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An Approximate Analysis of the Formation of a Buoyant Solid Sphere in a Supercooled Melt

A. D. Solomon
D. G. Wilson
V. Alexiades

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FOR THE UNITED STATES
DEPARTMENT OF ENERGY

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes—Printed Copy: A02 Microfiche A01

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ORNL-6212

Engineering Physics and Mathematics Division

Mathematical Sciences Section

AN APPROXIMATE ANALYSIS OF THE FORMATION OF A BUOYANT SOLID SPHERE IN A SUPERCOOLED MELT

A. D. Solomon

D. G. Wilson

V. Alexiades

Date Published: March 1986

Research sponsored by the
Applied Mathematical Sciences Research Program
U.S. Department of Energy
Office of Energy Research
and
Microgravity Science and Applications Division
of the
National Aeronautics and Space Administration
nder contract H-79749B

Prepared by the
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831
operated by
Martin Marietta Energy Systems, Inc.
for the
U.S. DEPARTMENT OF ENERGY
under Contract No. DE-AC05-84OR21400

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ABSTRACT

A mathematical model is presented for the idealized formation and development of a bouyant sphere solidifying in an infinite pool of supercooled liquid. The solid and liquid are of the same pure material and the solid is less dense than the liquid. Initially the liquid is at a uniform temperature that is below its equilibrium freezing temperature, T_{cr}, but above the so called hypercooled temperature, $T_{cr} - H / c_L$. Here H and c_L are the latent heat of solidification and the specific heat of the liquid respectively. An approximate solution is derived based on the Megerlin approximation method.

INTRODUCTION

In an earlier paper [1], we have described the process of solidification of an alloy system (HgCdTe) in a one-dimensional cylindrically symmetric case. The HgCdTe system is so constituted that the solid phase is poorer in its mercury content than the liquid phase with which it is in equilibrium. In the following lines, we begin to look at the implications of this fact in the context of the following process:

> A cylindrical ampule of liquid alloy is subjected to an axially symmetric heat sink. Soon after this occurs, we find a thin solid layer moving inward from the ampule boundary. At the same time, the liquid has become constitutionally supercooled. As a result, solid crystal "nodules" form in the liquid; since the solid is poorer than its surrounding liquid in mercury content, the solid is less dense than the liquid and the nodules rise upward. As they rise, the nodules continue to grow in

size. After some additional time interval, the collection of rising and growing nodules becomes so large that the flow cannot continue. Hence, the resulting composition of the solidified alloy is far more complex than would be that obtained from simple inward solidification.

A number of different factors are present in this process. The first is the model of the alloy solidification process, and, in particular, the treatment of the material when it is constitutionally supercooled. This can be dealt with as in [1-3]. Another factor is the manner in which nodules of given radial size (less than and greater than critical size) are formed. Relations for the crystal nucleation frequency in a binary metallic melt can be found, for instance, in [4] and for pure material in [5]. The actual buoyant rise of the less dense nodule has not, to our knowledge, been dealt with. Nevertheless, the "inverse" problem of thermosolutal convection in the liquid from a planar solid front is studied in [6]. The buoyant rise of bubbles in liquid has been widely treated (see e.g. [7]). Do all the solid nodules observed at some later time in the melt come into existence as a result of homogeneous nucleation? For certain materials it has been observed that solid pieces are broken off from other solid pieces [8]. It is believed that this is not a significant factor for the HgCdTe system. Similarly, at this state of our efforts, we are ignoring the effects of natural convection in the melt [9] for a variety of reasons related to ampule size and the manner of cooling used.

In this paper we take an initial step towards the simulation of the above process by considering the much-simpler case of the rise of a growing solid sphere in a denser, supercooled liquid of the same pure material. After formulating the problem, we develop some simple approximations for the size of the sphere and its velocity. These relations are incorporated into a "global" ampule model in a later study.

NOMENCLATURE

c	specific heat (kJ/kg-°C)
m	mass of sphere (kg)
g	gravitational constant (m/s^2)
H	latent heat (kJ/kg)
v	sphere velocity (m/s)
T	temperature (°C)
k	thermal conductivity (kJ/m-s-°C)
T_0	initial melt temperature (°C)
T_{cr}	melting temperature (°C)
F	force (kg-m/s^2)
ν	kinematic viscosity (m/s^2)
α	diffusivity (m^2/s)
ρ	density (kg/m^3)

Subscripts

S	solid
L	liquid

MODEL DEFINITION

Suppose that at time $t = 0$, an infinite pool of material is in a liquid, supercooled state at a uniform temperature $T_0 < T_{cr}$. We will also assume that it is not hypercooled; hence

$$T_0 > T_{cr} - H / c_L \ . \tag{1.1}$$

At the initial time a solid sphere of some radius $R_0 \geqslant 0$ is formed spontaneously. We assume that the solid density is less than that of the liquid,

$$\rho_S < \rho_L \ ; \tag{1.2}$$

hence the sphere will rise. At any later time $t > 0$, let $R(t)$, $\mathrm{v}(t)$ denote the radius and rise-velocity of the sphere, respectively. Let $m(t)$ be its mass,

$$m(t) = \frac{4}{3} \pi \rho_S R^3(t) \ .$$

From Newton's Law,

$$\frac{d}{dt} (m(t)\, \mathrm{v}(t)) = F_1(t) - F_2(t) \ , \tag{1.3}$$

with $F_1(t)$ the buoyant force (due to (1.2)) and $F_2(t)$ the resistive force to the sphere's movement. Then

$$F_1(t) = \frac{4}{3} \pi g (\rho_L - \rho_S) R^3(t) \ ,$$

while by Stokes Law

$$F_2(t) = 6 \pi \nu \, \mathrm{v}(t) R(t) \rho_L \ .$$

Hence from (1.3), we obtain

$$\frac{d}{dt} (\mathrm{v}(t) R^3(t)) = g \left(\frac{\rho_L}{\rho_S} - 1 \right) R^3(t) - \frac{9}{2} \frac{R(t) \nu \, \mathrm{v}(t) \rho_L}{\rho_S} \ . \tag{1.4}$$

This is the relation linking velocity and buoyancy of the sphere.

