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QUATIFICATION OF PRIMARY PHASE UNDERCOOLING OF RAPIDLY SOLIDIFIED DROPLETS WITH 3D MICROTOMOGRAPHY

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Abstract

Powders of different compositions of Al-Cu alloys were atomized in helium and nitrogen and the microstructure of the atomized droplets was examined using X-ray micro-tomography. A method was developed to remove X-ray artifacts and background noise from the particles images. The method developed involves creating a clean mask file using MATLAB image toolbox, followed by applying the mask file to the original image to achieve clean images for the particle of interest. Separate features of interest in the droplets, such as region of initial growth and primary dendrites, were investigated at the various stages of solidification. The data is used to estimate the primary phase undercooling of the droplets, which will be used in a solidification model as an input to estimate the phase fractions. The results will then be compared with the experimental results.

Introduction

Rapid solidification techniques are of great importance due to their ability to produce various structures which are not possible to achieve in conventional solidification processes [1]. Characterization techniques such as 2D microscopy can be used to provide valuable information about solidification microstructure, but they are limited by the number of sections that can be studied from a sample; therefore, 3D visualization techniques have been utilized to gather information about the entire solidified volume. X-ray tomography is one such 3D visualization technique in which the X-ray beams from a synchrotron facility are used to construct the 3D image of the solidified body. Prasad et al. [2] used this technique to study solidification microstructure of Impulse Atomized particles of Al-5wt%Cu and Al-17wt%Cu alloys. Using this technique, nucleation sites in different particles were identified and recalescence boundaries were seen in the vicinity of the nucleation sites [3]. In this paper a methodology will be described to estimate the primary phase undercooling from the volume fraction of the recalescence region measured using tomography data.

Experimental Procedure

All particles studied in this research were produced using Impulse Atomization (IA) technique. IA is a single fluid atomization technique in which the molten metal is pushed through several orifices by applying recurring forces. As a result, liquid ligaments emanate from the orifices and fall into a drop tube filled with inert gas. The ligaments then break into spherical droplets due to Rayleigh instability, and lose heat to the surrounding gas while falling. Eventually, the solidified droplets are collected at the bottom of the tube. IA has been used to produce rapidly solidified metallic (or non-metallic) powders. This process can reproducibly yield powders of uniform size-distribution with cooling rate as high as 10^5 K/s [4]. For this research, powders of Al-5wt%Cu and Al-17wt%Cu were produced using nitrogen as cooling gas.

X-Ray beams from synchrotron facilities were used to carry out computer tomography work. Observations were made on the droplets using X-ray beams of 1 μm resolution at European Synchrotron Radiation Facility (ESRF, France) and 0.37 μm resolution at Swiss Light Source (SLS, Switzerland). This non-destructive technique generated RAW files for the 3-dimensional section of the droplet. The size of each RAW file ranges from 1GB to 16GB. Analysis of the data sets involves the following steps:

1. Generate a histogram of the data
2. Locate the minimum data value between peaks. This corresponds to particle and exterior ranges of data values.
3. Do a thresholding operation on the data using the value from step 2. This generates a new data set contain 2 values, one for the particle and one for the exterior. Note that this new data set contains a variety of structures which must be cleaned.
4. Clean the thresholded data set by locating connected regions and filling in the small structures, both on the inside and outside. The result is the mask file which allows for separating the original data into particle and non-particle regions. This operation is done on each slice of the data set using 2D connection information. For more details of the technique used see [5].

