ISS-EXPERIMENTS OF COLUMNAR-TO-EQUIAXED TRANSITION IN SOLIDIFICATION PROCESSING

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Abstract

The main topic of the research project CETSOL in the framework of the Microgravity Application Promotion (MAP) programme of the European Space Agency (ESA) is the investigation of the transition from columnar to equiaxed grain growth during solidification. Microgravity environment allows for suppression of buoyancy-driven melt flow and for growth of equiaxed grains free of sedimentation and buoyancy effects. This contribution will present first experimental results obtained in microgravity using hypo-eutectic AlSi alloys in the Materials Science Laboratory (MSL) on-board the International Space Station (ISS). The analysis of the experiments confirms the existence of a columnar to equiaxed transition, especially in the refined alloy. Temperature evolution and grain structure analysis provide critical values for the position, the temperature gradient and the solidification velocity at the columnar to equiaxed transition. These data will be used to improve modeling of solidification microstructures and grain structure on different lengths scales.

Introduction

Casting of metallic alloys often results in a structure which consists of several dendritic grains. This grain structures is the result of a competition between the growth of several arrays of dendrites that develop under constrained and unconstrained conditions. In case of unidirectional solidification typically columnar dendritic grains exist. At high cooling rates or low temperature gradients nucleation and growth of equiaxed grains in the undercooled melt may occur. The often observed change in grain structure is described as a columnar-to-equiaxed transition (CET) [1-4].

The effect of CET was intensively investigated in the last decades because it's of high relevance in industrial application. Therefore, several computational models of the CET were developed. Volume-averaged multi-phase/-scale models [5-8] calculate the transport phenomena on the scale of an entire casting. Here, the CET can be determined based on the volume fractions of columnar and equiaxed grains. Meso-scale models track the growth of the envelope of each individual grain. Examples are the cellular-automaton finite-element (CAFE) model [9] or the front-tracking models [10-12]. Micro-scale models and phase field models resolve details of the solid-liquid interface and, thus, do not require a separate expression for dendrite tip growth or a criterion for the CET [13, 14]. In spite of the fact that these numerical models are able to describe the CET quite well, there are some shortcomings in the modeling studies. Especially the treatment of nucleation and growth of equiaxed grains is the purpose of numerical studies. In the presence of a gravitational field, settling or floatation of solid particles in the melt and their interaction with the development of the columnar grain structure as well as natural convection of the melt itself must be taken into account in order to predict the grain structure of a casting.

Here, a microgravity environment allows for suppression of buoyancy-driven melt flow and so for growth of equiaxed grains free of sedimentation and buoyancy effects. Therefore, experiments in microgravity provide unique data for testing fundamental theories of grain structure formation. To carry out such experiments and to model the process of columnar to equiaxed transition is the topic 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). Some results of experiments performed in microgravity on the International Space Station (ISS) are shown in the paper.

Set-up of the microgravity experiments

To investigate the columnar-to-equiaxed transition under diffusive conditions for heat and mass transport, experiments in microgravity were performed in the Materials Science Laboratory (MSL) with the Low Gradient Furnace (LGF) module onboard the International Space Station (ISS). For a first batch, six samples from Al-7wt%Si alloy (refined and non-refined) were processed. Rod-like samples of diameter 7.8mm and length 245.0mm were integrated in tantalum cartridges, which were also equipped with 12 thermocouples to measure the axial temperature distribution along the sample. **Figure 1** shows such a fully integrated MSL-SCA cartridge. More details are given in [15]. For sample processing the MSL-SCA set-up is inserted into the furnace, which consists of a 'cold zone' with 3 heaters, a 'hot zone' with 4 heaters, separated by an "adiabatic zone". By controlling the temperatures of the cold and the hot zone a temperature gradient along the sample axis is applied. Melting or solidification of the metallic alloy is realized by a movement of the furnace insert along the axis of the fixed sample with a defined speed.

