Validation of a Front-Tracking Model of the Columnar to Equiaxed Transition using Solidification Results from the Maxus 7 Microgravity Platform

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Abstract
To study the columnar-to-equiaxed transition (CET) in alloy castings, three aluminum-silicon samples were solidified onboard the MAXUS 7 sounding rocket. Temperature measurements were made during the flight and the samples were retrieved and analyzed for their solidified macrostructure. Two of the samples produced a CET while the third sample produced a mixed structure with grains nucleating on the crucible walls. A novel front tracking model of solidification and a model of the MACE test apparatus are presented. Validation of the solidification code is carried out. Cooling curves from the experiments are predicted. A simulation of the solidified macrostructure is also given for each experiment. The CET positions are predicted in agreement with the experimental results.

1. Introduction
When casting metallic alloys, the solid crystals (called dendrites) that grow from the liquid create the grain structure of the casting. This grain structure, sometimes called the macrostructure, is important because it can influence the final properties and the quality of the casting.

The Columnar to Equiaxed Transition (CET) is an important phenomenon that can occur in a casting’s macrostructure during the solidification stage. Columnar dendrites in a casting usually nucleate at the mould walls and grow in a direction parallel with, but in the opposite direction to, the heat flow. The columnar dendrites form columnar grains in the macrostructure. The columnar grains are recognizable with their large aspect ratio and elongated shape. Dendrites can also nucleate within the bulk liquid. Under low temperature gradient conditions, these dendrites grow and impinge with neighboring dendrites to create an equiaxed grain structure. Equiaxed grains have a lower aspect ratio than columnar grains and have random crystal orientations. At some point in the macrostructure of the casting, a CET will exist if sufficient equiaxed dendrites nucleate ahead of the columnar dendrites, thus blocking their progress.

In some engineering applications, columnar macrostructures are desired, for example, in turbine blades where high-temperature creep resistance is required. In other applications, equiaxed structures are preferred for the improvements they provide in increasing a component’s strength and reducing the likelihood of internal shrinkage defects\textsuperscript{5}. Thus, understanding the origin of the CET is of considerable importance in materials processing.

Much terrestrial work has been performed on understanding the CET\textsuperscript{5}. The European Space Agency has coordinated a specific Microgravity Application Promotion (MAP) project called CETSOL to use microgravity conditions to study the development of CET in castings\textsuperscript{3}. The objectives of the CETSOL program are to obtain both microgravity and terrestrial-gravity experimental results on CET formation and these results enabled validation of various modeling approaches adopted within CETSOL, now in its third phase. The MAXUS 7 sounding rocket was used as the microgravity platform for CETSOL. Three sample experiments on aluminum silicon alloys were launched onboard MAXUS 7 in May 2006. This experimental program was called Metallic Alloys in Columnar Equiaxed solidification or simply MACE. The experiment was launched with collaboration from the CETSOL partners, EADS-Astrium, Swedish Space Corporation, and ESA.
loss to the heat sink, thereby giving a controlled cooling rate, \( C_R \), in the sample.

Because each heater was controlled separately, two distinct regions with different axial temperature gradient values were achieved in the sample. An axial temperature gradient, \( G_1 \), was set up between heaters H1 and H2. Similarly an axial temperature gradient, \( G_2 \), was set up between heaters H2 and H3. Thus, the temperature profile of the sample was controlled along its length in the region between heaters H1 and H3, that is, from axial position 103 mm to 163 mm.

For observational reasons, additional thermocouples were mounted in the crucible walls at a distance of 0.5mm from the Al-Si sample. Fig. 1 shows the axial positions of these thermocouples.

Further details on the experimental setup and experimental procedure are available in other published works.4,5)

3. Test Samples

Three samples of Al-Si alloys were investigated on board the MAXUS 7, namely MACE A, MACE B, and MACE C. Table 1 summarizes the main controlled parameters achieved during the experiments.

