In situ solidification of eutectic Al-33wt%Cu droplets

J Valloton[1,a], N Mahdi[2], L Rabago[1], J Chung[1] and H Henein[1]

[1] Advanced Materials and Processing Laboratory, University of Alberta, Edmonton, Canada
[2] Norcada inc., Edmonton, Canada
[a] Corresponding author: valloton@ualberta.ca

Abstract. Al-33wt%Cu eutectic droplets were rapidly solidified using Impulse Atomization, a drop tube technique. The samples were then processed in situ in an SEM using an HTN-0101 MEMS heating chip manufactured by Norcada. The temperature of the chip, as well as the heating and cooling rates, can be easily set using the chip's control software, allowing for heating and cooling rates as fast as 1000°C/second. The droplets were heated above the eutectic melting temperature. Thanks to the oxide skin, the droplets retained their spherical shape, which allowed in situ solidification experiments. The live feed of the surface of the droplet obtained with the SEM was recorded during the experiments alongside the temperature-time profile. Various cooling rates were imposed and the liquid samples were shown to undercool prior to solidification. The resulting eutectic morphologies and spacings were then analyzed as a function of the cooling rates and undercoolings.

1. Introduction

High cooling rates and undercoolings stemming from rapid solidification of metallic alloys results in several favourable microstructural features. These include microstructure refinement, minimal segregation, solubility extension and the formation of metastable phases [1-6]. Thus, it is necessary to understand the relationship between the solidification parameters and the resulting microstructures in order to control the solidification path and resultant mechanical properties. However, it is sometimes difficult to relate the observed microstructure with process parameters due to experimental difficulties. These include measurements of critical variables such as cooling rate, undercooling and solidification rate, which are known to control the final solidification microstructure. Therefore, to understand the phenomena at play, solidification experiments with controlled parameters are useful. This contribution presents preliminary results of in situ solidification of rapidly solidified Al-33wt%Cu eutectic droplets.

2. Experimental method

Impulse atomization (IA) (a type of drop tube) is a containerless solidification technique (Figure 1) [7]. It consists in the transformation of a bulk liquid into a spray of liquid droplets of fairly uniform size. In this study, 300g of high purity Al (99.99%) and commercial purity Cu (99.9%) is added to a dense graphite crucible and melted by induction heating at a temperature 750°C (about 200°C above the equilibrium eutectic temperature of 548°C). The melt is held for 1 hour under argon atmosphere to ensure proper mixing. A plunger applies an impulse to the melt in order to push it through a nozzle plate with several orifices of known size and geometry. Liquid ligaments come out of each orifice and in turn break up into droplets by Rayleigh instability. Rapid cooling of the droplets then occurs during free fall by heat loss to the surrounding inert gas (Ar in this experimental run). The solidified samples are collected at the bottom of the atomization tower and subsequently sieved into different size ranges. A detailed description of the process is available in [7].

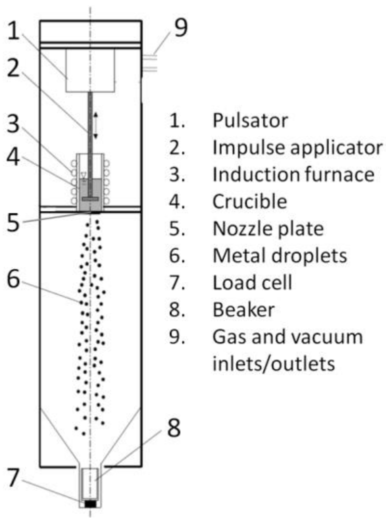

Figure 1: Schematic view of an Impulse Atomization apparatus [1].

One Al-33wt%Cu atomized droplet of diameter less than 90 µm is then mounted on an HTN-0101 MEMS heating chip designed and manufactured by Norcada (as close to the center as possible, Figure 2). The chip is inserted in a Norcada MEMS in situ heating stage made for ZEISS scanning electron microscopes (NHB-Z-SNL-001). The temperature of the chip, as well as the heating and cooling rates, can be easily set using the chip's control software, allowing for heating and cooling rates as fast as 1000 K/s. In this works, the Al-33wt%Cu were heated to 550°C at a rate of 2K/s and subsequently cooled down to 400°C at rates of 5, 10 or 20 K/s. The temperature-time profile is recorded during the in situ experiments alongside the SEM live feed of the surface of the droplet. This experimental setup enables to follow dynamically the change in the microstructure with time and ensures a stable and ultra-low drift in the target temperature.