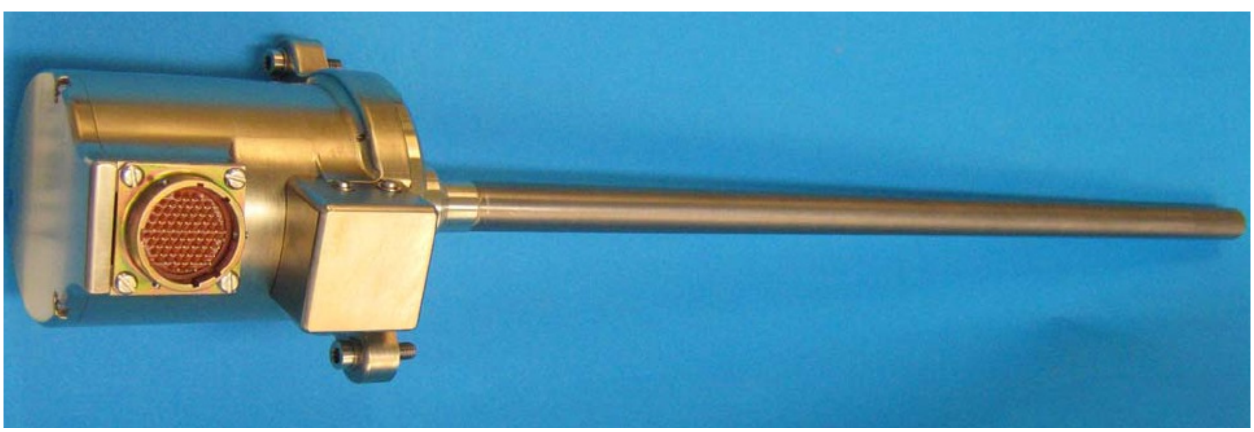

Figure 1: Fully integrated MSL-SCA cartridge to be processed in the MSL-LGF furnace on ISS.

The first phases of the experimental procedure were identical for all six samples FM#1 to FM#6 and result in an initial temperature gradient in the sample of 0.9K/mm. In samples FM#3 and FM#4 non-refined binary Al-7wt%Si alloys were used, while all other samples contain 0.5wt% of TiB_2 for grain refinement (AlSi7 + g.r.). The main process parameters are summarized in **Table 1**. The experiments differ mainly in the homogenization time t_H and in the parameters for the solidification phase 2. In this phase a transition from columnar dendritic growth to equiaxed growth (CET) should be triggered either by increasing the furnace velocity to $v2$=200µm/s or by decreasing the temperature gradient [15].

Table 1: Process parameters of the CETSOL1 flight experiments

Sample No.	Alloy	Homogenization time	Solidification phase 1		Solidification phase 2			Fast movement
		t_H (min)	$v1$ (µm/s)	$z1$ (mm)	$v2$ (µm/s)	$z2$ (mm)	dT/dt (K/min)	$v3$ (µm/s)
FM#1	AlSi7+g.r.	10	10	20	200	50	-4	3000
FM#2	AlSi7+g.r.	300	10	20	200	50	-4	3000
FM#3	AlSi7	300	10	20	200	50	-4	3000
FM#4	AlSi7	300	10	20	10	20	-4	3000
FM#5	AlSi7+g.r.	10	10	20	10	20	-4	3000
FM#6	AlSi7+g.r.	300	10	20	10	20	-4	3000

Grain structure evaluation

As an example, **Figure 2** shows the processed sample CETSOL1 FM#2. The maximum melting position at z=68mm can be identified, which separates the non-molten region and the mushy zone region. The adjacent solidification phases are also shown qualitatively (see also Table 1).

