GENERALIZATION OF INTERNAL CENTRIFUGAL
ZONE GROWTH OF METAL-CERAMIC COMPOSITES

Technical Report

Robert A. Hartzell

and

Robert F. Sekerka

Carnegie-Mellon University
Pittsburgh, Pennsylvania 15213

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

August 1976

Prepared for

THE U. S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

UNDER CONTRACT NO. E(11-1)-2407

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ABSTRACT

The electric and temperature fields within inductively heated materials have been calculated in order to better understand the essential features of the crystal growth process called Internal Centrifugal Zone Growth (ICZG). A complicated two-dimensional problem has been studied via a simple one-dimensional model which applies to infinitely long samples heated by infinitely long induction coils. Two such one-dimensional models are presented in this report. In the first model, all material properties of the sample are assumed to be independent of temperature. The coupled differential equations determining the electric field and temperature distributions within the sample are solved analytically. Resulting profiles of temperature and electromagnetic fields within the sample provide a basis for more sophisticated models.

The second model deals with materials (e.g., oxides) whose electrical conductivities are very low at room temperature but increase significantly with temperature. The differential equations for electromagnetic and temperature fields are consequently strongly coupled, resulting in multiple steady state solutions, some of which are unstable. An understanding of this instability is of utmost importance with regard to crystal growth via ICZG, because it has been observed that an increase in the electrical conductivity of the sample when melting occurs can cause a catastrophic increase in surface temperature. The modeling predicts, however, that the instability can be controlled through a judicious choice of RF frequency, sample size, and sample alloying.

Progress has been made in solving the two-dimensional differential equations. Many problems are encountered that are not present in one-dimensional modeling. The method of solution is outlined, but no solutions have yet been obtained.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product or process disclosed or represents that its use would not infringe privately owned rights.

TABLE OF CONTENTS

INTRODUCTION.....	1
CONSTRUCTION OF A TRACTABLE MODEL OF ICZG.....	14
ONE-DIMENSIONAL SOLUTIONS.....	31
TWO-DIMENSIONAL SOLUTIONS.....	83
CONCLUSIONS.....	91

INTRODUCTION

The crystal growth process called Internal Centrifugal Zone Growth (ICZG) is being mathematically modeled to gain a better understanding of its essential features and to develop a predictive capacity that will enable improvement and extension of this technique as currently practiced. In this report, particular attention is focused on the phenomenon of internal heating associated with crystal growth processes that utilize RF induction heating.

The differential equations for the electric and temperature fields within inductively heated materials have been developed and successfully solved via numerical methods. Much of the modeling has concentrated on materials whose electrical conductivities are very low at room temperature but increase significantly with temperature. A temperature dependent electrical conductivity causes the differential equations to be strongly coupled in a nonlinear manner, resulting in multiple steady state solutions, some of which are unstable. This means that the material being induction heated is able to switch from a high temperature state to a low temperature state and vice versa with only small changes in impressed field. This is analogous to thermal switching, a topic that has received considerable theoretical attention.¹

History of ICZG

In the early 1960's at Oak Ridge National Laboratories, A. T. Chapman and G. W. Clark developed a crystal growth process that had the unique distinction of being able to create a liquid zone in the interior of a pressed powder compact. Radio frequency induction heating was used to create, sustain, and translate this molten zone within the pressed rod in a manner similar to the float zone technique, except the surface of the rod was kept solid by a judicious choice of operating frequency and sample size. The obvious advantage to an interior melt is that the sample serves as its own crucible, thereby eliminating the problem of crucible reaction with the melt, a major difficulty for high-melting point materials.

ICZG was originally developed to grow single crystals of UO_2 for possible use as reactor fuel cells. Later, when attempts were made to increase the electrical conductivity of UO_2 through doping with small additions of tungsten, it was discovered, through the fortuitous mistake of a lab technician, that tungsten in concentrations on the order of 13 mole percent resulted in UO_2 - W eutectics.² ICZG was then launched in a whole new direction—the growth of mixed oxide and oxide-metal systems.

