In-situ characterization of droplets during free fall in the drop tube-impulse system

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Abstract. Powders of copper were produced using a drop tube-impulse atomization technique. In this system, molten metal is pushed through orifices, forming ligaments, which eventually break down and spherodize into droplets. A 3-D translation stage was designed, constructed and installed in the drop tube to allow for measurements of velocity and droplet size in flight using a Shadowgraph and radiant energy using DPV-2000. A mathematical model of the evolution of droplet velocity and temperature for different sized copper droplets at various heights was developed. The experimental results from the Shadowgraph and the DPV-2000 are compared to the model’s results. In addition, the extent to which microgravity prevails during flight and droplet solidification was investigated by using the model and the Shadowgraph results. It was found that the acceleration of falling droplets near the melting point is close to gravitational acceleration and as a result the falling droplets do not reach their terminal velocity at their melting point. The results of in-situ measurements during the atomization of copper showed that the larger droplets have higher radiant energy than that of the smaller ones. Correlation between experimentally measured radiant energy and predicted temperature of falling droplets will be investigated. The current work is part of the NEQUISOL project supported by ESA within contract number 15236/02/NL/SH and CSA within contract number 9F007-08-0154 and SSEP Grant 2008.

1. Introduction
Solidification of alloys is a complex phenomenon arising in many modern experimental techniques and industrial technologies related to casting and surface processing. The variation of different solidification conditions (such as undercooling or cooling rate) provides the possibility to control the morphology and size of crystal structures, which substantially influences physical and chemical properties of alloys; more specifically, deep undercooling of alloys below the equilibrium liquidus and eutectic temperature results in rapid solidification and yields materials with improved mechanical, magnetic and electrical properties. Deep undercooling results in metastable phases or in high growth velocities which in turn leads to a finer microstructure.

A number of models have been reported for predicting or estimating the undercooling temperature of droplets in gas and single fluid atomization systems as well as drop tubes [1-3]. There is no feasible means available yet to directly validate these models. An indirect method of validation was presented by Prasad et al [4]. In this approach, microtomographic images are analyzed in order to locate the region of initial growth in a particle. The volume of this region is measured and compared with predictions.
obtained using a microsegregation model incorporating a variable undercooling of the primary phase [5, 6]. This approach has led to estimations of primary phase undercooling of about 15 to 20°C in Al-Cu droplets. Eutectic undercooling has also been estimated by measuring the fraction of eutectic in the solidified structure either using SEM images or neutron diffraction [5, 7]. The measured quantity is compared with the respective phase diagram of the system under study. Solidus and liquidus lines are extended until agreement is found between measured eutectic fraction and the extended lines on the phase diagram. The eutectic temperature corresponding to this agreement is taken to be the nucleation temperature of the eutectic. Gandin et al [8] reported temperature measurements of a droplet levitated in an electromagnetic field using a high speed pyrometer. They clearly report measurements on an Al-24 Cu droplet of 6 mm diameter of primary phase and eutectic undercooling.

A systematic effort on the investigation of thermal history of falling droplets in-situ during atomization is rare.[9] In addition to the thermal history, in-situ measurement of the droplet diameter, initial velocity and instantaneous velocity is crucial in validating solidification models. This study reports on an effort to make such measurements on droplets in flight and in situ during the atomization of copper. Two measuring instruments were used: a Shadowgraph to measure droplet velocity and diameter and a DPV-2000 (a two color pyrometer) for droplet radiation measurements.

One of the important parameters affecting the thermal behavior of falling droplets during atomization is the amount of microgravity achieved. It has been reported that the level of microgravity achievable using drop tube technique is relatively high, i.e. up to 1×10^5 of the earth’s gravity [10]. The problem involved with the drop tube system is that the duration of microgravity happens in the drop tubes is short and many of the samples cannot solidify during their free fall in an evacuated drop tube. For example, the temperature of a 2 mm droplet of aluminum with the superheat of 0.1T_m (melting point) decreases only 19K during 50 meters falling height [11]. Adding gases to the tube ensures us that the solidification of the droplets happens in the reasonable falling height. But, this can deteriorate the low gravity condition. In this situation, smaller droplet size, higher gas pressure and faster initial falling velocity result in lower microgravity condition. Drop-tube-Impulse Atomization is a method in which atmospheric gas pressure is used and it provides a containerless solidification environment for the droplets to solidify with a high cooling rate.

In this study, a correlation between radiant energy and temperature of falling droplets using experimental and a mathematical code will be studied in order to investigate the sensitivity of DPV-2000 for measuring undercooling during droplet solidification. Furthermore, the microgravity condition of the falling droplets in the drop-tube impulse atomization system will be investigated using the same code and the experimental velocity results.