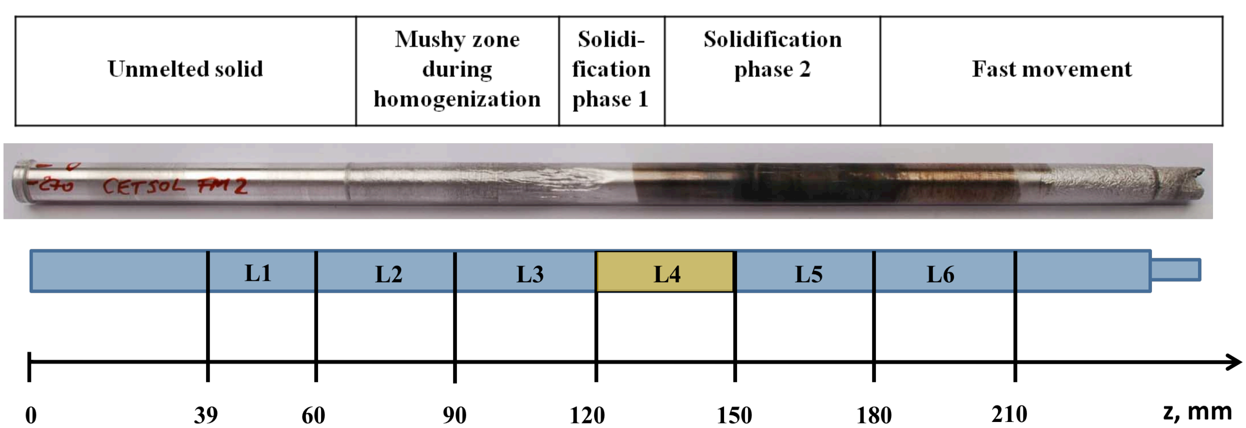

Figure 2: Image of the processed sample CETSOL1 FM#2 with indication of the different regions related to the solidification process (top) and markings for metallographic preparation of longitudinal cross-sections (below).

For analyzing the microstructure and the grain structure the samples have to be sectioned. First, the samples were cut into pieces of length 30mm to analyze the transversal cross-sections. Second, each of these pieces was sawed along the axis to get two halves for analysis of the longitudinal cross-sections L1 to L6 (see also Figure 2).

To determine the microstructure the samples were polished, slightly etched and observed with a microscope. **Figure 3** shows the longitudinal cross-section L4 of FM#2 sample. The length of the cross-section is 30mm, the position values are referred to the non-molten end of each sample. The left part of the cross-section shows a structure with rather large primary dendrites which corresponds to the solidification phase 1 with furnace velocity $v1$=10µm/s. The increase in furnace velocity to $v2$=200µm/s results in the development of a much finer dendritic microstructure.

To identify the grain structure qualitatively the cross-sections were electrolytically etched and analyzed in a polarized light microscope. Then, different colors represent different crystallographic orientations of the dendritic grains [16]. As a result, in cross-section L4 of sample FM#2 two large columnar grains can be identified on the left side (**Figure 4**). The increase of the furnace velocity from $v1$=10µm/s to $v2$=200µm/s results in the development of many smaller grains with different orientations.

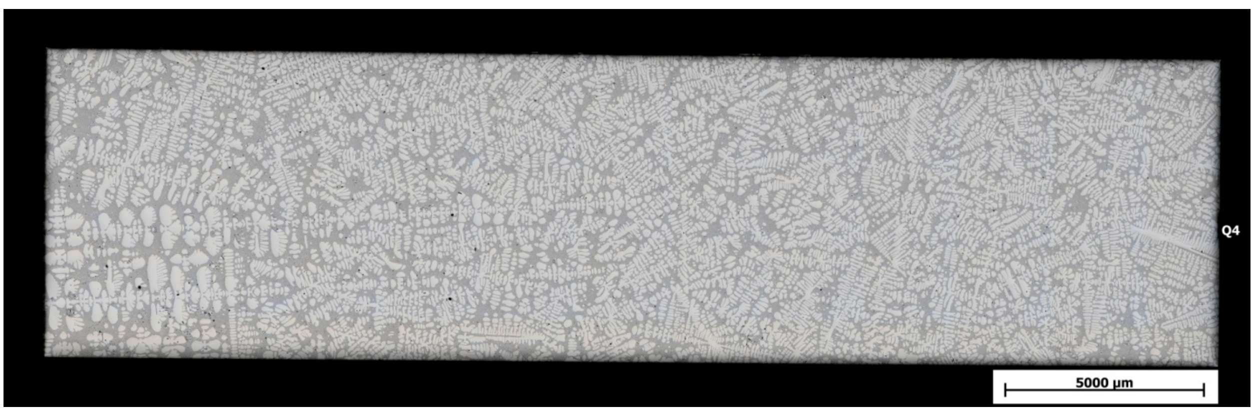

Figure 3: Microstructure in the longitudinal cross-section L4 (z =120mm to 150mm) of FM#2 flight sample; direction of solidification from left to right.

