Investigation of columnar-to-equiaxed transition in solidification processing of AlSi alloys in microgravity – The CETSOL project

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Abstract. Grain structures observed in most casting processes of metallic alloys are the result of a competition between the growth of several arrays of dendrites that develop under constrained and unconstrained conditions. Often this leads to a transition from columnar to equiaxed grain growth during solidification (CET). A microgravity environment results in suppression of buoyancy-driven melt flow and so enables growth of equiaxed grains free of sedimentation and buoyancy effects. This contribution presents first results obtained in experiments on-board the International Space Station (ISS), which were performed in the frame of the ESA-MAP programme CETSOL. Hypoeutectic aluminium-silicon alloys with and without grain refiners were processed successfully in a low gradient furnace (MSL-LGF). First analysis shows that in the non grain refined samples columnar dendritic growth exists, whereas CET is observed in the grain refined samples. From analysis of the thermal data and the grain structure the critical parameters for the temperature gradient and the cooling rate describing CET are determined. These data are used for initial numerical simulations to predict the position of the columnar-to-equiaxed transition and will form a unique database for calibration and further development of numerical CET-modeling.

1. Introduction
Grain structures observed in most casting processes of metallic alloys are the result of a competition between the growth of several arrays of dendrites that develop under constrained and unconstrained conditions. Figure 1 schematically shows the solidification of a metallic melt in a mould. Typically for a high cooling rate at the beginning directional growth of columnar dendritic grains from the side walls is initiated. During solidification the temperature difference between the melt and the mould is reduced and the cooling rate decreases. This often leads to nucleation and growth of equiaxed grains in the undercooled melt. This effect is described as a columnar-to-equiaxed transition (CET) and was investigated intensively in the last decades [1-12].
On ground a far-reaching effect of the buoyancy-driven flow is observed. This leads to relative transport of the equiaxed crystals and flow-induced macrosegregation. It results in difficulties in conducting precise investigations on the origin of the formation of the equiaxed crystals and their interaction with the development of the columnar grain structure.

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 [13-14]. 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). At present the CETSOL team consists of four European scientific partners assisted by five partners from industry.

Figure 1. Schematic drawing of columnar and equiaxed grain growth during solidification

2. 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 and will be performed. In 2006 the microgravity experiment MACE (Metallic Alloys for Columnar-Equiaxed Transition) was carried out in the experimental module TEM 01-2M during MAXUS-7 sounding rocket mission. Within this campaign three Al-7wt%Si samples were processed successfully. As a result the apparent effect of TiB$_2$ grain refiner particles in the alloy was to lower the critical undercooling for heterogeneous nucleation of primary equiaxed Al-dendrites in the bulk; CET was thus present at higher gradients and lower front velocities. Also, a significant delay of CET is observed on ground compared to the experiments performed in reduced gravity environment [15-19].

In continuation of these investigations experiments in microgravity were defined and successfully performed in the Materials Science Laboratory (MSL) with the Low Gradient Furnace (LGF) module onboard the International Space Station (ISS). For a Batch#1 the CETSOL team selected 6 experiments to investigate CET behaviour in microgravity. The AlSi7 sample material with and without grain refiner particles was provided by Hydro Aluminium Deutschland GmbH. Rod-like samples with diameter 7.8 mm and length 245 mm were integrated into flight cartridges by industry (EADS Astrium / Soterem). Figure 2 shows a sketch of the sample cartridge assembly (SCA) located
in the furnace (LGF). The metallic Al-7wt%Si alloy is enclosed in a protective alumina tubular crucible with Shapal plugs and boron nitride spacers at both ends. The conical shape of the upper Shapal plug acts as passive volume compensation device during heating-up and melting of the alloy. At the outer surface of the alumina tube 4 longitudinal grooves were manufactured. In each groove 3 thermocouples at different axial positions were placed. This allows measuring the temperature profile along the sample axis within the crucible walls. The whole set-up is inserted leak-tight in a tantalum tube and filled with 200 mbar helium. For sample processing the cartridge is inserted into the low gradient furnace (LGF). The main parts of the LGF are the relatively cold zone which consists of 3 heaters H1 to H3, the hot zone which consists of 4 heaters, and an adiabatic zone in between (see figure 2). By controlling the temperatures of the cold and the hot zone a temperature gradient along the sample axis develops. 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. The situation in figure 2 shows the furnace at its very left position (zero furnace position), which corresponds to the maximum melting position of the sample at the beginning of the solidification phase.

