Columnar and equiaxed solidification in a microgravity environment – The CETSOL project

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Abstract

The formation of the microstructure and the grain structure during solidification is affected by buoyancy-driven flow and movement of crystals growing in the melt. To allow for pure diffusive solidification conditions, i.e., suppressing melt flow and sedimentation and floatation effects, experiments in microgravity environment were performed in the frame of the ESA Map programme CETSOL (Columnar-to-Equiaxed Transition in SOLidification Processing). This contribution presents an overview of results obtained so far, focussing on 2 topics: First, to investigate the columnar-to-equiaxed transition (CET), refined and non-refined Al-7wt%Si alloys were solidified with different process parameters on-board the International Space Station ISS. From extensive analysis of the samples the microstructures and the grain structures were determined as well as critical parameters for CET. These results are compared to results obtained with different numerical models showing a fairly good agreement. Second, growth of equiaxed dendrites was investigated in-situ using the transparent model alloy Neopentylglycol-(d)Camphor in microgravity environment during the sounding rocket mission MASER-13. The growth of individual dendrites was directly observed with CCD cameras, providing information of the 3D shape of dendrites and of the dendrite morphology evolution.

Keywords: solidification, columnar-to-equiaxed transition, equiaxed dendrite, microgravity

1. Introduction

To allow for pure diffusive solidification condition with suppressed melt flow and without sedimentation or floatation effects, experiments in microgravity environment were performed, which provide unique data for testing fundamental theories of grain and microstructure formation.

Such activities are the task of the research project Columnar-to-Equiaxed Transition in SOLidification Processing (CETSOL) in the framework of the Microgravity Application Promotion (MAP) programme of the European Space Agency (ESA). At present, the CETSOL team consists of seven European scientific partners, three partners from the USA and seven partners from industry (see list of co-authors). This paper summarizes some main results achieved in the frame of the CETSOL project.

2. Microgravity experiments with Al-7wt%Si alloys

In the Materials Science Laboratory (MSL) on-board the International Space Station (ISS) several experiments in microgravity were carried out successfully to investigate especially the columnar-to-equiaxed transition under diffusive conditions for heat and mass transport. Within a Batch1 and a Batch2a six solidification experiments were performed in the Low Gradient Furnace (LGF) module and seven experiments in the Solidification and Quenching Furnace (SQF) module, respectively [1-5]. Rod-like samples made of Al-7wt%Si alloys with and without grain refiner particles were used. A main difference in processing are initial axial temperature gradients in the sample of about 1K/mm and 3-4K/mm in LGF and SQF, respectively. Columnar-to-equiaxed transition is triggered either by a sudden increase of the solidification velocity or by a continuous decrease of the temperature gradient.

As an example, results of experiment B2-FM1 (from Batch2a) analysis are given [6]. The recorded cooling curves, measured by thermocouples TC5 to TC12 along the sample axis, are shown in Figure 1. The solidification stage I with v_1=20µm/s for z_1=20mm is expected to generate a columnar dendritic growth. A transition to equiaxed growth should be triggered in the solidification stage II by increasing the furnace movement to v_2=200µm/s and by applying a cooling rate of dT/dt=-8K/min at the heaters of the furnace. Finally, sample quenching is applied in stage III. The average liquidus isotherm velocities and the temperature gradients ahead of the liquidus isotherm were deduced as function of time (Figure 2).

From longitudinal cross-section, the microstructure and the grain structure in the refined Al-7wt%Si sample were identified. Columnar structures are obtained for stage I, followed by equiaxed and finer dendrite grains in stage II. Quantitative evaluation of the microstructure showed the development of eutectic percentage as well as the dendrite arm spacing (DAS) along the sample axis (Figure 3). Due to the finer dendritic structure and the absence of large eutectic regions, the eutectic proportion is lower in stage II.