Results and Discussion

The crystallization of an undercooled melt occurs in two steps. Once nucleation has initiated solidification, the subsequent growth of the solid phase leads to the release of the heat of fusion ΔH_f. In the case of rapid crystal growth a steep rise of temperature takes place, called recalescence. Assuming that solidification during recalescence occurs under near-adiabatic conditions the volume fraction solidifying during recalescence under non-equilibrium conditions can be determined by Eq. 1 [6].

$$f_R = \frac{\Delta T}{\Delta T_{hyp}} \quad (1)$$

where $\Delta T_{hyp} = \frac{\Delta H_f}{C_p^l}$ is the hypercooling limit and C_p^l is the specific heat of the liquid. Prasad et al. [3] using X-ray tomography for particles of Al-Cu of both compositions identified the region of initial growth and recalescence boundaries (primary trunks). Also, the volume fraction of these regions in the droplets was evaluated. There were differences in the volume fraction of these regions observed in the Al-5wt%Cu and Al-17wt%Cu. This was believed to be caused by the difference in growth rates between the two compositions for a given undercooling.

Figure 1 shows a 2D image from tomography on a particle of Al-5wt%Cu. The regions of ultra-fine structure, representative of nucleation region, and the primary dendrite trunks are highlighted in the figure.

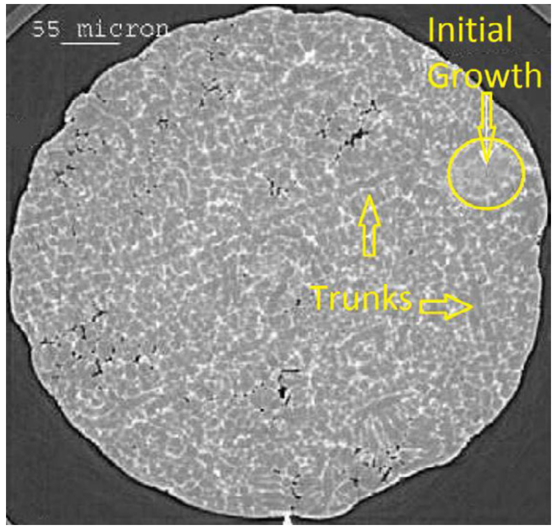

Figure 1. A 2D image from tomography on a particle of Al-5wt%Cu. The regions of ultra-fine structure and the primary dendrite trunks are highlighted in the figure.

Using stereological analysis [7] on the tomography images, the average volume fraction of nucleation and recalescence region within the particle was quantified for all the droplets (one Al-5wt%Cu droplet and four Al-17wt%Cu droplets). The calculation was done using the Cavalieri method [8]. It basically involves selecting the volumetric region containing the region of interest within the droplet and dividing that region into a fixed number of slices of known thickness. The area fraction of the feature of interest on each slice is then measured and the volume of that region for each slice is given by the product of area fraction and the slice thickness. A manual point count technique was employed for measuring the area fraction on each slice. Finally, the summation of the volume fraction for each slice gives the total volume fraction of the recalescence region for the entire droplet. Table 1 presents the results of recalescence volume fraction measurement, as well as corresponding calculated undercooling. Also, the properties of alloys used for this calculation are shown in Table 2.

Table 1.Measured recalescence volume fraction and corresponding calculated primary phase undercooling.

Droplet Details	Al-5wt%Cu	Al-17wt%Cu (1)	Al-17wt%Cu (2)	Al-17wt%Cu (3)	Al-17wt%Cu (4)
Diameter (μm)	660	480	490	450	420
Recalescence vol. %	19.4 ± 0.6	27.8 ± 2.4	33.3 ± 2.3	44.1 ± 2.2	32.1 ± 3.1
Primary phase undercooling (K)	64.9 ± 2.0	94.6 ± 8.2	113.3 ± 7.8	150.0 ± 7.5	109.2 ± 10.6

Table 2. Properties of alloys used in the calculation of primary phase undercooling.