![Figure 2](image-url) **Figure 2.** Sketch of a sample cartridge (MSL-SCA). Its position inside the low gradient furnace (LGF) corresponds to the beginning of the solidification phase.

Table 1 summarizes the main parameters of the flight experiments. In samples FM#3 and FM#4 non-refined AlSi7 alloys were used, while all other samples contain 0.5wt% of TiB2 for grain refinement. At furnace position 80mm the temperatures of all heaters were increased to 500°C, followed by a further increase to 540°C for the 3 heaters of the cold zone and to 660°C for the 4 heaters of the hot zone. With this a temperature profile was established along the sample axis resulting in a partly melting of the Al-7wt%Si alloy in the hot zone region. Moving the furnace at a speed of 80µm/s to the end position (as shown in figure 2), directional melting of the sample was initiated. With an additional increase of the temperatures of all heaters via telecommand by 30K the solid-liquid region of the sample was centred in the adiabatic zone of the furnace. From the temperature measurements along the sample axis a temperature gradient of about 0.9K/mm at the liquidus temperature of 618°C was determined. This procedure was identical for all flight experiments to allow meaningful comparison of the experimental results.

After reaching thermal equilibrium the solidification process was started following different thermal homogenization times $t_H$. A short homogenization time of about 10 minutes just stabilizes the temperature profile. During a waiting period of 5 hours a diffusive coarsening process in the mushy zone region is expected, which may result in an axial segregation of silicon due to a temperature gradient zone melting (TGZM) effect [20].
The first solidification phase was identical for all experiments (see table 1). A furnace movement with \( v_1 = 10 \mu m/s \) for \( z_1 = 20 \) mm is expected to generate a columnar dendritic growth. A transition to equiaxed growth should be triggered in the second 50mm solidification phase of furnace movement. Therefore the temperatures of the heaters in the hot zone were decreased at \( dT/dt = 4 \) K/min. Additionally, in the experiments FM#1 to FM#3 the furnace speed is increased to \( v_2 = 200 \mu m/s \). Finally a fast movement of the furnace with \( v_3 = 3000 \mu m/s \) to its end position and a shut down of the heaters stops the experiment.

| Sample No. | Alloy          | Homogenization time \( t_H \) (min) | Solidification phase 1 | Solidification phase 2 | Fast movement |
|------------|----------------|--------------------------------------|------------------------|------------------------|---------------|
| FM#1       | AlSi7+g.r.     | 10                                   | 10                      | 20                     |               |
| FM#2       | AlSi7+g.r.     | 300                                  | 10                      | 20                     | 200           |
| FM#3       | AlSi7          | 300                                  | 10                      | 20                     | 200           |
| FM#4       | AlSi7          | 300                                  | 10                      | 20                     | 200           |
| FM#5       | AlSi7+g.r.     | 10                                   | 10                      | 20                     | 200           |
| FM#6       | AlSi7+g.r.     | 300                                  | 10                      | 20                     | 200           |

The flight experiments have been performed from November 2009 until April 2010 on the ISS. The processed cartridges have been downloaded with Space Shuttles and de-integrated by industry, followed by scientific evaluation of the CETSOL team. First results are presented in the next chapter.

3. CETSOL1 Batch#1 microgravity experiments

The performance of the flight samples consists of several steps which were agreed by the CETSOL team partners to achieve as much scientific output as possible. The main items can be summarized as follows:

3.1. Process parameters

The critical phases of each microgravity experiment, i.e. the homogenization and solidification phases, were performed during sleeping periods of the astronauts on the ISS. Thereby a microgravity level without larger disturbances was expected. The evaluation of the data of the gravity sensors close to the MSL rack confirms that a gravity level below \( \pm 0.5 \times 10^{-3} \) \( g_0 \) was achieved during all experiments, where \( g_0 = 9.8m/s^2 \) is the standard terrestrial value.