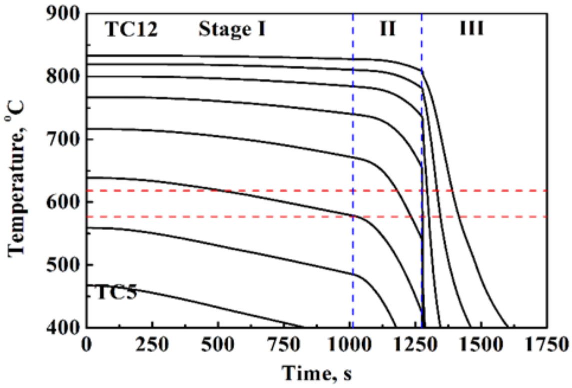

Figure 1: Measured temperature evolutions at thermocouple positions TC5 to TC12 for B2-FM1.

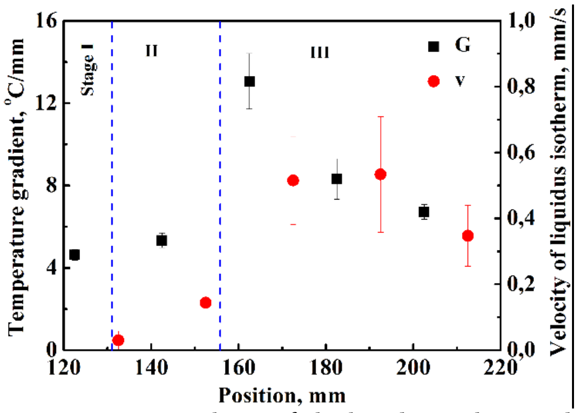

Figure 2: Average velocity of the liquidus isotherm and temperature gradient ahead of the liquidus isotherm.

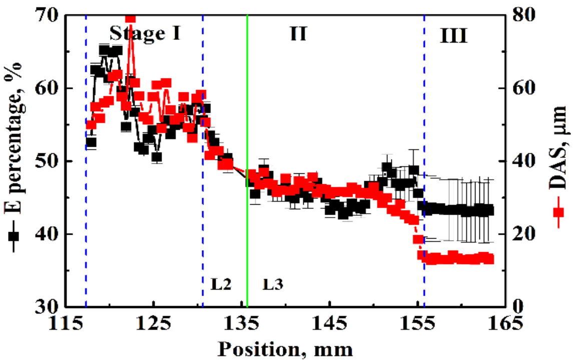

Figure 3: Mean eutectic percentage and DAS vs. position along the longitudinal direction of the sample

For determination of the CET position, the grain elongation factor (EF) and the equivalent diameter (ED) were evaluated quantitatively using EBSD measurements. The grain contour maps are constructed and displayed in Figure 4 (top). In these maps, each grain consists of a number of indexed pixels that share close crystallographic orientations. The average EF and ED of the grains along the sample axis are also given in Figure 4. The ED decreases from stage I to stage II, while the EF stays close to but above 2 after CET_{min} at z=134mm up to z=156 mm. Thus, equiaxed grains are elongated defining a progressive CET. It should be noted that a deviation of the elongation factor is observed at about z=148 mm, which is due to the existence of a side porosity [6].