The creation of UO_2 eutectics was not without some practical use. A. T. Chapman lead a team of scientists at Georgia Tech in the development of oxide-metal eutectics grown by ICZG for use as powerful electron field

emitters.³ At present, the group at Oak Ridge is developing a Cr_2O_3 -Mo eutectic that promises to have many practical high temperature applications.⁴

Description of the ICZG Process

An illustration of the crystal growth apparatus used at C-MU to perform ICZG is provided in Figures (1-1) and (1-2). The sample is shown centered on a ceramic pedestal which is attached to a rod that can rotate and translate vertically; a vertical centering rod restrains the sample from toppling over. The induction coil is introduced into the pyrex enclosure through the bottom base plate via a coaxial arrangement of the leads with ground on the outside, and a stainless steel sheet rolled into a cylinder forms a splash/radiation shield between the sample and pyrex. Susceptors can be attached to the structure holding the centering rod and also to a sliding rod inserted through the bottom base plate.

Susceptors must be used to initiate the induction heating process if the electrical conductivity of the oxide material is too low at room temperature. The susceptors are heated by the induction coil and in turn radiantly heat the sample to a temperature at which the electrical conductivity of the sample is large enough to cause appreciable induction heating when the susceptors are parted. For large diameter samples, it is more convenient to use internal susceptors--small chips of metal embedded in the powder

Figure (1-1)

Crystal growth apparatus used a C-MU to perform ICZG. The pressed powder compact is caused to melt internally by direct coupling with the RF field through a gap in the molybdenum susceptors. The sample is then rotated and translated, causing crystal growth. The top and bottom metal base plates which are water cooled press against a pyrex cylinder, fitted with boot gaskets, to form the gas containment chamber.

Figure (1-2)

An enlargement of the sample in the previous figure. Within the sample, there are four distinct regions--crystal, unmelted solid, a melt zone, and a void which grows in size as solid is transformed into crystal.

Figure (1-1)

Figure (1-2)

compact before pressing. Internal heating originates at these high-conductivity chips and spreads to the oxide matrix as its conductivity increases. A closely related crystal growth process, called Skull Melting,⁵ also utilizes internal susceptors to raise the initial conductivity of the charge.

After the pre-heat stage, the coil current is increased until a molten zone forms, usually indicated by a sudden increase or decrease in surface temperature of the sample. If the frequency is not well suited to the sample size and the electrical conductivity of the sample material, it is very difficult to maintain a solid surface. If the frequency is too high, melting occurs on the surface rather than in the interior; whereas, if the frequency is too low, a heating instability can occur which sometimes results in a catastrophic increase in surface temperature. Much of the modeling so far has concentrated on this instability.

The rod is positioned vertically so the zone melts a small portion of a seed crystal inserted in the base of the sample. Once a stable zone is established, the sample is slowly raised through the influence of the localized RF field, causing solid to change into liquid ahead of the interior zone and liquid to transform to single crystal behind the moving zone. The resolidification of the pressed rod causes densification and the growth of a central void above the zone as shown in Figure (1-2).

Physical, Electrical, and Thermal Changes Occurring in the Sample

The sample is the focus of attention as far as the modeling is concerned, and it is the physical, electrical, and thermal changes occurring within the sample that are the subject of this investigation. Not all the changes, however, need be incorporated into the modeling; in fact, it is virtually impossible to model processes as they actually exist. Some compromise must be made with reality in order that the modeling represents a tractable problem. An idea of the complexity of the physical changes occurring within the sample as it heats is conveyed by the following:

1. When a sintered powder compact which is less than fully dense is internally melted, a central void forms and increases in volume with further melting.

2. There is often extensive plastic deformation due to stresses from large thermal gradients, density changes, and internal gas formation.

3. Internal voids in the solid migrate and coalesce, forming cracks and weakening the wall containing the melt.

4. Internal radiation in semi-transparent materials plays an important role in determining the internal temperature profiles.⁶

5. The transport properties of the melt are not easily characterized because of the complex nature of the convection currents associated with magnetic

stirring⁷, turbulent mixing, and free convection.

6. If a solute is present, the process of zone refining, and more generally, segregation may cause significant changes in electrical and thermal properties.

