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Arvind Prasad, Hani Henein, Charles-André Gandin. The Effect of Eutectic Undercooling on Microsegregation of Rapidly Solidified Al-Cu Droplets. *Journal Of The Japanese Society Of Microgravity Applications*, 2008, Proceedings of Third International Symposium on Physical Science in Space 2008, 25 (3), pp.449-452. <hal-01570949>

**HAL Id: hal-01570949**

**<https://minesparis-psl.hal.science/hal-01570949v1>**

Submitted on 1 Aug 2017

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# The Effect of Eutectic Undercooling on Microsegregation of Rapidly Solidified Al-Cu Droplets

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## Abstract

Al-Cu alloys of (nominal) compositions 4.3 wt%Cu, 5 wt%Cu, 10 wt%Cu and 17 wt%Cu were atomized using a drop tower type technique under N<sub>2</sub> and He gas. The atomized droplets were characterized using X-Ray tomography, Neutron Diffraction and Stereology calculations on Scanning Electron Microscope images. From the results of these experiments, the position of nucleus within an individual droplet, the volume percent of initial growth, recalescence and eutectic fraction were determined. Some of this data was used to initialize the solidification model developed for microsegregation during rapid solidification. The model solves the heat balance equation as the droplet loses heat to the gas in the atomizing chamber and phase transformation occurs during solidification. Starting with an off-center nucleation site and an initial user-defined undercooling for the nucleation of the primary phase, the model predicts the thermal history, fraction solidified and microsegregation in the droplet as a function of time. The calculated amount of eutectic in the solidified droplet is compared with the measured values. It will be shown that accounting for eutectic undercooling is critical to the agreement between experimental and predicted results.

## 1. Introduction

A project has been initiated to model microsegregation in industrially relevant alloys viz. Al and Ni based systems. The model aims to capture the effect of fluid convection on heat and solute transport during solidification. The experimental data required to validate this model would come from parabolic in-flight and ground-based levitation experiments and terrestrial atomization experiments.

Impulse Atomization experiments on Al-Cu binary alloys were carried out and a simple microsegregation model was developed at the Advanced Materials and Processing Laboratory, Canada. This paper presents the experimental results and the comparison between the experiment and the microsegregation model results.

## 2. Experimental

Impulse Atomization is a drop tube type technique wherein a liquid metal stream is pushed through a nozzle of known size. As the liquid emanates from the nozzle it enters a domain of quiescent gas where it disintegrates into tiny droplets. Subsequently, these droplets then fall through the stagnant gas, lose heat and solidify. The droplets attain a free-fall situation in the initial stages and therefore there is no gravity induced convection in these droplets. Thus solidification analysis of the atomized droplets provides an opportunity for providing solidification studies under reduced convection. The schematic of the atomization set up is shown in Fig. 1.

Al-Cu alloys of (nominal) compositions 4.3 wt%Cu,

5 wt%Cu, 10 wt%Cu and 17 wt%Cu were atomized under N<sub>2</sub> and He gas<sup>1</sup>. The solidified atomized droplets were characterized using X-Ray tomography, Neutron Diffraction and Stereology calculations on Scanning Electron Microscope images. The experimental measurements of the phase constituents in the droplets were compared with the predictions from a microsegregation model developed and described briefly below.

Tomography was carried out at European Synchrotron Radiation Facility (ESRF), Grenoble, France. The

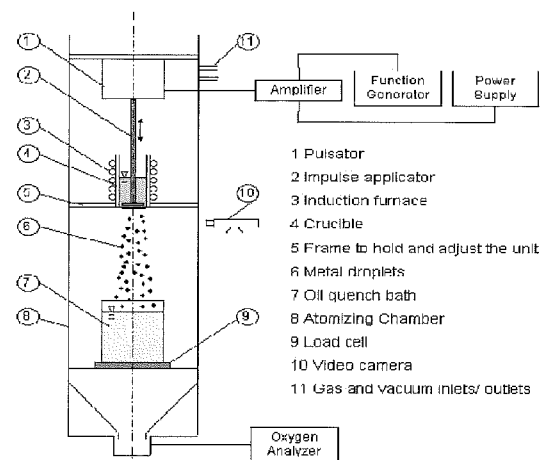


Fig. 1 Schematic of the Impulse Atomization unit. The numerical labels describe the different parts of the unit.