MACE A and MACE B have similar thermal conditions and differ only in their chemical constituents: in MACE B a grain refiner (215 \( \mu \)g/g titanium and 30\( \mu \)g/g boron) was introduced to see the effects of inoculation on CET. Both MACE A and MACE C were free of the inoculant; however, MACE C had lower axial temperature gradients and a lower cooling rate than MACE A.

4. Experimental Results

Detailed characterization and analyses of the sample microstructures are available elsewhere.4,5,6) A brief account of important findings are presented herein.

Because of the experimental procedure and the axial temperature gradient, the Al-Si rod was partially melted at the beginning of the test flight. Thus, part of the rod remained solid throughout the entire experiment and this helped contain the liquid at the hot end of the apparatus.

Three solid zones were distinguishable along the length of the final cross section of the recovered samples, namely, the un-melted solid, a partially-stabilized Temperature Gradient Zone Melted (TGZM) section, and the as-cast solidified structure. The boundary of each section along the sample was planar and transverse to the central axis. The TGZM section, which was formed from a semi-solid region, developed through the initial holding period prior to lift off. Studies have been performed on TGZM7) that show if a TGZM is allowed to fully stabilize into a solid by holding the temperature for long periods, then the composition profile in the remaining liquid will be influenced and the average composition of the liquid will be increased. However, the holding period for the MACE experiment was 22 minutes and this time was deemed much too short for the TGZM to fully stabilize and to influence the composition profile in the liquid. This assumption is supported by the presence of solidified, solute-rich droplets in the TGZM.

The vertical positions of the TGZM boundaries are

<table>
<thead>
<tr>
<th>Table 1 Samples flown on MAXUS 7</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>MACE A</strong></td>
</tr>
<tr>
<td>Sample composition:</td>
</tr>
<tr>
<td>Al-7wt.%Si</td>
</tr>
<tr>
<td>Axial temperature gradients:</td>
</tr>
<tr>
<td>( G_1 = 31.3 \text{ K/cm} ); ( G_2 = 7.6 \text{ K/cm} )</td>
</tr>
<tr>
<td>Cooling rate:</td>
</tr>
<tr>
<td>( C_R = 12 \text{ K/min} )</td>
</tr>
<tr>
<td><strong>MACE B</strong></td>
</tr>
<tr>
<td>Sample composition:</td>
</tr>
<tr>
<td>Al-7wt.%Si with Ti-B Grain Refiner</td>
</tr>
<tr>
<td>Axial temperature gradients:</td>
</tr>
<tr>
<td>( G_1 = 30.2 \text{ K/cm} ); ( G_2 = 7.4 \text{ K/cm} )</td>
</tr>
<tr>
<td>Cooling rate:</td>
</tr>
<tr>
<td>( C_R = 12 \text{ K/min} )</td>
</tr>
<tr>
<td><strong>MACE C</strong></td>
</tr>
<tr>
<td>Sample composition:</td>
</tr>
<tr>
<td>Al-7wt.%Si</td>
</tr>
<tr>
<td>Axial temperature gradients:</td>
</tr>
<tr>
<td>( G_1 = 15.9 \text{ K/cm} ); ( G_2 = 4.4 \text{ K/cm} )</td>
</tr>
<tr>
<td>Cooling rate:</td>
</tr>
<tr>
<td>( C_R = 6 \text{ K/min} )</td>
</tr>
</tbody>
</table>
Table 2  TGZM boundary positions and CET positions measured along the length of the sample (reference: the datum in Fig. 1)

<table>
<thead>
<tr>
<th></th>
<th>TGZM Lower Position (mm)</th>
<th>TGZM Upper Position (mm)</th>
<th>Approx. CET Position (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MACE A</td>
<td>99.0</td>
<td>111.4</td>
<td>147.9</td>
</tr>
<tr>
<td>MACE B</td>
<td>98.6</td>
<td>109.6</td>
<td>132.5</td>
</tr>
<tr>
<td>MACE C</td>
<td>112.0</td>
<td>135.2</td>
<td>No CET</td>
</tr>
</tbody>
</table>

The upper boundary of the TGZM distinguishes the extent of the initial liquid at the beginning of the microgravity experiment. It is this boundary that distinguishes the most interesting part of our solidification results, namely, the as-cast solidified region.