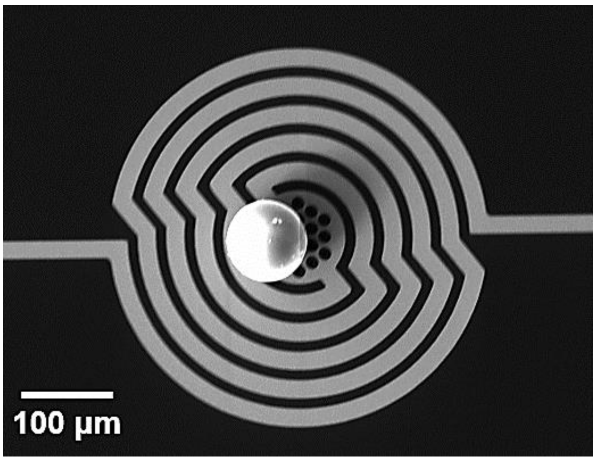

Figure 2: SEM view of Norcada HTN-0101 MEMS heating chip with an Al-33wt%Cu droplet placed at the center for in situ experiments.

3. Results and discussion

Figure 3a shows a snapshot of the SEM live feed after heating of the droplet above the eutectic temperature. Thanks to the oxide skin, the liquid does not spill over and the droplet becomes fully spherical. After cooling, the droplet has fully solidified, with visible solidification shrinkage (Figure 3b). The eutectic microstructure at the surface of the sample is clearly apparent, which allows for quantification. The dark grey phase is the α-Al solid solution while the lighter phase is the Al_2Cu intermetallic. Impurities on the surface of the samples are sometimes observed, but they do not seem to act as nucleation points.

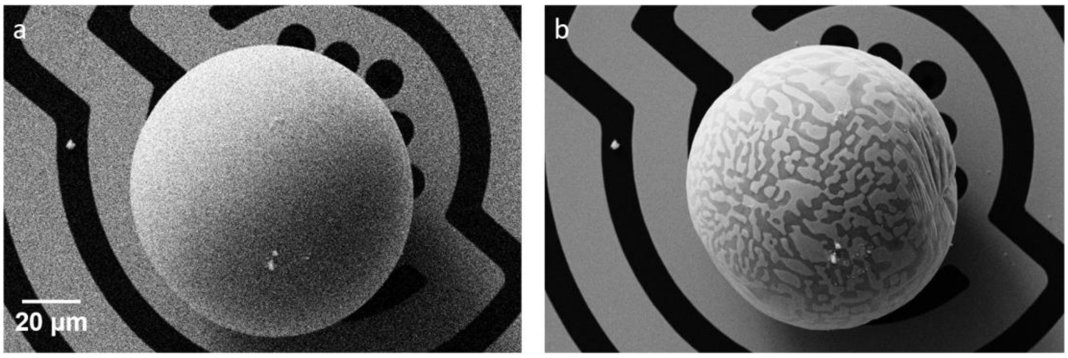

Figure 3: SEM micrographs of an Al-33Cu droplet when fully liquid (a) and after solidification (b).

Figure 4 (left) shows a typical temperature-time profile obtained during an in situ solidification experiment. In this case, heating of the droplet to 550°C was performed at a rate of 2 K/s and cooling to 400°C at a rate of 5 K/s. The temperature acquisition rate of the heating chip is 1 Hz. In all heating cycles, melting of the droplets was observed at 540±2°C. Even though this temperature is slightly lower than the equilibrium eutectic temperature (548°C), it can be used as a reference to see if the samples undercool prior to solidification. On the other hand, the nucleation temperature can only be identified approximately, especially at the higher cooling rates. To estimate this value, the frame from the recorded video feed that contains the first indication of solidification is identified and related to the frames corresponding to the temperature acquisitions directly prior to and following solidification (all the analyzed droplets fully solidified in less than a second). The nucleation temperature is then determined using a linear fit (Figure 4 (right)).