X-Ray beams used were monochromatic beam of  $30\text{keV} \pm 0.1\text{keV}$  energy. Tomography was carried out with a  $1\mu\text{m}$  resolution on a  $4.3\text{ wt}\%\text{Cu}$ ,  $660\mu\text{m}$  droplet atomized in  $\text{N}_2$  and  $17\text{ wt}\%\text{Cu}$ ,  $490\mu\text{m}$  droplet atomized in He. Only a single nucleation event was observed in each particle. Furthermore, a distinct gradation in length scale of the microstructure was seen in the particles which ranged from ultra-fine to coarse. The position of the nucleation site was calculated as well as the volume percent of finer and coarser structures was quantified and is shown in Table 1<sup>1,2</sup>. The gradation in the microstructure was related to the solidification during different regimes of droplet solidification i.e. nucleation, initial growth, recalescence, post-recalescence and eutectic solidification. The initial growth solidification occurs immediately after nucleation at some undercooled temperature. The dendrite growth rate is proportional to the undercooling and the presence of undercooling results in some finite growth rates of the dendrites. As the droplet continues to cool, the undercooling increases further. Consequently, the dendrite growth rate increases with increased rate of latent heat generation. At some point when the latent heat released is greater than the convective heat loss from the droplet, recalescence sets in, wherein the droplet temperature increases even during solidification. Since faster growth rates result in finer structures, therefore, the fine structure reported in the table is believed to correspond to the initial growth regime. Likewise, the relatively coarser region corresponds to the recalescence regime.

From the results of these experiments, the position of nucleus within an individual droplet, the volume percent of initial growth, recalescence and eutectic fraction were determined. Some of this data was used to initialize the solidification model developed for microsegregation during rapid solidification.

The results for weight percent  $\text{CuAl}_2$  from Neutron Diffraction (carried out at Chalk River, Canada) and volume percent eutectic from Stereology are given in Fig. 2a and 2b<sup>3</sup> respectively. Each data point in the

**Table 1** Volume percents of coarser and finer structures and position of the nucleation site (N) deduced from X-Ray Micro-Tomography.

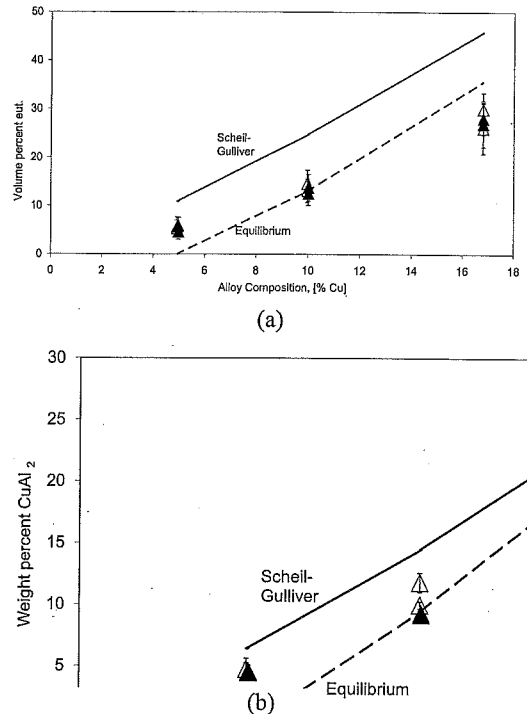
Sample (wt% Cu)	Atomization condition	N	Structure	
			Finer (%)	Coarser (%)
4.3	660 $\mu\text{m}$ , $\text{N}_2$ , 1123K	0.75 Rp*	0.1	19.4
			$\pm 0.09$	$\pm 0.6$
17	490 $\mu\text{m}$ , He, 1115K	0.63 Rp	14.5	33.2
			$\pm 2.6$	$\pm 2.3$

\*Rp – particle radius measured from centre to surface.

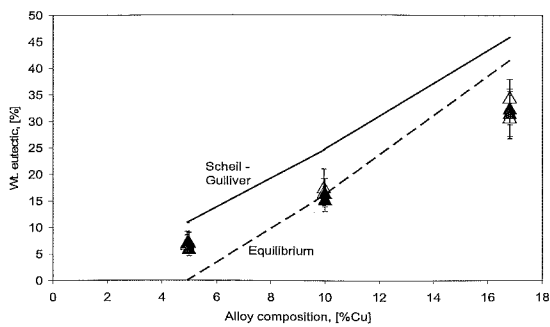
figure represents a droplet size atomized under a given gas type and therefore represents the effect of cooling rate. Solid data points represent He gas whereas the hollow points represent  $\text{N}_2$ . Scheil-Gulliver and equilibrium predictions are also presented in Fig. 2.