The housekeeping data – mainly heater and sample temperatures as well as the furnace movement – were controlled on-line during the experiments. This monitoring at the Microgravity User Support Centre (MUSC/DLR) in Cologne (Germany) also allowed the scientist to adjust the heater temperatures to shift the solid-liquid interface position in the metallic sample into the adiabatic zone of the MSL-LGF furnace. As an example figure 3 shows the temperatures of the 12 thermocouples which are placed along the sample with distances between two neighbouring thermocouples of 10mm. The position of the first thermocouple SC/Temp tc_01 is 72mm from the non-molten end of the metallic sample. The diagram axis on the right side in figure 3 indicates the furnace position \( s \). The graphs correspond to the solidification phases for experiment FM#2, starting with a furnace movement of \( v_1 = 10 \mu m/s \) at \( t = 27380s \), an increase to \( v_2 = 200 \mu m/s \) at \( t = 29395s \) and finally a fast movement at...
The evaluation of the data demonstrates that all microgravity experiments were performed nominally.

Figure 3. Temperatures of the 12 thermocouples along the sample and furnace position $s$ (axis to the right) during the different solidification phases for experiment FM#2.

3.2. Non-destructive sample analysis

The next stage of evaluation is a non-destructive analysis of the processed samples using 3D computer tomography. As a result no macroscopic shrinkage holes were detected. This indicates that the volume compensation mechanism worked reliably. In the samples with short homogenization times of about 10 min some pores with sizes up to 500 $\mu$m were identified, mainly in the mushy zone region. This fact has to be taken into account for further analysis.

Figure 4 shows an image of the processed FM#2 flight sample after extraction from the alumina crucible. The maximum melting position at $z=68$ mm can be identified, which separates the non-molten region and the mushy zone region. The adjacent solidification phases are also shown qualitatively.

Figure 4. Processed FM#2 flight sample with related experiment phases (top) and identification of cross-sections for the metallographic preparation (below).
3.3. Microstructure and grain structure evaluation

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 Qi (see figure 4). Second, each of these pieces was sawed along the axis to get two halves for analysis of the longitudinal cross-sections Li. To determine the microstructure the samples were polished and etched and observed with a microscope. To identify the grain structure the cross-sections were electrolytically etched and analysed in a polarized light microscope. Then, different colours represent different crystallographic orientations of the dendritic grains [21].

Figure 5 shows the longitudinal cross-sections L3 of FM#2 and FM#3 flight samples (for notation see figure 4). The lengths of the cross-sections are 30mm, the position values are referred to the non-molten end of each sample with a difference of 7mm between the FM#2 and FM#3 flight samples. Such a difference can be explained by small temperature differences (about 6K) within the two samples in the very flat gradient. The left part of each cross-section shows the developed mushy zone region. As a result of the long thermal homogenization of 5 hours in a temperature gradient (see table 1) coarsening of the primary Al-rich dendrites exists. Between refined (FM#2) and non-refined (FM#3) alloy no significant difference of the mushy zone structure is detected. After the thermal homogenization the directional solidification with $v_1=10\mu m/s$ was started. The development of a dendritic microstructure is shown in the right part of the cross-sections (figure 5). Starting from several nuclei in both samples, the non-refined alloy (FM#3) shows a more pronounced columnar structure than the refined alloy (FM#2). Concerning the grain size and grain density no substantial difference is found.

![FM#3 cross-section](image1)

**FM#3 (L3: $z = 97mm$ to $127mm$)**

![FM#2 cross-section](image2)

**FM#2 (L3: $z = 90mm$ to $120mm$)**

**Figure 5.** Microstructure and grain structure in longitudinal cross-sections L3 of FM#2 and FM#3 flight samples showing the mushy zone region (left) and the onset of columnar dendritic growth.
Figure 6 shows the cross-sections L4 of samples FM#2 and FM#3. The increase of the furnace velocity from $v_1=10\,\mu\text{m/s}$ to $v_2=200\,\mu\text{m/s}$ results in the development of a finer columnar dendritic microstructure in the non-refined alloy (FM#3) whereas the grain refined alloy (FM#2) shows a transition to equiaxed grain growth.