These changes are interlinked with each other and, in particular, to the thermal and electrical state of the sample and its environment.

Apart from the purely physical changes, the sample is saturated with an intense electric field which produces the substantial heating effect. The temperature of the sample and the electromagnetic field are, in a most sensitive manner, coupled to each other, primarily through the electrical conductivity of the sample; concisely stated, the electric field produces the heating and the heating shapes the field. Clearly, most or all of these changes occurring within the heated sample and its environment are interrelated to each other. With regard to modeling the system, this implies that the mathematical descriptions of these interrelated changes are likewise coupled.

It should not be difficult to appreciate that the system in its true state of complexity cannot be modeled in toto. Instead, a selection must be made from all the competing and coupled phenomena for the few that can be regarded as the important and essential features of ICZG, and it is these features which, combined, form the basis for an idealized model of ICZG. It is quite obvious that the two aspects of

ICZG that most fully characterize the process are the electromagnetic penetration and the thermal transport within the sample. It is these two coupled phenomena, to the exclusion of others, which are being modeled. The idealized model that focuses attention on the thermal and electrical aspects of the process and which forms the basis for the work in this report consists of the following:

1. The sample is homogeneous in composition and circular in cross-section.
2. The density of the sample material remains constant even upon melting.
3. The material is optically opaque even at elevated temperature.
4. The conductive properties of the melt are such that the boundary layer thickness in the melt is constant all along the interface.
5. The electrical and thermal properties are explicit functions only of temperature.
6. Surface radiation heat losses are proportional to the fourth power of the absolute temperature (i.e., the emissivity is independent of temperature).
7. The induction coil encircling the sample is concentric with the sample and equidistant from the ends of the sample. Moreover, the leads to the coil and the pitch of its turns do not affect the electromagnetic field within the sample.

At this stage of the modeling, only the internal melting phenomenon is being investigated; the sample is presumed to be stationary relative to the induction coil. For an actual crystal growth process in which the sample is translated slowly, a quasi-steady state develops and the solution for a stationary sample is still approximately applicable.

Predictive Capacities of the Modeling

The mechanics of modeling are first to state the mathematical equations defining the idealized model and second to solve the equations. In the next chapter, the differential equations which describe the temperature and electric fields within the sample are developed and simplified when deemed appropriate. In subsequent chapters, solutions are obtained by numerical methods (except for certain restrictive cases where analytical solutions are possible).

Once the model has been defined and the differential equations describing the model have been developed and solved, the model becomes a tool to investigate various aspects of the internal heating phenomena associated with ICZG. There are quite a number of variables in the above model, and the objective of this report is to study their effect upon such important considerations as internal temperature profiles, zone size and shape, and stability of the steady state solution with respect to small perturbations in the coil current.

One area of prime interest with regard to crystal growth is the configuration of the solid/liquid interface, because it, possibly more than anything else, affects the degree of perfection of crystals grown. Several subjects of possible investigation are listed below:

1. A relatively flat solid/liquid interface, normal to the growth direction, is essential to the growth of eutectic crystals in order that rods and lamellar plates grow parallel to each other; otherwise, dislocations and segmented rod or plate growth (colony structure) results.⁸ A very slight negative curvature to the growth interface is helpful in eliminating growth that is competitive to the orientation of the seed crystal.

2. If the induction coil geometry is not suitably matched to the electrical characteristics of a particular sample material, a torriodal-shaped zone can result.

3. The thermal gradient at the interface determines, in part, if there is morphological instability and hence possibly dendritic solidification.⁹

The model is capable of addressing itself to these questions of interest. Interface shape and size can be investigated as a function of induction coil geometry, sample length, skin depth, etc. The predictive capabilities of the

modeling are quite far reaching.

Another subject of concern is zone formation. The properties of the solid and liquid differ greatly, in oxides especially, and it is logical to expect significant changes occurring at melting. Dr. Clark's group at Oak Ridge in their work with Cr_2O_3 have been plagued by the problems associated with zone formation; once liquid begins to form, melting can occur so rapidly and violently that the sample is literally destroyed. Modeling has shown that this behavior is a result of a heating instability that can be controlled and even eliminated, through the proper choice of operating frequency, sample size, and sample alloying. In fact, the major emphasis of this report focuses on this heating instability because of its overall importance, not only towards a better understanding of ICZG, but to instability phenomena in general.