2. Experiments

Copper shots with 99.99% purity (manufactured by Alfa Aesar) were melted in an induction field of a drop tube-impulse atomization technique to produce copper powder. In this system, molten metal is pushed through orifices, forming ligaments, which eventually break down and spherodize into droplets according to Rayleigh instability [12]. After heating the copper shots to superheat of 1400°C inside a graphite crucible using an induction furnace, atomization was then started. During atomization, the liquid copper is pushed through 37 orifices with the diameter of 400 μm on a nozzle plate at the bottom of the crucible via a vibrating plunger. Falling droplets were cooled in argon atmosphere having a maximum oxygen content of 90 ppm and lose heat to the surrounding gas.

A 3-D translation stage was designed, constructed and installed in the drop tube to allow for measurements of radiant energy using a DPV-2000 (Tecnar Automation Ltée) and velocity and droplet size in flight using a Shadowgraph (Sizing Master Shadow from LaVision GmbH in Gottingen, Germany).

DPV-2000 is an optical sensing device based on a patented technology developed by the National Research Council of Canada. DPV-2000 uses a dual slit optical sensor that can measure the radiant
energy of up to 800 droplets per second with the depth of field of 1.9 mm, and it can measure radiant energy of droplets at two different wavelengths [13].

Sizing Master Shadow from LaVision is using the shadowgraph technique (backlighting) to visualize droplets for image analysis. A light source, in this case a pulsed laser combined with the diffuser optics, illuminates in-flight droplets 5 times per second. Backlight of droplets inside the measurement volume of 6×6×6 mm is captured by a high resolution imaging system consists in a high-speed camera. It is possible to investigate droplet sizes down to 5μm using the shadowgraph device. Figure 1 shows a schematic of the shadowgraph system [14].

Particle size distribution of falling droplets (D50 and D90) and the velocity were measured via the shadowgraph device. Also, using the 3-D stage, the droplet size, velocity and radiant energy can be measured at different distances from the nozzle plate. In this study, these measurement instruments were located at 10 cm below the nozzle plate. The focal point of both shadowgraph and DPV-2000 was set to an imaginary vertical line going through the center of the nozzle plate; therefore, any unfocused droplets would be rejected by the criteria set by the operator in the respective software of each instrument.

3. Modeling

In order to verify the experimental results, a mathematical model of the evolution of droplet velocity and temperature for different sized copper droplets at different heights was developed. The model calculates the droplet velocity and temperature as a function of time and distance traveled. For the purposes of this paper, the cooling and velocity of droplets up to their melting point is considered.

Droplets begin their downward trajectory with an initial velocity. The subsequent trajectory of the droplets depends on this initial velocity, and the forces of gravity, buoyancy and drag as shown schematically in Figure 2.
Applying Newton’s Second Law on the droplet, the instantaneous acceleration can be found as follows [15].

\[
\frac{dv}{dt} = g - \frac{3 \rho_g C_d d}{4 \rho_p d} v^2
\]  

(1)

where \( v \) is the relative velocity between the droplet and the atomization gas, (which in the case of Impulse Atomization \( v_{gas}=0 \)) and \( \rho_p \) and \( \rho_g \) are the densities of the droplet and atomization gas, respectively. The gravitational acceleration is given by \( g \) and \( d \) is the droplet diameter. The discharge coefficient, \( C_d \), for droplets in laminar flow is given by: [16]

\[
C_d = \frac{18.5}{Re^{0.5}}
\]  

(2)

The Reynolds number, \( Re \), in Equation (2) is given by:

\[
Re = \frac{\rho_g v d}{\mu_g}
\]  

(3)

where \( \mu_g \) is the gas viscosity.

To calculate the thermal history of a falling droplet, an equation describing the rate of heat energy lost from the surface of the droplet to the surrounding gas is used and is given by:

\[
q = h_{eff} A (T_m - T_0)
\]  

(4)

where \( h_{eff} \) is the effective heat transfer and consisting of the additive contribution of convection, conduction and radiation heat transfer mechanisms, \( A \) is the surface area of the droplet, \( T_m \) is the droplet surface temperature and \( T_0 \) is the gas temperature. It is assumed that the surface temperature of the droplet represents the entire droplet temperature, since the internal temperature gradient within the droplets are negligible (Biot <0.1) [17]. It is also assumed that the temperature increase due to surface oxidation is negligible (see appendix 1). The effective heat transfer coefficient is mostly dominated by convection in low temperature alloys but for the high temperature alloys, radiation heat transfer can be significant.

One approach to quantify the convective component (\( H_c \)) of \( h_{eff} \) has been through the use of semi-empirical equations (e.g. Ranz-Marshall or Whitaker) [18, 19] where the Nusselt number (\( Nu \)) is averaged over the entire droplet surface (Equation 5).

\[
Nu = \frac{h_{cd}}{k_g} = A + B \cdot Pr^m \cdot Re^n
\]  

(5)

where the Prandtl number, \( Pr \), in Equation (5) is given by

\[
Pr = \frac{C_p \mu_g}{k_g}
\]  

(6)

and \( d \) is the droplet diameter, \( k_g, C_p, \rho_g \) and \( \mu_g \) are the conductivity, heat capacity, density and viscosity of the gas, respectively, and all other parameters are constants.