The differences in the microstructures and grain structures are also evident in the transversal cross-sections Q4 given in figure 7. Sample FM#3 shows a regular dendritic microstructure. The uniform color of the electrolytically etched image indicates the columnar growth of nearly one single grain. In the grain refined sample FM#2 however many grains with different orientations were identified.

![Microstructure and grain structure in longitudinal cross-sections L4 of FM#2 and FM#3 flight samples showing columnar growth (FM#3) and transition to equiaxed grain growth (FM#2)](image)

**Figure 6.** Microstructure and grain structure in longitudinal cross-sections L4 of FM#2 and FM#3 flight samples showing columnar growth (FM#3) and transition to equiaxed grain growth (FM#2)

### 3.4. Columnar-to-equiaxed transition

For determination of the columnar-to-equiaxed transition in Batch#1 FM#2 flight sample the grain structure was evaluated quantitatively. From the longitudinal cross-sections L3 to L5 the grain sizes were measured using digital image analysis. The average grain size along the sample axis is given in figure 8, averaging was performed over all grains being totally or partly within a sheet of $\pm2\,\text{mm}$ 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 position of CET is determined at $z=126\pm2\,\text{mm}$.
Figure 7. Transversal cross-sections Q4 of the Batch#1 flight samples FM#3 (top) and FM#2 showing the dendritic microstructure (left) and the corresponding grain structure (right).

The relevant parameters – critical temperature gradient and critical velocity of the isotherm at CET – have to be determined from the temperature measurements along the sample axis.

To estimate the critical isotherm velocity the local cooling rate is determined. Figure 9 shows the temperature and calculated cooling rate of thermocouple SCAtemp_tc_07. This thermocouple is located at position $z=132$mm in the alumina crucible. This position is about 6mm ahead of the measured position of CET in the sample and is assumed to be representative for the thermal behaviour in the undercooled region ahead of the columnar front at CET. At about $t=29500$ s the temperature of the thermocouple reaches the liquidus temperature of the alloy ($T_L=618^\circ$C). Assuming a front undercooling of about 6K this temperature is reached at about $t=29600$ s. The corresponding cooling rate in this time period is rather constant at 0.065K/s.
Figure 8: Grain size obtained from longitudinal cross-sections L3 to L5 of Batch#1 flight sample FM#2

Figure 9: Temperature profile and cooling rate at thermocouple SCAtemp_tc_07 during the solidification phases

Figure 10 shows the temperature gradient at the liquidus-temperature during the solidification phases. The gradient is constant at $G=0.95\text{K/mm}$ in the first solidification phase with $v_l=10\mu\text{m/s}$. 
During the second solidification phase with a furnace movement of \(v_2=200\mu m/s\) a slight decrease of the temperature gradient is measured. Assuming CET at \(t=29600\) s the critical temperature gradient is about \(G_c=0.75K/mm\). As a result the critical isotherm velocity is calculated from the critical values for the gradient and the cooling rate as about \(v_c=87\mu m/s\).

![Figure 10: Temperature gradient at the solid-liquid interface for sample FM#2 during the solidification phases \((v_1=10\mu m/s, v_2=200\mu m/s)\) ](image)

3.5 Numerical prediction of CET position

The modelling approach [12] that combines Front Tracking (FT) of columnar growth [5] and an equiaxed volume averaging method [22] was employed to simulate the FM#2 experiment. Among the key advantages related to this approach are: the ability to explicitly track the undercooled columnar front; the capacity to model equiaxed nucleation from grain refiners and subsequent unconstrained growth; high computational efficiency for modeling equiaxed solidification in this range of equiaxed grain sizes; and the macroscopic nature of CET.