Alloy properties	Al-5wt%Cu	Al-17wt%Cu
Density (kg m^{-3})	2760	2870
Specific heat (J $K^{-1}kg^{-1}$)	1176.3	1152.9
Latent heat (J kg^{-1})	393699.3	392247.4

The primary phase undercooling values shown in Table 1 were used in a solidification model to predict the phase fractions after solidification of IA particles. The details of the solidification model can be found elsewhere [9, 10]. In brief, the model assumes a uniform temperature for the entire droplet. The droplet starts falling at the atomization temperature with an initial velocity of

0.5 m/s. While the droplet continues to fall through the quiescent gas, it loses heat mainly through convection. At some user defined temperature, the nucleation of a microstructure is assumed to occur in the droplet center and to develop by radial growth of a spherical zone. At first, a dendritic microstructure is modeled, followed by a eutectic microstructure. This diffusion-based microsegregation model is coupled with diffusion in the extradendritic liquid following the model proposed by Wang and Beckermann [11]. Solidification results in latent heat release proportional to the solidification rate, and as a result the net heat content of the droplet is given by the difference between the latent heat generated and the convective heat loss. The latent heat is released until solidification is complete. Prasad et al. [10] using the interpolation of the eutectic undecoolings reported in [9] found values for the primary phase and eutectic undercooling (see Table 3). Then they used those values as input for the solidification model. The model's outcome for weight percent of eutectic microstructure was then compared with that from the experiment and reasonable match between prediction and measurements was achieved.

Table 3. Values of primary and eutectic undercooling used in [10].

Dendritic nucleation undercooling, ΔT_N^p [K]		15
Eutectic nucleation undercooling, ΔT_N^e [K]	5wt%Cu	20
	10wt%Cu	20
	17wt%Cu	27.5

They also showed that the primary phase undercooling plays minor role in the phase fractions observed in the solidified particles. However, as it is shown in Table 1 the primary phase undercooling achieved was significantly higher than that was predicted by the previous authors (Table 3). The effect of this increase in the amount of primary phase undercooling is discussed next.

The particle size and primary phase undercoolings for the 4 particles of Al-17wt%Cu were averaged (116.8K) and used as input for the mathematical model. Also, the standard deviation of this calculation was used to find lower and higher limit for primary phase undercooling (i.e. 95K and 138K). To calculate the eutectic fraction, several different values for eutectic undercooling were also used. For Al-5wt%Cu, the eutectic undercooling reported in [10] was used along with the primary phase undercooling found in Table 1. However, for Al-17wt%Cu, the amount of eutectic undercooling used in [10], i.e. 27.5K, along with any of the primary phase undercoolings shown in Table 1 will result in complete suppression of the primary phase. This shows that the eutectic started to occur at lower temperatures than that assumed in [10]. Table 4 and 5 show the modeling results for eutectic weight fraction in Al-5wt%Cu and Al-17wt%Cu, respectively. If one does not take into account the 100 % eutectics values as they are not relevant and also does not take into account the 0 undercooling because it is expected to be some, the average wt% eutectic is 27.9% ± 2.8% for Al-17wt%Cu and is 6.3 ± 0.4% for Al-5wt%Cu. One then can average the corresponding undercooling values for primary undercooling and eutectic undercooling. The calculated values from Table 4 and 5 were then compared with that from experimental, reported in [10], equilibrium and Scheil prediction in Figure 2. From this figure it is evident that for Al-5wt%Cu, the eutectic fraction is not significantly sensitive to any of the primary phase and eutectic phase undercooling values and the model can closely predict the phase fractions. For Al-17wt%Cu, however, it seems that using 95 and 50K undercooling for primary and eutectic undercooling, respectively, results in closest eutectic fraction to that from experimental measurements. While no undercooling for both primary and eutectic phase results in overestimation of eutectic fraction, increasing the amount of undercooling for both primary and eutectic (above 95 and 50, respectively) resulted in underestimation of eutectic fraction.

Table 4. Undercooling values used as input for simulation and corresponding results of calculated primary phase and eutectic fraction for Al-5wt%Cu.

Diameter=660μm	Gas=N2		
Primary undercooling (K)	**Eutectic undercooling (K)**	**wt% primary phase**	**wt% eutectic**
0	0	92.8	7.2
0	20	93.1	6.9
65	0	93.7	6.3
65	20	94.0	6.0

Table 5. Undercooling values used as input for simulation and corresponding results of calculated primary phase and eutectic fraction for Al-17wt%Cu.