Table 2 also shows the position of the CET found in the samples. The CET for MACE A was quite clearly observed from the longitudinal cross section of the sample. Because of the inoculation, MACE B had a higher grain density, and, therefore, the CET in MACE B was not as easy to observe as it was in MACE A. However, statistical processing of the data showed that a trend in grain size along the length of MACE B.

MACE C showed no observable trend to indicate the presence of a CET. Instead, the macrostructure was a mixture of columnar and equiaxed grains. It appeared that grains nucleated at the crucible walls and grew towards the centre of the sample. Transverse sections of MACE C were also examined and some dendrites at the crucible walls were pointing inwards to the centre of the sample, thus supporting the hypothesis that grains nucleated at the crucible walls.

5. Modeling with the Front Tracking Method
A 2-D front tracking model was developed through the CETSOL program, which describes binary alloy solidification under diffusion-controlled solidification. This front tracking model simulates each growing dendrite within an envelope; thus, the model simulates at the mesoscale of the grain and excludes microscopic crystallographic details. The latent heat released from each growing grain is simulated according to the Scheil equation. The properties of the material (specific heat and conductivity) change as function of temperature and phase fraction, thus giving realistic thermophysical property variation.

The nucleation model of Quested and Greer is used in the front tracking model. Inoculant particles are distributed through the modeling domain with a uniform probability distribution. The inoculant size, which determines the potency of the seed particle, is set to follow a log-normal statistical distribution. The number of seed particles in the simulation can be increased to obtain higher inoculation levels.

Because microscopic detail of the crystal is ignored, the modeling technique provides results of the predicted grain structure in short run times with standard desktop hardware.

6. MACE Apparatus Model
In addition to the front tracking model of solidification, it is necessary to have a mathematical model of the MACE apparatus. As already mentioned, the axial temperature gradient and cooling rate of the MACE furnace was controlled in the section between heaters H1 and H3 (between 103mm and 163mm from the datum). This is the only section of the apparatus that we have modeled.

Fig. 2 shows the truncated section of the apparatus that we are interested in. The overall model domain is rectangular and it consists of three parts: the Al-Si rod, the alumina tube, and the nickel tube. Two boundaries in Fig. 2 are labeled B1 and B2. The boundary at the bottom B1 is at the colder end of the sample. The boundary at the top B2 is at the hotter end of the sample. B1 is at a distance 103mm from the datum; B2 is at a distance 163mm from the datum. Hence, the domain of interest is 60mm long.

Three circular markers in Fig. 2 show the positions of thermocouples built into the crucible walls. In the diagram, the thermocouples are at positions 113mm, 133mm and 163mm from the datum. The temperature readings taken at these thermocouple positions during the MAXUS 7 flight will be used to define the boundary conditions so that we can reconstruct the temperature conditions of the MACE experiments in the model.

The boundary condition at B1 was selected to be a Neumann boundary, that is, the temperature gradient at B1 was set in the model. Equation (1) gives Fourier’s law for the heat flux at a boundary.
\[
\frac{dT}{dy}\big|_{B_1} = -\frac{q}{K}
\]

where \( T \) is the temperature, \( y \) is the vertical coordinate, \( q \) is the heat flux at the boundary, and \( K \) is the thermal conductivity. Because the material changes along the boundary, the thermal conductivity of the control volume adjacent to the boundary changes in accordance with the material and the temperature of the control volume.