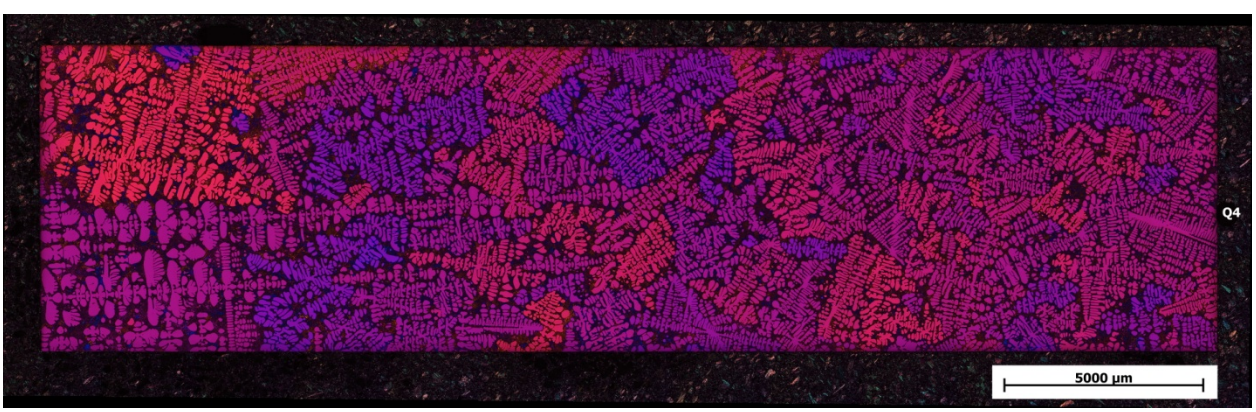

Figure 4: Grain structure in cross-section L4 (z =120mm to 150mm) of FM#2 sample obtained from electrolytical etching.

For quantitative evaluation of the grain structure the cross-section was vibratory polished for many hours. This allows for a surface which is exceptional flat and nearly free of stress. Grain orientation measurements were performed in a scanning electron microscope using an electron backscatter diffraction device (SEM-EBSD) [17]. The size of the field of view is typically 4mm x 8mm. From 8 subsequent measured fields of view the grain structure in the cross-sections L4 of FM#2 sample was analysed quantitatively. For each grain the fibre direction (pole 100) in transverse direction was determined. Related to the rod-like sample this direction corresponds to the sample axis. **Figure 5** shows the resulting grain structure in cross-sections L4 of FM#2 sample. The grey level indicates the deviation of the crystallographic axis of each grain from the sample axis. Black colour corresponds to 0° deviation angle and white colour to the maximum deviation angle of 54.74°. The different grey levels indicate a variety of grains with different orientations, which is characteristic for equiaxed grain growth.

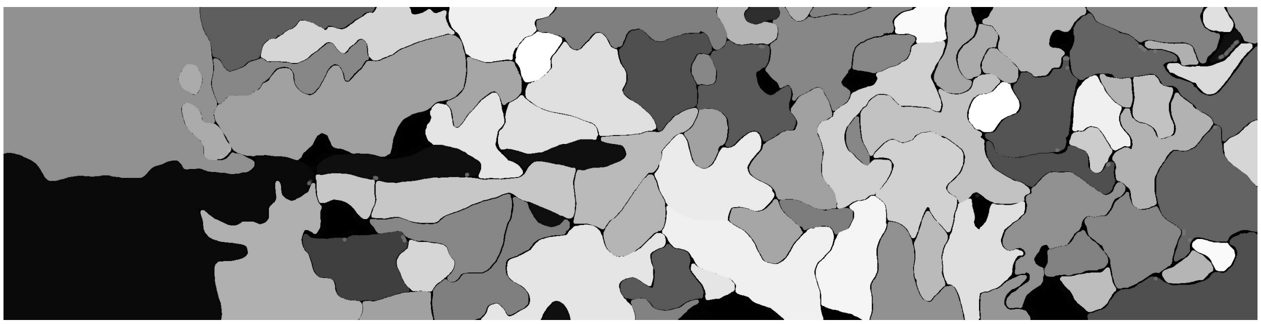

Figure 5: Grain structure in cross-sections L4 (z =120mm to 150mm) of FM#2 sample obtained from EBSD measurements with crystallographic orientation related to the sample axis (white: deviation angle 0°; black: maximum deviation angle 54.74°).