Wiskel et al. [17] modified the Whitaker correlation in a way that it takes into account the variability of the gas conductivity across the boundary layer. Equation 7 shows the modified Whitaker correlation:
\[ \text{Nu} = \frac{h_c D_s}{k_g} = 2 \frac{C}{k_s (m + 1)} \left( \frac{T_{m+1} - T_g}{T_s - T_g} \right) + \left( 0.4 \text{Re}^{1/2} + 0.06 \text{Re}^{2/3} \right) \text{Pr}^{0.14} \left( \frac{\mu_e}{\mu_s} \right) \]  

(7)

where from the variation of gas conductivity with temperature \((k_g=C\times T^m)\) for argon \(C = 1.86 \times 10^{-4}\) and \(m = 0.7915\) [19], the model was able to closely predict the range of time and distance in which the droplets completely solidified under the condition that \(k_s\) be evaluated at the metal droplet surface temperature and the \(Re\) and \(Pr\) numbers evaluated at the free stream gas temperature. Table 1 lists the properties of pure copper and argon which were used in the model.

| Temperature (°C) | Thermal conductivity (W/K.m) | Specific heat (J/kg.K) | Density (kg/m³) | Viscosity (m²/s) |
|-----------------|-----------------------------|------------------------|----------------|-----------------|
| Copper          | 1400                        | 325                    | 532            | 7722            |
| Argon           | 40                          | 0.01853                | 20.80          | 0.287           | 0.021           |

4. Results and Discussion

4.1. Online measurements

The mean droplet diameter \((\text{D50}=565\mu m)\) and the standard deviation \((\sigma=1.52)\) were measured using shadowgraph. The velocity versus droplet size of the falling droplets as measured by the shadowgraph is shown in Figure 3. For this measurement, the shadowgraph was continuously collecting droplet size and velocity data at 5Hz frequency. A total number of 6100 droplets were measured during the entire atomization time of two minutes. From Figure 3, each droplet size can have a range of velocities, with larger droplets having a smaller range than smaller ones.

![Figure 3](image-url) Velocity as a function of droplet size of falling droplets collected by shadowgraph system at 10cm below the nozzle plate.
While the shadowgraph was collecting the velocity and diameter of falling droplets at the center of the plume, the DPV-2000 was measuring the radiant energy of the droplets from the same location. Figure 4(a) shows the signal counts of each droplet that DPV-2000 measured at two wavelengths. The area under the curves shown in Figure 4(a) represents the radiant energy and as such it is a dimensionless value. The radiant energies measured for different droplet sizes at two different wavelengths are shown in Figure 4(b). The measurement was done at 10cm below the nozzle plate. It can be seen that larger droplets have higher radiant energy.

![Figure 4](image)

**Figure 4.** (a) The signal counts measured at two different wavelengths for a single droplet, (b) Radiant energy vs. droplet diameter measured by DPV-2000 at 10cm below the nozzle plate.

### 4.2. Droplets temperature calculations

To model the temperature of a given droplet size at different distances from the nozzle plate, the droplet’s initial temperature (i.e. atomization temperature) and initial velocity (i.e. the exit velocity from the nozzle plate) are required. The initial temperature in this experiment was 1400°C, and the initial velocity of the droplets can be found using the instantaneous velocities measured by shadowgraph (Figure 3) and the model discussed in section 3. The mean droplet diameter (D50=565μm) is used as an example to show the process of droplet temperature calculations.

A range of velocities was measured for each droplet size, as shown in Figure 3. Running the model under different initial velocities reveals the initial velocities which result in the measured values at the 10 cm distance for a 565 μm droplet size (Figure 5). Thus, the initial velocity for this droplet size ranges from 0.52 to 1.36 m s⁻¹. For a given droplet size, based on the frequency of velocity measurements obtained by the shadowgraph (Figure 3), a mean initial velocity and a standard deviation is determined.
**Figure 5.** Velocity as a function of distance of droplets from the nozzle plate for the 565μm droplet size, generated from the model.

Using the range of calculated initial velocities and the initial temperature of melt in the model, the temperature of a 565μm droplet at different distances from the nozzle plate can be determined. Figure 6 shows the temperature-distance relationship of a 565μm droplet with different initial velocities. It is evident that different initial velocities result in different temperature at a given distance. The average of these different temperatures will be used to represent the temperature of each droplet size.