Diameter = 460μm	Gas=He		
Primary undercooling (K)	**Eutectic undercooling (K)**	**wt% primary phase**	**wt% eutectic**
0	0	64.3	35.7
95	50	68.1	31.9
95	60	69.6	30.4
95	70	71.3	28.7
116	50	0.0	100.0
116	60	70.3	29.7
116	70	72.0	28.0
116	80	73.9	26.1
138	70	0.0	100.0
138	80	74.8	25.2
138	90	76.5	23.5

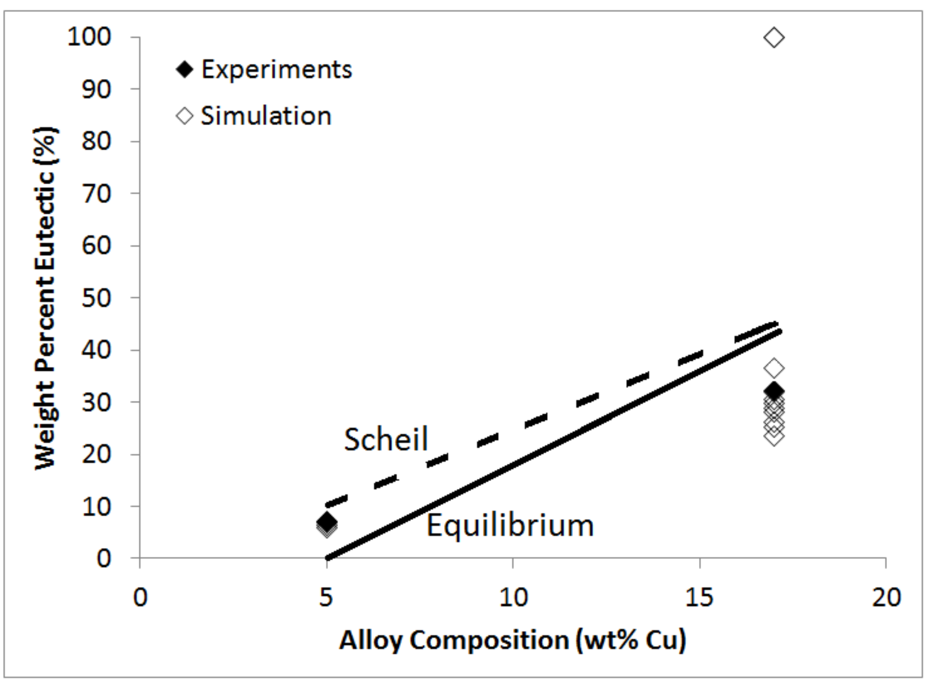

Figure 2. Comparison of weight fraction of eutectic from experimental with that from modeling, Scheil-Gulliver and equilibrium. Al-5wt%Cu particles were atomized in nitrogen and particles of Al-17wt%Cu were atomized in helium.

On the other hand, the difference between the experimental values and those predicted by Scheil-Gulliver and equilibrium shows that the primary phase and eutectic undercooling play a very important role in the final percent of eutectic in the droplets.

In order to reduce the variations in the measured volume percentage of recalescence region, higher resolution tomography is beneficial. Also, Focused Ion Beam (FIB) is being used to create multiple slices of region of interest at higher resolution.

Summary

X-ray tomography was used to study the 3D microstructure of Al-5 and 17 wt%Cu powders. The volume fraction of recalescence region was measured and used to estimate the primary phase undercooling. It was found that the particles with higher Cu content achieve higher primary and eutectic undercooling. The results were then used as input in a solidification model to estimate the weight fraction of phases formed during solidification. Comparison between model prediction and experiments is performed considering the eutectic percent. It is found that a reasonable match between prediction and measurements can be reached by using certain levels of primary and eutectic undercooling

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