Determination of CET

For determination of the columnar-to-equiaxed transition in FM#2 flight sample the grain structure was evaluated quantitatively from the electrolytically etched longitudinal cross-sections L3 to L5 (**Figure 6**). The grain sizes were measured using digital image analysis. Averaging was performed over all grains being totally or partly within a sheet of ±2mm around the actual position. The maximum value corresponds to columnar growth of just two large grains in the first solidification phase. Equiaxed growth is characterized by a significant decrease of the average grain size. Thus, based on this criterion and this type of evaluation, the region of CET is determined and is centered at z=126±2mm.

The critical parameters for CET, i.e. a critical temperature gradient of about G_c=0.75K/mm and a critical velocity of the isotherm of about v_c=87µm/s, were determined from the temperature measurements along the sample axis and were already given in [15].

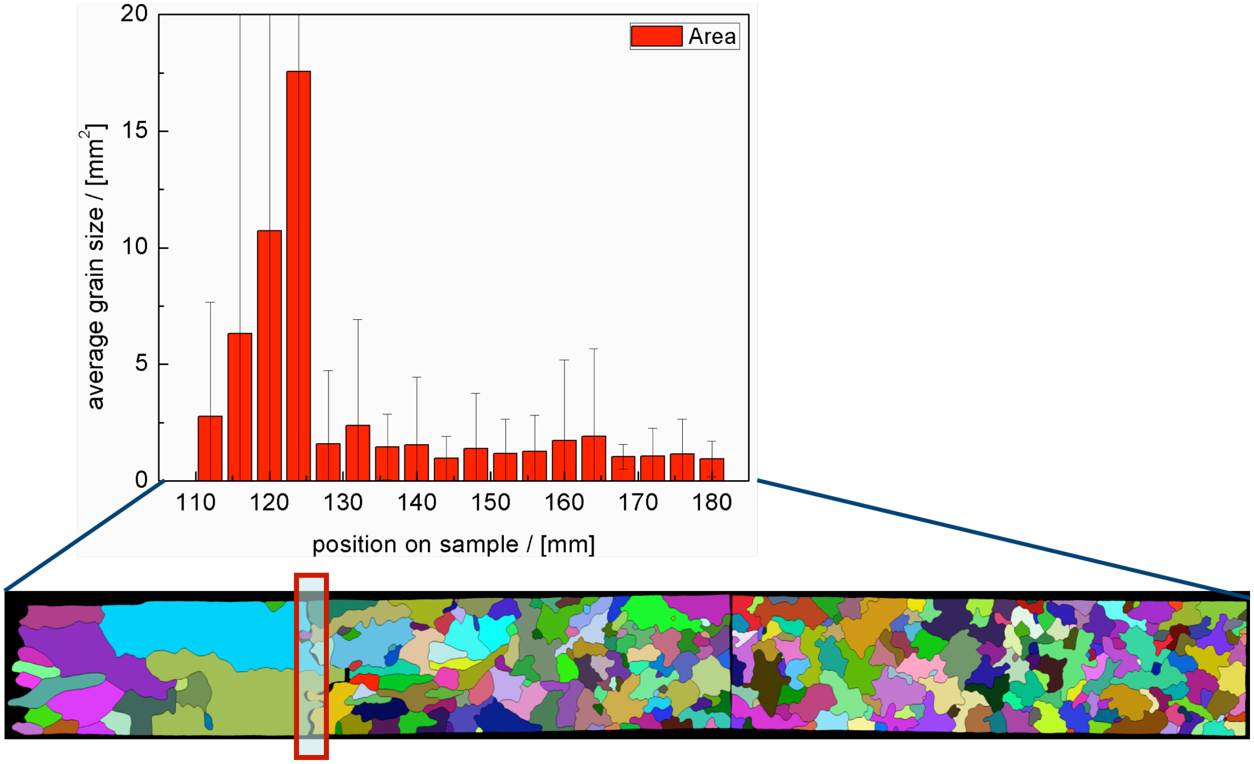

Figure 6: Grain structure identified from longitudinal cross-sections L3 to L5 (z =90mm to 180mm) of FM#2 sample (below) and determination of averaged grain size (top).