**Figure 6.** Temperature vs. distance from nozzle plate (plotted using the model). Dashed line shows distance from the nozzle plate.
4.3. **Droplets acceleration calculations**

In order to investigate the level of gravity of falling droplets during solidification, the acceleration of the droplets must be calculated. For this purpose using the average initial velocity, the velocity-time profile of a 565μm droplet is plotted in Figure 7. Furthermore, the temperature-time profile of the same droplet as a function of time is included. At the melting point of copper (1084°C), the corresponding time (0.26s) to reach this equilibrium solidification temperature is determined. At this time of the trajectory, the derivative of the velocity results (shown in Figure 7) is used to calculate the acceleration of the droplet. This calculation was performed on different droplet sizes at their respective times to reach the solidification temperature. The results are shown in Figure 8 and compared with the gravitational acceleration.

![Figure 7](image1.png)

**Figure 7.** Temperature and velocity as a function of droplet traveling time, for 565μm droplet.

![Figure 8](image2.png)

**Figure 8.** Acceleration of different sized droplets at a time in which the droplets temperatures reach to the melting point. Dashed line shows the gravitational acceleration.
The variation in the amount of acceleration experienced by each droplet size in Figure 7 is due to different frequencies of velocities for each particle size. It can be seen that the droplets acceleration are smaller than the gravitational acceleration. This is due to the buoyancy and drag forces applied from the surrounding gas atmosphere on the falling droplets. Buoyancy and drag forces resist the droplets falling and decrease the effective acceleration on the droplets applied by the gravitational acceleration. Also, the droplet acceleration of about 7 m/s² shows that the droplets do not experience their terminal velocities at the time of the solidification. This means that the gravity is still the dominant force acting on the droplets until they reach their equilibrium solidification temperature.

4.4. Calculated temperature and measured radiant energy correlation
As was shown in section 4.1, the radiant energy measured by DPV-2000 is a dimensionless value and is dependent on a number of variables in the system, including emissivity, view factor, and the nature of the droplet surface. It is therefore important at this stage of the study to determine if there is a correlation between the calculated droplet temperatures and the measured radiant energies measured by the DPV-2000. Figure 9 shows the preliminary results of temperature-energy values achieved at 10 cm below the nozzle plate for different droplet sizes. It can be seen that at a 10 cm distance from the nozzle plate, larger droplets have higher temperature than the smaller ones. The measured radiant energy also follows the same trend. Ongoing research is directed at investigating the sensitivity of the DPV-2000 in measuring of the rate of change of the radiant energies at the two wave lengths and comparing them to the predicted cooling rate of liquid droplets. Subsequent efforts will focus on exploring the ability of the DVP 2000 to detect the undercooling of the droplets.

![Figure 9](image)

**Figure 9.** Temperature and Energy as a function of droplet diameter, in 10cm below the nozzle plate.

5. Summary
1- Particle size, particle size distribution and velocity of Impulse Atomized droplets have been measured using a Shadowgraph device.
2- Radiant energy of molten copper droplets was measured with DPV-2000 at 10 cm below the nozzle plate.
3- Acceleration of falling droplets near the melting point is close to gravitational acceleration.
4- Predicted droplet temperature at 10cm from nozzle plate decreases with decreasing the droplet size. Measured E(λ₁) and E(λ₂) also decreases with decreasing droplet size.

6. Acknowledgments

The authors would like to thank Natural Sciences and Engineering Research Council of Canada (NSERC) and Canadian Space Agency (CSA) for the financial support and Nils Ellendt for the modeling.

7. Appendix

In the model, it was assumed that the temperature increase due to oxidation is negligible. Here, the validity of this assumption will be investigated.

As mentioned in the experimental section, the amount of oxygen in the atomization tower at the start of the run is about 90ppm. The oxidation of the droplets during atomization under this condition is thermodynamically favorable and is exothermic. Thus, it may result in a temperature increase of the droplet. An estimate of the maximum increase in the droplet temperature is performed.

Liquid copper oxidizes according to the following reaction: [21]

\[ 2\text{Cu}(l) + \frac{1}{2}\text{O}_2 = \text{Cu}_2\text{O(s)}, \Delta H= -188.3 \text{kJ/mole} \] (8)

Volume of the atomization tower is about 1.67m³ and total oxygen content of the tower atmosphere is 90ppm. This gives 6.14×10³ moles of O₂ available to oxidize 900g of Cu used for atomization. This amount of Cu would produce about 1.2 x 10⁶ droplets of 565μm (D50) (a monosized droplets distribution is assumed).

Considering the total number of droplets, total available oxygen in the tower and the oxidation reaction, the average thickness of oxide layer on each droplet may be calculated to be about 240nm. This amount of oxidation was found to increase the temperature of a single Cu droplet by 5°C. This temperature increase is less than the error bars shown in Figure 9. Thus the assumption of neglecting surface droplet oxidation in the model is valid.

7. References

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