The combined model needs to solve the macroscopic energy equation with appropriate latent heat source terms which come from the simulation of columnar and equiaxed growth. The columnar growth initiation from initial solid/mush is considered. The model tracks the evolution of the columnar dendritic front over the relevant period. In this simulation, the columnar front is assumed to commence its growth from the mushy zone (at the liquidus isotherm of the Al-7wt%Si alloy) that exists when cooling (solidification) begins. Therefore, the initial position of the computational markers that represent the columnar front was set at the computed temperature profile’s liquidus isotherm. In the undercooled liquid, ahead of the columnar front, the possibility of equiaxed nucleation from grain refiners is considered. A log-normal distribution for grain refiner size [23, 24] is used to compute the number of potential equiaxed nucleation sites at a given time using the computed site temperature. More details of the equiaxed nucleation model employed are available elsewhere [22].

The simulation domain is a 132mm long section from the experimental sample and starts at position \(z=61.5\) mm (zero position corresponds to solid sample end). To define the thermal boundary conditions for the simulations, thermal data recorded during the experiment on the ISS was used. As
mentioned in the experimental section, thermal data is collected from 12 thermocouple positions at 10 second intervals. Intermediate spatial and temporal input data values used to feed the model simulation were linearly interpolated. A pre-experiment chemical analysis showed that the FM#2 sample contained 30 ppm boron and 225 ppm Ti. We assumed that (i) all boron atoms appeared in TiB$_2$ grain refiner particles and (ii) a nearly 1% efficiency of such TiB$_2$ grain refiners. The effective TiB$_2$ density in the sample was computed as 1.25/mm$^3$ using the previous literature [23, 24]. Considering that a slice with thickness equal to half of the average equiaxed dendrite diameter contributes to the 2D view, the input grain refiner density for the 2D model is obtained via multiplying the 3D grain refiner density value by a factor equal to 0.75mm$^{-1}$ (the as-cast average equiaxed dendrite diameter is approximately 1.5mm).

Alloy related thermo-physical properties, for both solid and liquid phases, and the dendrite kinetics growth law (tip velocity-undercooling relationship) were taken from ref. [4]. Predicted as-cast columnar and equiaxed volume fractions together with CET position from the model simulation are presented in figure 11. The zoomed-in section of figure 11 can be directly compared with the FM#2 - L4 section of figure 6.

![Figure 11](image1.jpg)

**Figure 11.** Predicted final as-cast columnar and equiaxed volume fractions in simulation domain. The section between $z=120$mm to $z=150$mm is magnified.

As mentioned in chapter 3.4, CET for FM#2 experiment was determined at $z=126\pm2$ mm. In the model simulation, CET is predicted at a distance of $z=127.5$mm. Hence a strong agreement between model simulation and experiment was found. The columnar length is approximately equal to the distance that the furnace moved at the lower velocity ($v_1$) and relatively high thermal gradient (see figure 10) suggesting that CET is related to the velocity jump. However, under a similar velocity jump, FM#3 has not shown a CET. Therefore, the influence of grain refiners needs to be evaluated carefully via with further modeling.

4. Summary
This paper reports on first results obtained during metallic alloy solidification experiments onboard the International Space Station in the Materials Science Laboratory using the Low Gradient Furnace module. Within Batch#1 a set of six experiments with Al-7wt%Si alloy were performed successfully to investigate columnar-to-equiaxed (CET) solidification behaviour in microgravity. Mainly the impact of grain refiner particles in the alloy and different cooling rates on CET were investigated. First evaluations show that in the non grain refined samples columnar dendritic growth persists throughout,
whereas 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 determination of critical parameters for CET from analysis of thermal data and the grain structure will be used for calibration and further development of numerical model predicting CET.

At present a second set of microgravity experiments is in preparation. Within the Batch#2a a set of seven experiments with Al-7wt%Si alloy will be processed in the MSL using the Solidification and Quenching Furnace module (SQF). This furnace allows for higher temperature gradients and cooling rates and therefore resulting in an extension of the data basis concerning CET.

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