CONSTRUCTION OF A TRACTABLE MODEL OF ICZG

In this chapter, the differential equations that describe the two aspects of ICZG which most completely characterize the internal heating phenomena--the heat transport and penetration of the electromagnetic fields within the sample--are developed from first principles. In later chapters, these equations are solved via numerical methods.

Electromagnetic Fields

The electric field produced by the induction coil is generated by an oscillating potential difference across the coil. This potential difference produces both a current through the coil and an electrical charge distribution on the surface of the coil. The current through the coil creates a magnetic field whose oscillatory nature generates an electric field (non-conservative); the charge distribution on the surface of the coil produces an electric field (conservative) much like that of a series of capacitor plates spaced apart.¹⁰ The non-conservative electric field flows essentially in circles concentric with the axis of the coil; whereas, the conservative electric field is multi-directional in the vicinity of the coil. The electric field in the following equations represents the total electric field, produced by both the oscillating current

and the electrical charges on the surface of the coil.

The electric and magnetic fields within the sample and its environment are governed by partial differential equations, constructed from the fundamental relationships * of electrodynamics:

$$\text{curl } E = -\frac{\partial B}{\partial t} \quad \text{Faraday's law} \quad (2-1)$$

$$\text{curl } H = J + \frac{\partial D}{\partial t} \quad \text{Maxwell's form of Ampere's Law} \quad (2-2)$$

$$J = \sigma E \quad \text{Ohm's Law} \quad (2-3)$$

$$H = \frac{B}{\mu} \quad \text{Constitutive relations} \quad (2-4)$$

$$D = \epsilon E \quad (2-5)$$

The wave equations for the electric field, Eq.(2-6), and the magnetic field, Eq.(2-7), are derived, for constant magnetic

* All electrodynamic quantities are expressed in MKS units.

E = electric intensity field.
B = magnetic induction field
H = magnetic intensity field
J = current density
D = electric displacement field
 σ = electrical conductivity
 μ = magnetic permeability
 ϵ = dielectric constant

permeability, from the above equations.

$$\text{curl} (\text{curl} E) + \mu\sigma \frac{\partial E}{\partial t} + \mu\epsilon \frac{\partial^2 E}{\partial t^2} = 0 \quad (2-6)$$

$$\text{curl} (\text{curl} B) + \mu\sigma \frac{\partial B}{\partial t} + \mu\epsilon \frac{\partial^2 B}{\partial t^2} = 0 \quad (2-7)$$

These two differential equations are derived in most books on electricity and magnetism. Both contain damping terms proportional to the electrical conductivity of the medium. The third terms in Eqs. (2-6) and (2-7), arising from displacement currents *, are generally negligible for frequencies in the range of induction heating and will be neglected henceforth.

Under steady state conditions, the electric and magnetic fields are assumed to be harmonically varying functions of time ($\sim e^{i\omega t}$) **. With the time derivatives in eqs. (2-6) and (2-7) evaluated, the steady state wave equations are

$$\text{curl} (\text{curl} E) + i\mu\sigma\omega E = 0 \quad (2-8)$$

$$\text{curl} (\text{curl} B) + i\mu\sigma\omega B = 0 \quad (2-9)$$

* The term $\partial D/\partial t$ added to Ampere's Law by Maxwell, and called the displacement current, corresponds to the current that flows in space between two charged condenser plates. A displacement current would give rise to electromagnetic radiation.

**The electric field is represented as a complex quantity, but the actual electric field is

Real($E(r,t)$) which is equivalent to
Real($E(r)$) $\cos \omega t$ - Imaginary($E(r)$) $\sin \omega t$.

Electric Field Boundary Conditions

The electric field within the sample is largely determined by the geometry of the induction coil. To relate the electric field in the sample to its source--the coil current--it is necessary to evaluate the electric field inside the copper tubing that forms the induction coil and the field outside the coil. The field outside is very difficult to evaluate, because the differential equation describing the field, Eq. (2-8), is a vector equation which is actually three coupled differential equations, one for each component of the field.