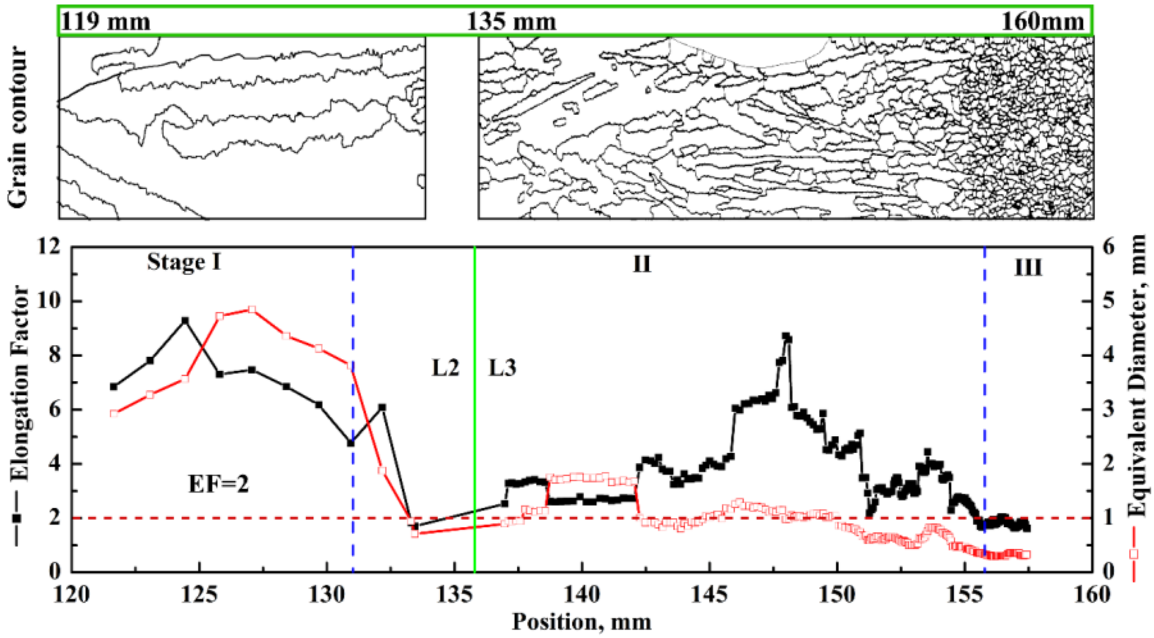

Figure 4: Grain contours revealed by EBSD maps for longitudinal cross-sections L2 to L3 (top) and longitudinal profiles of averaged EF and ED (bottom).

The evaluation of the microgravity experiments so far shows that in non-refined alloy no CET was observed, but columnar dendritic growth with some fragmentation of dendrite structures. CET was detected only for refined alloys. In case of the sudden increase of the solidification velocity a sharp CET within a few mm distance was observed, whereas in case of a decrease in temperature gradient a progressive CET mode exists.

The unique experimental data was used for numerical modelling of CET by three different methods:

(i) A columnar front tracking model [7, 8] was adapted to include an equiaxed growth model (based on JMAK theory) to predict columnar dendrite tip undercooling, growth rate, and the temperature gradient with respect time; the model allows for sharp or progressive CET prediction. Good agreement was found between the simulated and experimentally measured CET position.

(ii) A dendritic needle network (DNN) model [9] was applied to simulate CET in flight samples. Already in a thin sample approach of the DNN model both sharp and progressive CET in refined Al-7wt%Si alloy could be reproduced. For further improvement this effective modelling will be extended in 3D.

(iii) Using a CAFE model both segregation and grain structures, as well as CET, were numerically modelled in 3D. The CET transition mode, be it sharp or progressive, is retrieved. Distributions of the grain EF and the ED are fairly reproduced. Moreover, the CET positions are predicted accurately and precisely [8]. At present a multiscale modeling of dendritic growth is under development [10].

3. Microgravity experiments with NPG-DC alloys

Within the CETSOL project the experiment MEDI (Multiple Equiaxed Dendrite Interaction) was performed on the sounding rocket mission MASER-13 [11]. It focuses on equiaxed dendrite nucleation, growth and interaction via solutal fields in the transparent organic model alloy Neopentylglycol-(d)Camphor (NPG-DC). The experimental volume has dimensions of height of about 10 mm, depth of 3 mm and width of 13 mm. Starting in a temperature gradient of G=0.3 K/mm between hot and cold side, a cooling-rate of -0.75 K/min was applied.

With the chosen experimental parameters, an equiaxed dendritic front occurs by successive nucleation and growth, moving from bottom to top. In-situ and real-time observation of this process is performed by two different light-optical systems, providing overview digital image (Figure 5) and detail images using a microscope optic (Figure 6). In the detail image the field-of-view could be selected in the sample volume by manual movement and, in addition, stacks of images in 1 mm thickness within 30 s (scan forth and back) with a stack-to-stack distance of about 6 µm were acquired.