Fig. 2a shows the results obtained using Stereology measurements on 2-D section in an electron microscope while Figure 2b is the result of  $\text{CuAl}_2$  measured using Neutron Diffraction. Clearly, there is a distinct relative decrease in microsegregation with increasing composition as seen by the increasing difference between experimentally determined amount of eutectic or  $\text{CuAl}_2$  and the Scheil-Gulliver prediction. Furthermore, there is only a small effect of cooling rate on the microsegregation though it is not as significant as the effect of alloy composition on microsegregation. The effect of cooling rate is seen in the variance between the solid and hollow points in Fig. 2 for each composition.

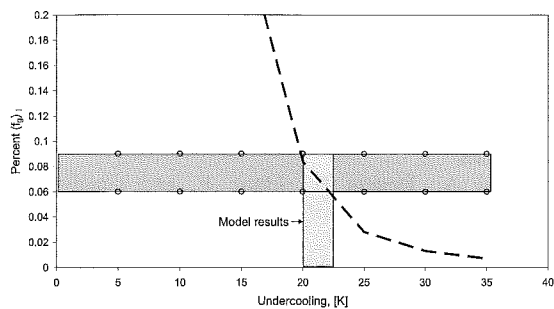
The microsegregation model developed gives weight percent eutectic and therefore to compare the model results with experimental results, it was necessary to convert the volume percent eutectic from Stereology measurements into weight percent eutectic. This was achieved by performing a mass balance of the solute (Cu) in the droplets<sup>3</sup>. The results from this conversion are shown in Fig. 3. The trend is the same as for volume percent eutectic and weight percent  $\text{CuAl}_2$  as expected.



**Fig. 2** (a) Volume percent eutectic and (b) weight percent  $\text{CuAl}_2$  plotted as a function of alloy composition. Closed and open points are for droplets atomized in He and  $\text{N}_2$ , respectively. The dashed lines are for the equilibrium prediction of microsegregation and the solid lines are the Scheil-Gulliver predictions.



**Fig. 3** Weight percent eutectic in the atomized droplets as a function of alloy composition. Solid points are for the He atomized droplets while the open points represent droplets atomized under  $N_2$ .



**Fig. 4** Comparison of volume fraction solidified in the initial growth regime between model and the experiment. Comparison shows that the droplets underwent approximately 20K of undercooling.

### 3. Model and predictions

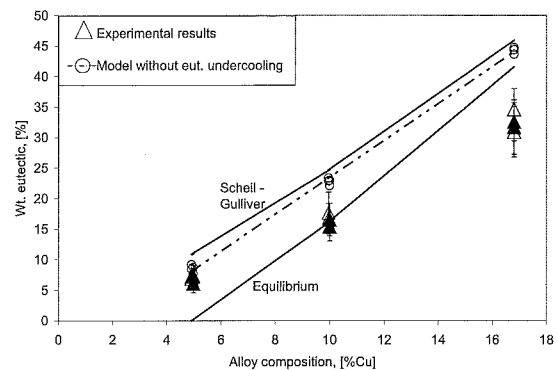
A droplet solidification model was developed to study microsegregation in the droplets. The droplet loses heat to the gas mainly by convection. The latent heat generation during solidification requires calculating the fraction solid formed as a function of time and is calculated using the Rappaz-Thévoz (R-T) model<sup>4,5</sup>. In this model nucleation occurs at a pre-selected undercooled temperature at the center of the droplet. Following nucleation, the dendrites grow equiaxially into the undercooled melt resulting in free (unconstrained) growth. The loci of the dendrite tips form the grain envelope  $R_g$  which grows at every time step. This dendrite growth is governed by the LKT dendrite kinetics<sup>6</sup>. The stability parameters in the LKT model were taken into consideration for the current model. This feature allows the model to take into account both low and high primary phase nucleation undercooling values.

