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## SOLIDIFICATION OF DROPS

## IN THE MSFC DROP TUBE

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# Solidification of Drops in the MSFC Drop Tube 

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## I. Abstract

Silver drops ( $99.9 \%, 7 \mathrm{~mm}$ diameter) were levitated, melted, and released to fall through the Marshall Space Flight Center's 105m drop tube in an He-6\%H atmosphere at 170 degrees superheat. The extent of solidification during the $\sim 4.6 \mathrm{~s}$ of free fall time prior to impact was measured experimentally and computed numerically using a newly developed solidification heat transfer model. Comparison of the experimental observation of the fraction of liquid transformed with the numerical solutions showed reasonable agreement. Possible modifications of the model, in an attempt to close the gap between the experiment and the model comparison are discussed.

## II. Introduction

The intent of this work is to investigate solidification behavior in "bulk" spheres, i.e. those having diameters greater than 1 mm . An experimental study of drop solidification necessitates the sample to be free of a container. This constraint can be relaxed by levitating the sample in an electromagnetic field or, as utilized here, by letting it free fall a significant length through a controlled environment, i.e. a drop tube. Many studies have utilized drop tubes to investigate various aspects of solidification phenomena and processing. (See [1-3] and references therein.)

Szekeley and Fischer [4] among others [5,6] have numerically studied a simple model of the solidification behavior of pure materials with radiation as the transport mechanism for heat outward through the surface of the solidifying shell. McCoy et al. $[7,8]$ simultaneously solved the dynamical equations for drop position and velocity in order to provide an approximate value of the heat transfer coefficient at the surface of an isothermal drop as a function of the drop time, or distance covered, during free fall. Their results showed that the amount of microgravity time the drop actually experienced was very short. In addition, allowance was made for the influence of heat transfer on solidification.

The goal of this work is to present experimental observation showing the extent of solidification of a liquid Ag sphere of diameter 7 mm and 170 degrees (K) superheat, and to correlate this finding with the solution to a model of droplet solidification during free fall. The details of the heat flow and the dynamics of the interface motion within the sphere, as well as the external environmental influence on the convective heat transfer adjacent to the sphere surface, are explicitly included in the numerical procedure. By comparing the experimental results with theory, insight into the
important physical parameters affecting the solidification microstructure of drops can be illuminated.

## III. Experimental Procedure

Ag shot ( $99.9 \%$ purity) was weighed with the intent of producing a 7 mm diameter spherical drop [1-3]. The shot was placed in a graphite crucible and melted with the aid of an hydrogen torch. The subsequent sample was generally shiny and oblately spherical in appearance. The sample was placed in a pedestal in the bell jar atop Marshall Space Flight Center's 105 meter drop. After a mixed He-6\% atmosphere was established within the tube, at an overall pressure of $\sim 690$ torr, the individual sample was raised within the coil of an electromagnetic levitator (EM). A two color pyrometer continually recorded the temperature of the levitating sample. The thermal arrest which was observed during melting of the $\mathrm{Ag}\left(\mathrm{T}_{\mathrm{M}}=1233.8 \mathrm{~K}\right)$ sample served as a calibration reference. After melting, the sample oscillated slightly and proceeded to heat at a rate of $\sim 60 \mathrm{~K} / \mathrm{s}$. Once a preset superheat temperature was reached power to the coil was automatically cut and the sample fell the length of the tube; the dropped sample was retrieved after the run. Review of the temperature versus time plot shows the release temperature to be within $\pm 5 \mathrm{~K}$ of that stated. Finally as indicated by the purity of the Ag used, no attempt was made to promote undercooling during free fall, and no recalescence was observed. The physical state of the 7 mm drop was found by an examination chunks of solid pieces retrieved after impact. The sample dropped with little if any supercooling in the liquid prior to solidification.

## IV. Model Development

A numerical procedure is developed to solve equations governing heat transfer within the drop, heat transfer from the drop to the surrounding medium and the kinetics of solidification within the drop. After release of the molten drop, it is assumed the sample remains a sphere of radius $\mathrm{R}_{\text {out }}$ throughout free fall. During this time, heat may be expelled into the surrounding medium by a combination of convection and radiation.

After the drop is released and sufficient heat has been transported to the surrounding medium, such that the outer surface temperature reaches the bulk melting point, solidification begins uniformly along the outer surface. Within the solid or liquid material, radially symmetric heat flow is the governed by the heat equations, each phase having unique but constant values of thermal diffusivity. At the solid-liquid interface, the standard conservation of heat condition balances the latent heat released to the jump in heat fluxes at the solid-liquid interface. In this work, the interfacial temperature is assumed to remain at the bulk melting point. Solidification continues until impact ( $\sim 4.6 s$ ) at the drop tube bottom. More details are provided in references [13].

The heat conducted to the outer radius from within the sphere is transported to the environment by convection and/or radiation using a standard mixed boundary condition. The value of the heat transfer coefficient is assumed to depend on the Prandtl and Reynold's numbers for the free falling drops. Since $\operatorname{Re}$ and $\operatorname{Pr}$ depend upon
the free fall velocity of the drop the dynamical equations for the drop position and velocity must be solved simultaneously with the temperatures throughout the sphere as a function of time. The expressions for the drop speed and acceleration are as in references $[7-9]$. The internal solid-liquid interface position is determined when solidification is occurring and used to compute fraction transformed.

## V. Results

For a given Ag drop of diameter, 7 mm in an $\mathrm{He}-6 \% \mathrm{H}$ atmosphere the calculation were was carried out for the superheat of 170K. In Figure 1, results for the 7 mm drop are shown. During the first 2.78 seconds of free fall the drop has lost its 170K superheat and established a nearly uniform temperature profile. In the remaining time before impact ( $2.78 \mathrm{~s}-4.62 \mathrm{~s}$ ), the solid-liquid interface propagates from the outer surface of the drop inward. Comparison of the shell thickness computed numerically and measured experimentally agree to within 50 percent, with the numerical results underestimating the fraction transformed.

Ag spherical drops inftally superheated $170^{\circ} \mathrm{K}$


Figure 1: The radius of the liquid interior to the sphere as a function of time measured from the time of release of the drop. Radii are plotted for a 7 mm diameter sphere. The time indicated on the plot corresponds to the time at which solidification commenced at $\mathrm{R}_{\text {out }}$. The initial temperature of the drop was 170 K above the melting point.

## VI. Conclusions

Liquid silver spheres having diameter 7 mm were released into free fall in a gaseous atmosphere of $\mathrm{He}-6 \% \mathrm{H}$ at 170 degrees superheating. Observations of and measurements of the retrieved sample remnants were compared to the results of a fully transient, spherically symmetric model of the cooling and solidification which occurs during free fall. The model accounts for the variation in the gas properties and explicitly tracks the solid-liquid interface as it progresses through the liquid during the solidification phase.

The results compared favorably. Differences in the observed solidification structure with the results of the calculations indicate additional factors need future consideration in modeling efforts to provide a more quantitative comparison. During solidification the temperature in the interior liquid was seen to remain constant, whereas a nearly linear temperature gradient developed in the solid shell. This has implications related to the morphological instability and the subsequent development of dendritic microstructures which may develop in alloy drops.

## VII. Acknowledgments

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