Denote the surface of the sphere by $S(t)$. At this surface (ignoring effects of surface tension, surface transfer, etc. [10]), we now derive an energy conservation relation. Let us employ cylindrical coordinates (r,z) with the sphere assumed to be moving along the vertical z axis. If the center of the sphere is at the height

$$z = \zeta(t)\,, \tag{1.5}$$

then the sphere can be represented parametrically as

$$(z - \zeta(t))^2 + r^2 = R^2(t)\,. \tag{1.6}$$

Energy conservation at the surface S, together with the assumption that it is a T_{cr}-isotherm, implies that the rate of energy release from the growing sphere is

$$\frac{d}{dt}\left\{\frac{4}{3}\,\pi\rho_L\,H\,R^3(t)\right\} = \int\int_{S(t)} k_L\,|\mathrm{grad}T|\,d\sigma \tag{1.7}$$

$$= 4\pi k_L\,R^2(t)\,|\,\mathrm{grad}_{\mathrm{avg}}T\,|\,. \tag{1.7a}$$

Here, the subscript "avg" denotes a surface mean value.

The conditions (1.4), (1.7) and

$$T\,|_S \equiv T_{cr} \tag{1.8}$$

constitute the front conditions for our problem. Within the sphere $T \equiv T_{cr}$; on the other hand, in a neighborhood of the sphere, the liquid has a (varying) temperature distribution.

In the next section we will find an approximation for the functions v, R and T.

AN APPROXIMATE SOLUTION

A triplet of functions $R(t)$, $\mathrm{v}(t)$, $\zeta(t)$ satisfying the velocity-buoyancy relation (1.4) can be found in the form

$$R(t) = a\sqrt{t} \ , \tag{2.1a}$$

$$v(t) = 2\,b\,t \ , \tag{2.1b}$$

$$\zeta(t) = b\,t^2 \ , \tag{2.1c}$$

with a, b satisfying the relation

$$b\left\{5a^2 + \frac{9\,\nu\,\rho_L}{\rho_S}\right\} = a^2 g\left(\frac{\rho_L}{\rho_S} - 1\right). \tag{2.2}$$

In the liquid we will seek a temperature approximation $T(r\,,z\,,t)$ of the form

$$T(r,z,t) = T_{cr} + (T_{cr} - T_0)\left\{\exp\{-A(t)\,[r^2 + (z - \zeta(t))^2 - R^2\,]\} - 1\right\}, \tag{2.3}$$

with $A(t)$ an unknown function of t. $A(t)$ will be sought so that

i) the heat equation holds at the front S;

ii) energy conservation holds at S;

iii) the buoyancy-velocity relation is obeyed.

Note that i represents the basic assumption of the Megerlin approximation method [11].

By a simple calculation, we find that at the surface $S(t)$,

$$\frac{1}{r}\,(r\,T_r)_r = 4A^2r^2\,\Delta T - 4A\,\Delta T \ , \tag{2.4a}$$

$$T_{zz} = -2A\,\Delta T + 4A^2(z - \zeta)^2\,\Delta T \ , \tag{2.4b}$$

$$T_t = 2A\,v\,\Delta T\,(z - \zeta) + 2A\,\Delta T\,R\,R' \ , \tag{2.4c}$$

$$T_r = -2A\,r\,\Delta T \ , \tag{2.4d}$$

$$T_z = -2A\,\Delta T\,(z - \zeta) \ . \tag{2.4e}$$

Here $\Delta T \equiv T_{cr} - T_0$.

From the exact buoyancy relation (1.7) we find

$$A(t) = \frac{\rho_L H R'}{2 k_L \Delta T R} . \tag{2.5}$$

Similarly, from the heat equation

$$T_t = \alpha \left\{ \frac{1}{r} (r T_r)_r + T_{zz} \right\}$$

evaluated at the front S, we find

$$v(z - \zeta) + R R' = 2 \alpha A R^2 - 3 \alpha . \tag{2.6}$$

Here, taking the mean value of $z - \zeta$ as zero yields

$$A = \frac{R R' + 3 \alpha}{2 \alpha R^2} . \tag{2.7}$$

But $R(t) = a \sqrt{t}$: hence we obtain

$$a = \left\{ \frac{6 \alpha_L St}{1 - St} \right\}^{1/2} , \tag{2.8}$$

$$A(t) = \frac{a^2 + 6 \alpha_L}{4 \alpha_L a^2 t} . \tag{2.9}$$

Note that the assumption that the liquid is not hypercooled implies that

$$St = \frac{c_L (T_{cr} - T_0)}{H} < 1.$$

Knowing a, we now may obtain b from (2.2), and so the derivation of the approximation is complete.

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145. Dr. Hatten S. Yoder Jr., Director, Geophysical Laboratory, Carnegie Institute of Washington, 2801 Upton Street NW, Washington, DC 20008

146. Professor G. P. Youvaraj, Department of Mathematics, I. I. T. Kanpur - 208016, INDIA

147. Office of Assistant Manager for Energy Research and Development, U. S. Department of Energy, Oak Ridge Operations Office, Oak Ridge, TN 37830

148-175. Technical Information Center