Conclusions

This paper reports on results obtained during metallic alloy solidification experiments onboard the International Space Station in the Materials Science Laboratory using the Low Gradient Furnace module. Within a first batch six experiments with Al-7wt%Si alloy were performed successfully to investigate columnar-to-equiaxed (CET) solidification behaviour in microgravity. Qualitative and quantitative grain structure analysis using electrolytical etching and EBSD-technique show that CET is observed in the grain refined samples. Here, the microgravity environment allows for pure diffusive conditions for heat and mass transfer in the melt and therefore for investigation of CET without buoyancy convection and sedimentation of equiaxed grains in the melt. The critical parameters for CET were determined from analysis of thermal data and taking into account the grain structure. These data basis will be used for calibration and further development of numerical model predicting CET.

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References

1. J.D. Hunt, ”A numerical analysis of time dependent isolated dendritic growth for conditions

near the steady state," *Acta Metall. Mater*., 38 (1990), 411-418.
2. Ch.-A. Gandin, "From constrained to unconstrained growth during directional solidification," *Acta Materialia* 48 (2000), 2483-2501.
3. D.J. Browne and J.D. Hunt, "A fixed grid front-tracking model of the growth of a columnar front and an equiaxed grain during solidification of an alloy," *Numerical Heat Transfer, Part B: Fundamentals* 45 (2004), 395-419.
4. L. Sturz and G. Zimmermann, "Investigations on Columnar-to-Equiaxed Transition in Binary Al Alloys with and without Grain Refiners," *Materials Science Forum* 508 (2006), 419-424.
5. C. Beckermann and C.Y. Wang, "Multi-Phase/-Scale Modeling of Transport Phenomena in Alloy Solidification," *Annual Review of Heat Transfer VI*, ed. C.L. Tien, Begell House, New York, NY, (1995), 115-198.
6. M.A. Martorano et al., "A solutal interaction mechanism for the columnar-to-equiaxed transition in alloy solidification," *Metall. Mater. Trans. A*, 34A (2003), 1657-1674.
7. A. Ludwig and M. Wu, "Modeling the columnar-to-equiaxed transition with a three-phase Eulerian approach," *Mater. Sci. Eng.*, A413-414 (2005), 109-114.
8. A. Noeppel et al., "Numerical modelling of columnar to equiaxed transition – application to microgravity experiments," *Int. Journal of Cast Metals Research* 22 (2009), 34-38.
9. Ch.-A. Gandin and M. Rappaz, "A coupled finite element-cellular automaton model for the prediction of dendritic grain structures in solidification processes," *Acta Metall. Mater.* 42, (1994), 2233-2246.
10. I. Steinbach et al., "Three-dimensional modeling of equiaxed dendritic growth on a mesoscopic scale," *Acta Mater.* 47, (1999), 971-982.
11. P. Delaleau et al., "Mesoscopic Simulation of Dendritic Growth Observed in X-ray Video Microscopy During Directional Solidification of Al–Cu Alloys," *ISIJ International* 50 (2010), 1886-1894.
12. J. Banaszek et al., "Natural Convection and Columnar-to-Equiaxed Transition Prediction in a Front-Tracking Model of Alloy Solidification," *Metallurgical Materials Transact.* A38 (2007), 1476-1484.
13. H.B. Dong and P.D. Lee, "Simulation of the columnar-to-equiaxed transition in directionally solidified Al–Cu alloys," *Acta Mater*. 53 (2005), 659-668.
14. A. Badillo and C. Beckermann, "Phase-field simulation of the columnar-to-equiaxed transition in alloy solidification," *Acta Mater*. 54 (2006), 2015-2026.
15. G. Zimmermann et al., "Investigation of columnar-to-equiaxed transition in solidification processing of AlSi alloys in microgravity – The CETSOL project", accepted in *IOP Conference Series Materials Science and Engineering, ISPS4.*
16. E. Schaberger, F. Grote and A. Schievenbusch, "Farbätzung und Farbbildanalyse - Ein Weg zur Charakterisierung von Gefügen innovativer Gusswerkstoffe," *Prakt. Metallographie 37* (2000), 419-434.
17. A.J. Schwartz et al., *Electron Backscatter Diffraction in Materials Science* (Springer Science+Business Media, 2nd edition, 2009)