It would be difficult, if not impossible, to solve three coupled partial differential equations in three dimensions in addition to the partial differential equation for the temperature.

The problem is quite tractable, however, when it is realized that the electric field within the sample is primarily a result of the magnetic field rather than surface charges on the induction coil. For samples with electrical conductivities sufficiently large to allow induction heating, the only important field to consider in the sample is the non-conservative electric field which flows approximately in circles concentric with the axis of the sample and coil; the equation describing this field is Eq. (2-8) in scalar form (where $E=E_\theta$):

$$\frac{\partial^2 E}{\partial r^2} + \frac{1}{r} \frac{\partial E}{\partial r} - \frac{E}{r^2} + \frac{\partial^2 E}{\partial z^2} - i\omega\sigma\mu E = 0 \quad (2-10)$$

In order to relate this angular electric field outside the coil to the current through the coil, two boundary conditions at the surface of the turns of the coil must be applied. Since the total electric field outside the coil is not being evaluated, it is not possible to calculate either the electric field inside the induction coil tubing or the angular electric field distribution at the coil surface. The neglect of surface charge effects results in some loss of information regarding all vector components of the electric field inside and outside of the coil.

Nevertheless, with the use of an approximate boundary condition, the angular component of the electric field can be uniquely determined. It should be noted that each turn represents only a small fraction of the total volume being modeled, so it is reasonable to assign to each turn an average value for the angular electric field, a different value for each turn. Each value is chosen such that Maxwell's equations, specifically,

$$\oint_{\Gamma} \mathbf{B} \cdot d\mathbf{l} = I\mu_0 \quad (2-11)$$

and

$$\text{curl } \mathbf{E} = - \frac{\partial \mathbf{B}}{\partial t} = -i\omega \mathbf{B} \quad (2-12)$$

are satisfied at that respective turn. In combined form, they constitute a boundary condition for each turn:

$$\oint_{\Gamma} \text{curl } \mathbf{E} \cdot d\mathbf{l} = -i\omega I\mu_0 \quad (2-13)$$

The electric field at each turn of the coil is assumed to

have a constant value, a different value for each turn, but curl E varies at the surface of each turn. The constant value for the electric field at each turn is chosen such that Eq. (2-13) is satisfied.

In addition to the boundary condition at the coil, the electric field must be specified at some outer boundary. In theory, the electric field is zero infinitely far away from its source, but a boundary condition of that nature is unsuitable for solutions obtained by numerical techniques. In an actual experimental apparatus, the electric field is zero within a much shorter distance due to the shielding effect of the radiation/splash shield and base plates enclosing the sample and coil. Therefore, for the purposes of this modeling, the angular electric field is considered to be zero at the inside surface of the base plate/shield metal enclosure.

Skin Depth Phenomenon

An important parameter that is often mentioned with regard to the induction heating of a material at a particular frequency is its skin depth ($= \sqrt{2/\sigma\omega\mu}$), the approximate depth to which the electric field penetrates into the sample material. The skin depth phenomenon arises from the combined effect of the diffusion of the electric field into the sample material as modulated by the reversing polarity of the field; the skin depth is the distance the field can diffuse into the sample in the time it takes for

the field polarity to change.

In materials with high electrical conductivity like metals, the induced eddy currents flow in a region on the order of the skin depth near the surface and, for an infinitely long coil and sample, act like a solenoid, creating an induced magnetic field that is uniform over most of the interior of the material and zero outside the material. The sum of the imposed and induced magnetic fields, comprising the total magnetic field inside the sample, exponentially decreases from the sample surface and is zero at the center. In materials with low electrical conductivity (like oxides at low temperature), there is virtually no interaction between the imposed magnetic field and the sample material, and therefore there is only a very small induced electric or magnetic field. Figure (2-1) illustrates the variation of the total magnetic field within each of two materials, one with a large and the other with a small skin depth, but both with the same surface magnetic field.