Equation (2) gives the heat flux, \( q_x(s) \), at a given \( x \) position along the boundary \( B_1 \). It is written more succinctly in the Laplace domain using the complex frequency variable, \( s \), instead of the time variable.

\[
q_x(s) = \frac{K_x}{K_{ref}} \left( e_1(s) \left[ k_P + \frac{k_I}{s} + \frac{k_D s}{1 + s^2} \right] \right)
\]

(2)

Here \( K_x \) is the thermal conductivity at the boundary at \( x \); \( K_{ref} \) is a reference value for the thermal conductivity; \( k_P \), \( k_I \) and \( k_D \) are the proportional, integral, and derivative terms of a PID feedback controller; and \( \tau \) is the break period of a first-order lag that helps to filter any noise on the differential channel.

The error term, \( e(s) \), is simply the difference between the temperature recorded at the 113mm-position, \( T^{\text{meas}} \) and the temperature computed at the same location within the model, \( T^{\text{comp}} \).

\[
e_1(s) = T^{\text{meas}} - T^{\text{comp}}
\]

(3)

Equations (2) and (3) describe a PID feedback control system that has been adapted to solve the inverse heat conduction problem. This formulation represents a dynamic inverse heat conduction method, thus no iteration during time steps is required. The PID terms, \( k_P \), \( k_I \) and \( k_D \) were selected by tuning the system response with the Ziegler-Nichols method.\(^{11}\) The value of \( \tau \) was selected as:

\[
\tau = 0.1 \frac{k_P}{k_D}
\]

(4)

The reference value for the thermal conductivity, \( K_{ref} \), was taken as the value of the thermal conductivity at the boundary \( B_1 \) at the radial position corresponding to the thermocouple position.

The boundary condition at the hotter end, \( B_2 \), was assumed to be a Dirichlet boundary condition. The temperature across the boundary is fixed at the same value as measured at the thermocouple located at the 163mm position, which is on the boundary \( B_2 \).

The heat flux across the crucible walls is separated into regions. The heat loss at the nickel tubing is simply described by the flux value of \( q_L \). The user can set this value once an estimate of the heat loss is made.

The heat flux at the heaters, \( q_H \), was automatically adjusted in the model so that we achieve the same temperature at the 133-mm position that was recorded at the same location by the thermocouple during the flight. Again using a PID control system we have the equation for the heat flux at the heater contact surface

\[
q_H(s) = e_2(s) \left[ k_P + \frac{k_I}{s} + \frac{k_D s}{1 + s^2} \right]
\]

(5)

where, in this case, the error term \( e_2(s) \) is the difference between the recorded thermocouple measurement and the computed temperature at the 133mm thermocouple position. Because a heater can only act as a heat source, equation (5) holds only if the calculated heat flux is greater than some passive heat loss value, that is, if \( q_H > q_L \). If equation (5) gives a value for \( q_H \) that is less than \( q_L \) then \( q_H \) is set equal to \( q_L \).

The heat flux was assumed constant across the surface of the heater in contact with the nickel tube. The PID coefficients of this controller are also tuned using the Ziegler-Nichols criterion. This control system model for the heater action is a realistic model of the controller used in the experiment. It is assumed that optimum tuning was achieved for the controller before the flight.

The boundary conditions on the crucible walls were assumed symmetric about the central axis, (see figure 2).

The thermal resistance of each control volume is given by the inverse of the conductivity. Between the Al-Si sample and the alumina tube we have assumed an additional thermal resistance due to a gap formation between the sample and the crucible. The resistance of this gap, \( R_{gap} \), must be set by user of the model. This additional thermal resistance is added in series with thermal resistance of the control volumes directly adjacent to the gap.\(^{12}\)

7. Model Parameters and Properties

The thermo physical properties for the Al-7wt%Si (both liquid and solid) were taken from the literature.\(^{13}\) The thermal conductivity (W/cmK) of the alumina crucible material and the nickel tube were given by the fourth-order polynomial in temperature

\[
K = a_1 T^4 + a_2 T^3 + a_3 T^2 + a_4 T + a_5
\]

(6)