However, for the current situation of comparison with atomization experiments, it was found that the droplets undergo a primary nucleation undercooling of approximately 20K. As the primary phase nucleation undercooling changes, the dendrite growth rate changes, and thereby changing the volume fraction of grain envelope in the initial growth regime. Thus a

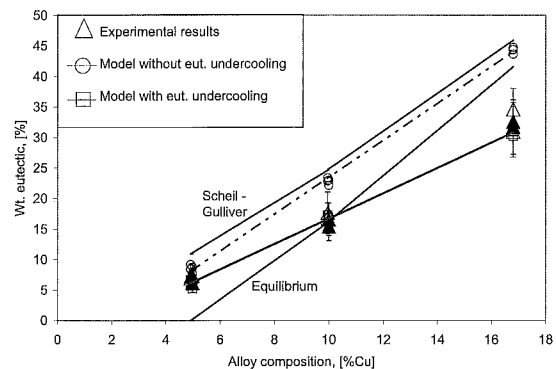
comparison of the model and experimental results for volume fraction solidified during the initial growth regime, provides an estimate of the primary phase nucleation undercooling in the droplets. The volume fraction of initial growth regime quantified from the tomography experiments and the volume fraction of grain envelope  $R_g$  in the initial growth regime obtained from the model were compared. **Fig. 4** shows the comparison and the approximate primary phase nucleation temperature. The result is for the 660  $\mu m$ , Al4.3wt%Cu droplet atomized in  $N_2$ .

LKT dendrite kinetics also requires phase diagram information (partition coefficient). In addition, a linear Al-Cu phase diagram was assumed. From the temperature evaluated at each time step, the phase diagram is also used to get information on the liquid and solid phase solute concentration. Finally, the original R-T model with nucleation taking place at the center of the grain was modified<sup>7</sup> to include an off-center nucleation site as was observed from tomography (Table I).

The model results are shown in **Fig. 5** and **Fig. 6**. **Fig. 5** shows the model prediction compared to the experimental values for weight percent eutectic. In this case the simulations were run with eutectic occurring at



**Fig. 5** Comparison of weight percent eutectic results from the model and the experiment. Eutectic was assumed to take place at the equilibrium temperature.



**Fig. 6** Comparison of weight percent eutectic results from the model and the experiment with eutectic undercooling taken into account in the model simulations.

the equilibrium temperature. As can be seen, the model prediction deviates from the experimental results as the alloy composition is increased. Herlach et al<sup>8)</sup> have recently performed levitation experiments on Al-Cu droplets and observed eutectic undercooling in these droplets under unseeded undercooling conditions. The alloy compositions used in their experiments were Al-4wt%Cu, Al-14wt%Cu and Al-24wt%Cu. The eutectic undercooling values they measured and reported were 28.5, 40.3 and 66.8K respectively for the three compositions. Clearly, the eutectic undercooling increased with the alloy composition. For the droplets in this work, eutectic undercooling values were interpolated for compositions of 5, 10 and 17 wt%Cu. Thus the eutectic undercooling values obtained were 29, 33.8 and 46.7K respectively for the three alloy compositions.

These eutectic undercooling values were used in the current model. This was done by using the undercooled eutectic temperature in the model and assuming that the remaining liquid at that temperature undergoes complete eutectic transformation isothermally. Fig. 6 shows the simulation results with eutectic undercooling incorporated. As can be clearly seen, the model results compares favorably with the experimental measurements only when eutectic undercooling is included in the model.

#### 4. Conclusions

Results from novel experimental techniques and model results were combined to study the rapid solidification under unconstrained growth conditions. Volumetric 3-D information was obtained using X-ray micro-tomography and neutron diffraction. The experimental results show that alloy composition plays

a more important role in the scale of the microstructure and the quantity of these fine scale structures than cooling rate. The model incorporates the position of the nucleation site.

The model with no eutectic undercooling calculates reasonably well the different regimes (Initial Growth, recalescence and eutectic) as seen experimentally for the low alloy binary system (4.3wt% Cu). However, with increasing alloy composition the model with no eutectic undercooling overpredicts microsegregation. When eutectic undercooling is accounted for, excellent agreement is obtained between model and experimental results.

#### Acknowledgments

The authors would like to acknowledge the grants received from Canadian Space Agency (CSA) and European Space Agency (ESA) for this work. We would also like to acknowledge the support from Killam Fellowship received by Dr. Hani Henein.

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