One would expect that elements of the behavior for both large and small skin depths would be present when the electrical conductivity varies greatly within a sample. Consider, for example, an oxide in which the center is near the melting point and the surface is 1000 C° cooler. In the surface region, the electrical conductivity is so small that the magnetic field is not expected to attenuate significantly (i.e., large skin depth limit). In the center region, the electrical conductivity is large, possibly as

Figure (2-1)

The variation of magnetic field within an inductively heated material as a function of radial position for the two limiting cases, $\delta \gg R_s$ and $\delta \ll R_s$, where δ is the skin depth and R_s is the sample radius.

much as 100 times greater than at the surface; the magnetic field is expected to be reduced considerably in this region. It will be shown later via actual solutions to the coupled differential equations that this anticipated behavior is, indeed, correct.

In general, the electrical conductivity in Eqs. (2-8) and (2-9) is a function of temperature, so solutions for the electric and magnetic fields are dependent upon how the temperature varies within the material. There are no apparent analytic solutions for the fields if the electrical conductivity varies within the sample.

Internal Temperature Distribution

Knowledge of the electromagnetic fields within the material being induction heated is only of indirect importance in the modeling of a crystal growth process like ICZG. It is actually the temperature distribution that is of primary interest. The steady state temperature distribution is the solution to the following differential equation:

$$\nabla \cdot (k \nabla T) + |E|^2 \frac{\sigma}{2} = 0 \quad * \quad (2-14)$$

where the second term is the power per unit volume induced within the sample. The instantaneous power per unit volume is equal to $(\text{Real}\{E\})^2 \sigma$; yet, because the material

* k is the thermal conductivity of the material.

has considerable thermal inertia, the effective power density is $|E|^2 \frac{\sigma}{2}$ which is a time average of the instantaneous power density.

If the sample is rotated while it is being induction heated, the internal temperature distribution is less sensitive to the asymmetrical variations in the electric field. The power density, appearing in Eq. (2-14) should then be the angular average of the power density, and Eq. (2-14) should contain partial derivatives with respect to only two of the three coordinate variables, namely r and z :

$$\frac{1}{r} \frac{\partial}{\partial r} (rk \frac{\partial T}{\partial r}) + \frac{\partial}{\partial z} (k \frac{\partial T}{\partial z}) + \frac{1}{2\pi} \int_0^{2\pi} |E|^2 \frac{\sigma}{2} d\theta \quad (2-15)$$

The formulation of a proper boundary condition for the temperature at the surface of the sample is rather straight-forward because, at high temperatures, heat loss is principally by radiation. The temperature environment of the sample is not mathematically important unless there is considerable back radiation from external susceptors. There is some heat loss through end supports, but if the contact area between the sample and supports is small, this effect is minimal.

Internal Melting

If the temperature within the sample being induction heated exceeds its melting temperature, a second phase is

formed. The thermal and electrical properties of the liquid phase can be quite different from those of the solid phase. In addition, there are convection currents and possibly turbulent mixing and magnetic stirring within the melt. Consequently, modeling of the two-phase problem is considerably more involved.

A separate set of differential equations for the temperature and electric field must be solved for the liquid phase because the thermal and electrical properties change discontinuously at the melting temperature. These additional differential equations are the same in form as Eqs. (2-10) and (2-15).

With a second phase present, a very important boundary has been added to the problem with its own special set of boundary conditions. The solid/liquid interface defines the separation between the solid and liquid phases, and at this interface, there are boundary conditions for the temperature as well as for the electric field.

The boundary conditions for the temperature at the interface are 1) the temperature is equal to the melting temperature and 2) the heat flow normal to the interface is continuous. Stated mathematically,

$$T_{\text{solid}} = T_{\text{melting}} ; T_{\text{liquid}} = T_{\text{melting}} \quad (2-16a)$$

$$k_{\text{solid}} \frac{\partial T}{\partial n} \Big|_{\text{solid}} = k_{\text{liquid}} \frac{\partial T}{\partial n} \Big|_{\text{liquid}} \quad (2-16b)$$