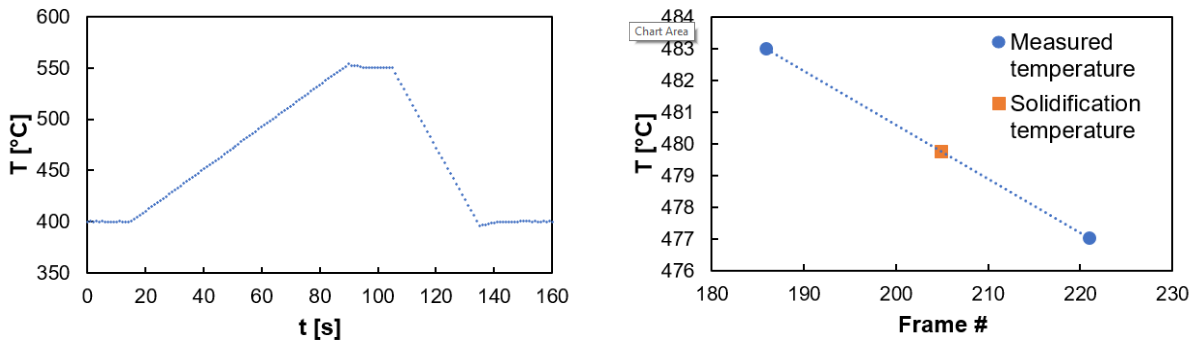

Figure 4: Left: temperature-time profile of a droplet heated at 2 K/s and cooled at 5 K/s. Right: estimation of solidification temperature

Figure 5 shows the surface of Al-33wt%Cu samples that have been solidified in situ with imposed cooling rates of 5, 10 and 20 K/s (a, b and c respectively). Looking at the solidification temperatures, all droplets experienced a certain degree of undercooling prior to nucleation, which increases with the cooling rate (Table 1). However, due to the small size of the droplets, no recalescence event is visible on the temperature-time profiles. Lamellar spacings were measured on the 3 samples shown in Figure 5. To minimize measurement errors due to the near spherical nature of the samples, measurements were only carried out at the center of the samples. As expected, the lamellar spacing decreases as the cooling rate and the undercooling increase. Further experiments will be carried out in order to obtain different undercooling values for a same cooling in order to clarify the role of undercooling on the refinement of the microstructure. Interestingly, the sample with the coarsest eutectic (Figure 5a) does not exhibit a typical lamellar eutectic structure. Some regions see large chunks of Al_2Cu while others are rod-like.

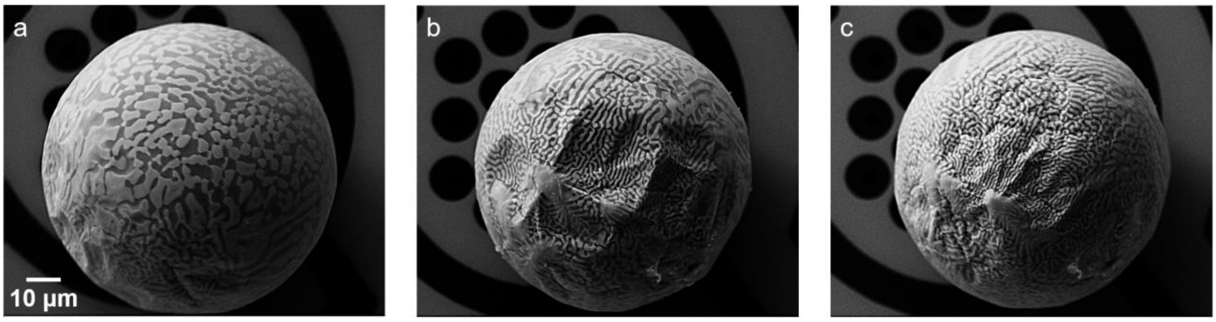

Figure 5: SEM micrographs of Al-33Cu droplets solidified at 5 K/s (a), 10 K/s (b) and 20 K/s (c).

Table 1: Cooling rate, undercooling and lamellar spacing of the samples shown in Figure 5.

Sample	Cooling rate [K/s]	Undercooling [K]	Lamellar spacing [μm]
a	5	68	3.3 ± 0.6
b	10	75	1.5 ± 0.2
c	20	97	1.0 ± 0.2

4. Conclusions

In situ experiments were carried out on Al-33wt%Cu impulse atomized droplets. The samples were processed in a scanning electron microscope using Norcada MEMS heating stage. When the sample is brought over the eutectic temperature, the oxide skin allows the liquid droplet to keep its spherical shape. This enables in situ solidification experiments. First tests show that the droplets undercools prior to solidification. The undercoolings increase as the imposed cooling rates increase. The resulting microstructure is thus refined.

The Norcada stage for MEMS in situ heating and cooling of samples in a scanning electron microscope shows great potential for aging and solidification experiments that will be explored further.

Acknowledgments

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