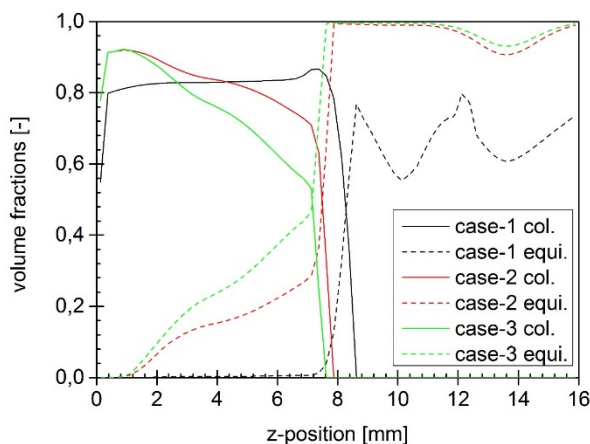


Figure 8. Volume fractions (here only columnar and equiaxed) for cases 1-3.

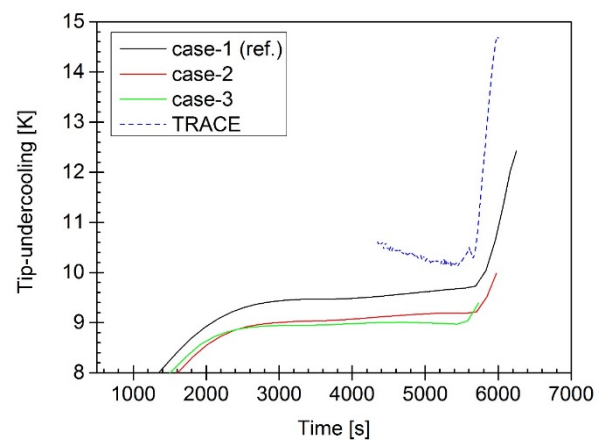


Figure 9. Front undercooling for cases 1-3 in comparison to experimental data.

Furthermore an oscillation in equiaxed volume fraction is achieved in case-1. Interestingly, such behaviour is also found in the experiment “TRACE+” with NPG-20.0wt.-%DC, flown on TEXUS-49 with the CET not fully achieved [17].

The evolution of the columnar and equiaxed volume fractions in time for case-1 are shown in figures 10&11 in log-scale. CET is achieved at $t=6270$ s and position $z=8.4$ mm. The significant increase in the equiaxed volume fraction and thus finally the CET is observable between $t=5520$ s and $t=6270$ s in the simulation. Experimentally we find a similar situation, showing columnar growth at $t=5480$ s in figure 12 and the CET happened at $t=5990$ s in figure 13. The experimental position of CET is at $z=9.5$ mm.

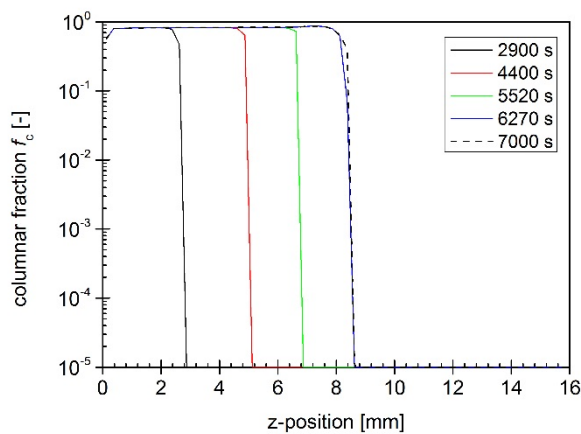


Figure 10. Evolution of columnar volume fraction in time for simulation case-1.

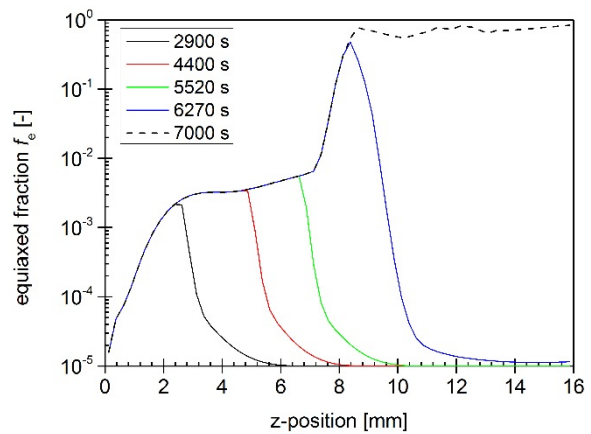


Figure 11. Evolution of equiaxed volume fraction in time for simulation case-1.



Figure 12. Macro-image of columnar solidification in “TRACE” at $t=5480$ s (50 s before rocket lift-off, 110 s before increase of cooling-rate=start of μg -time). The lower dark front corresponds to the eutectic temperature.



Figure 13. Macro-image of columnar-equiaxed solidification in “TRACE” at $t=5990$ s (10 s before end of μg -time). The lower dark front corresponds to the eutectic temperature.

4. Summary

Results from the low-gravity experiment “TRACE” investigating columnar and equiaxed dendritic solidification in the transparent alloy NPG-37.5wt.-%DC were compared to predictions from a 5-phase-volume averaging model. By adapting the columnar growth kinetics, morphological parameters, thermo-physical properties and nucleation parameters from experimental analysis a fairly good agreement was achieved for front undercooling and the position of CET in time and space. Further improvement is expected by adjusting heat transfer coefficients and the dendrite selection constant σ^* . For the later a four times higher value than determined experimentally was used and the CET-position was found to be at 8.4 mm (9.5 mm in experiment). The results from simulation case-2 show a later CET in space and time and a higher undercooling, when decreasing σ^* from the standard KGT/LGK-value of 0.0253 to 0.0076. A similar effect is observed when decreasing the maximum nucleation density (case-3).

Acknowledgement

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