For alumina \( a_1 = 2.7e-13, a_2 = -1.1e-9, a_3 = 1.6e-6, a_4 = -1.1e-3, a_5 = 0.39. \) For nickel \( a_1 = 4.1e-13, a_2 = -1.9e-9, a_3 = 2.9e-6, a_4 = -1.6e-3, a_5 = 0.95. \)

The specific heat capacity (J/cmK) for alumina was also taken as a fourth-order polynomial in Temperature with coefficients \( b_1 = -2.5e-12, b_2 = 8.9e-9, b_3 = -1.2e-5, b_4 = 7.5e-3, b_5 = 2.9. \) The specific heat capacity of nickel was taken as 3.916 J/cmK.

The parameters for the distribution of inoculant particle size were taken from literature.\(^{14}\) The number of seeds, \( N_{in} \), is quoted with each simulation run.

8. Results

The initial simulation has a low number of seed particles (\( N_s = 100 \)) distributed in the initial liquid part of the domain. This scenario is an approximation for MACE A where no grain refiner was added to the sample. This low number of particles may be present due to impurities in the sample. The gap between the
sample and the crucible was assumed to have a thermal resistance, \( R_{gap} \), of 11.28 cm\(^2\)K/W. The heat loss through the nickel outside diameter, \( q_L \), was set at a constant flux of 0.25 W/cm\(^2\). The temperature recordings for MACE A were used to apply the heat flux at the boundary \( B_1 \) and at the H2 heater contact surface. Fig. 3 shows the cooling curves from the model and compares them to the experimental recordings for MACE A. Agreement is good, especially at the hotter thermocouples.

Fig. 4 (a) shows the final predicted macrostructure. Here we can see the upper boundary of the TGZM in the sample domain. We can also observe a columnar region consisting of three columnar grains in the lower portion of the sample and an equiaxed zone in the upper portion. The upper boundary of the TGZM is predicted at 108.5mm. The CET occurs at approximately 143mm. These values can be compared to those presented for MACE A in table 2.

Fig. 4 (b) shows the resulting grain structure when the number of inoculant particles is increased (\( N_o = 500 \)). The cooling conditions from MACE B were used (similar conditions to those used in MACE A). Here we see that the columnar region was shortened and the CET occurred at a lower position of approximately 137mm. The grain density in the columnar and equiaxed zones increased also. If we make comparisons to the macrostructures recorded from the MACE B sample, we see that the predicted CET occurred close to the measured position of CET. However, the grain density predicted in the columnar region is lower than that measured in the experimental sample. Thus, a higher quantity of nucleated particles in the columnar region is required in the model.

Fig. 5 shows the predicted macrostructure when the MACE C cooling curves were imposed upon the model. The level of inoculation used is the same as that used in Fig. 4 (a) results. The TGZM occurred further along the sample than in Fig. 4 results. The position of the lower TGZM boundary with un-melted solid was also predicted in the result. The simulated macrostructure from the solidified region showed no clear CET. The model simulated a mixture of columnar and equiaxed grains.

9. Conclusions
Three Al-Si samples were solidified in microgravity on-board the MAXUS 7 sounding rocket. The samples were retrieved and analyzed for their grain structure. Two of the samples produced a CET; the third produced a mixed structure.

A model of the experiment was created that incorporated temperature measurements from the MAXUS 7 flight. A front-tracking model was used to predict the grain structures. The cooling curves predicted from the model are in good agreement with the temperatures measured on the flight. Some agreement is found between the predicted macrostructures and the observed ones; however, more investigation into the model results is required.

Some input parameters to the model were unmeasured and remain unknown. For the inoculant, accurate data on the number of seeds and the average diameter of the particles was unavailable. Comparisons of the
macrostructures from MACE A and MACE B experiments clearly demonstrated the significant effect of inoculation. For the furnace the heat loss at the walls and the gap’s thermal resistance were unknown. Thus, it is necessary to do a sensitivity analysis on the model by varying the aforementioned parameters. The influence of these parameters on the CET and the cooling curves can be observed through modeling.

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