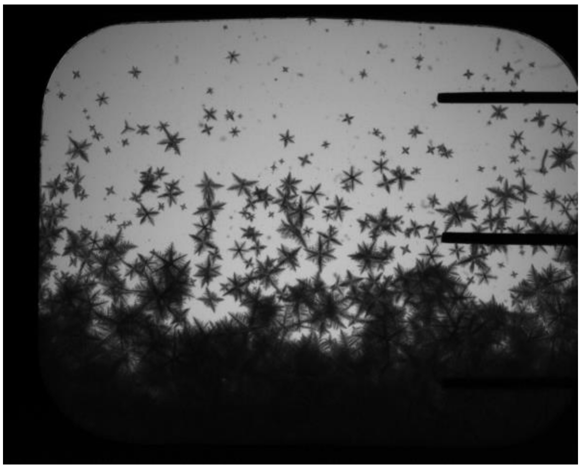

Figure 5: Overall view of the experimental cell showing equiaxed dendrites at t=460s after Lift-Off

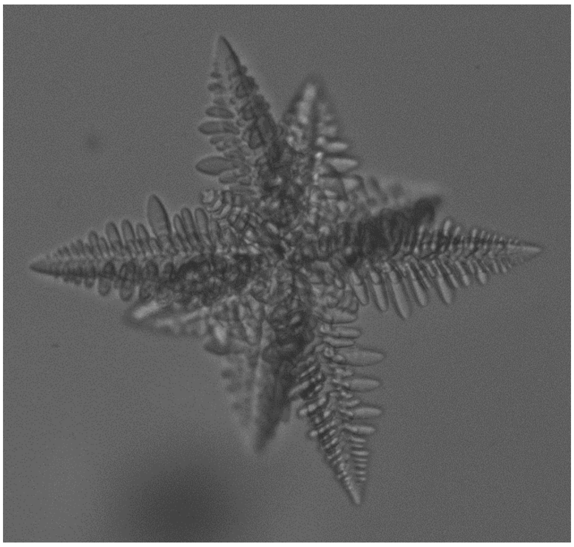

Figure 6: Equiaxed dendritic crystals taken from different focal series (FoV about 1.44 mm width by 1.08 mm height)

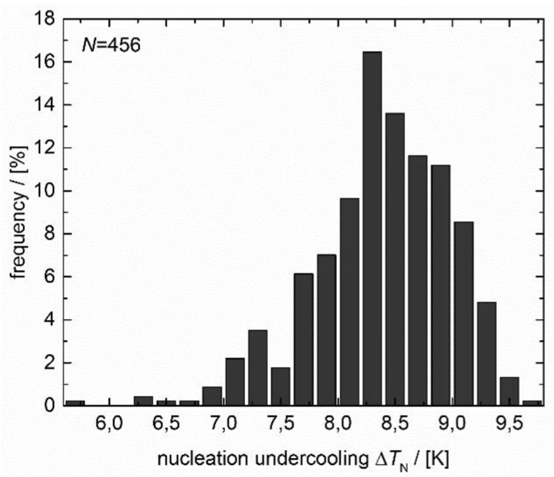

Figure 7: Distribution of estimated nucleation undercooling during transient nucleation after application of the cooling-rate of 0.75 K/min

The spatial position and corresponding interpolated temperature of each new observable equiaxed dendrite was determined from the synchronized overview images and thermocouple readings. The nucleation undercooling at the moment of each nucleation event could then be estimated, the distribution of which is shown in Figure 7 (average nucleation undercooling of 8.4 K for a total number of N=456 equiaxed crystals). A bespoke macroscale numerical model of heat flow, and equiaxed nucleation, growth and impingement is currently in development [8].

Further results for the dendrite morphology evolution, kinetic law, nucleation characteristics, evolution of dendrite density and interaction of dendrite pairs are given in [11].

4. Summary

The investigation of CET in refined Al-7wt%Si alloys processed with different parameters on-board the International Space Station ISS provides a huge amount of unique data for diffusive solidification condition. From extensive sample analysis the microstructures and the grain structures were determined as well as critical parameters for CET. These results are compared to results obtained with different numerical models showing a quite good agreement.

As part of the CETSOL project the sounding rocket experiment MEDI was performed on MASER-13 flight to study in-situ free dendrite growth and solutal interaction between close crystals under microgravity conditions without sedimentation of the equiaxed crystals and melt flow. Now, the experimental data serve as benchmark for modelling of equiaxed growth at different length scales.

In future further experiments on the ISS will be performed in the frame of the CETSOL project focussing on the investigation of CET and fragmentation, both in AlCu alloys and in NPD-DC alloys.

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