If there is complex fluid motion in the melt, it is not really possible to determine the temperature distribution

within the liquid; but, it is not entirely necessary either. Knowledge of the shape and size of the solid/liquid interface and the temperature distribution within the solid requires only information concerning the heat flux at the interface. If the temperature gradient at the interface is assumed to be relatively the same all along the interface, it is sufficient to be able to calculate the total power generated in the melt as the following equations demonstrate:

$$k_{\text{solid}} \frac{\partial T}{\partial n} \Big|_{\text{solid}} = k_{\text{liquid}} \frac{\partial T}{\partial n} \Big|_{\text{liquid}}$$

$$= \frac{1}{\text{zone area}} \int_{\text{zone}} |E|^2 \frac{\sigma}{2} d(\text{volume}) \quad (2-17)$$

From Maxwell's equations it can be shown that the tangential component of the electric field and the tangential component of the magnetic field (if the permeabilities of the solid and liquid are equal) are continuous across the interface. Since only the electric field and not the magnetic field is being calculated, the boundary condition for the magnetic field must be expressed as an additional boundary condition for the electric field at the interface. This is accomplished through one of Maxwell's equations, Eq. (2-1), which relates the magnetic field to the curl of the electric field. Therefore, for the magnetic field to be continuous across the interface, the curl of the electric field must be continuous across the interface. It can be shown that if the normal derivative of

the electric field is continuous at the interface, the curl is also continuous. The proof follows:

The curl, in a coordinate system whose axes are parallel and perpendicular to the interface at each point along the interface, consists of derivatives parallel and normal to the interface. The parallel derivatives in each phase are identically equal at the interface because the interface is common to each phase; therefore, for the curl to be continuous, it is only necessary to require that the normal derivative be continuous at the interface.

Consequently, the two boundary conditions for the electric field at the interface are

$$E_{\text{solid}} = E_{\text{liquid}} \quad (2-18a)$$

$$\frac{\partial E}{\partial n}|_{\text{solid}} = \frac{\partial E}{\partial n}|_{\text{liquid}} \quad (2-18b)$$

Reduction of Equations to Dimensionless Form

It is desirable to change all dimensional quantities to dimensionless quantities. This process is not unique, but a convenient transformation is the following:

temperature

$$\theta = \frac{T}{T_{\text{melting}}}$$

electric field

$$\phi = \frac{Ea}{\omega I \mu_0} R_S$$

(a is the average spacing between turns of the induction coil; R_S is the sample radius)

radial variable

$$\xi = \frac{r}{R_S}$$

axial variable

$$\zeta = \frac{z}{R_S}$$

electrical conductivity

$$\Omega = \frac{\sigma}{\sigma} \text{ in solid at } T \text{ melting}$$

skin depth

$$\delta = \sqrt{2/\mu\omega\sigma_{T_m}} R_S^2$$

radiation cooling coefficient

$$\tilde{h}_R = \frac{\text{emissivity} \cdot \text{Stefan Boltzmann} \cdot R_S \cdot T_{\text{melting}}^3}{k}$$

convection cooling coefficient

$$\tilde{h}_C = \frac{\text{convection cooling coefficient} \cdot R_S}{k}$$

ambient temperature

$$\tilde{T}_{\text{amb}} = \frac{T_{\text{ambient}}}{T_{\text{melting}}}$$

activation temperature

$$\tilde{T}_{\text{act}} = \frac{T_{\text{activation}}}{T_{\text{melting}}}$$

$$\text{e.g., } \sigma = \sigma_{\infty} \exp(-T_{\text{activation}}/T)$$

electrical conductivity in the liquid

$$\tilde{\sigma}_L = \frac{\sigma \text{ in liquid}}{\sigma \text{ in solid at } T_{\text{melting}}}$$

skin depth in the liquid

$$\delta_L = \sqrt{2/\mu\omega\sigma \text{ in liquid}} R_S^2$$

thermal conductivity in the liquid

$$\tilde{k}_L = \frac{k \text{ liquid}}{k \text{ solid}}$$

surface power density

$$\tilde{p}_V = \frac{\sigma_{T_m} R_S^4 \omega^2 I^2 \mu_0^2}{T_{\text{melting}} \cdot k \cdot a^2}$$

induction coil current (squared)

$$\tilde{I}^2 = \frac{2 I^2 R_S \omega \mu_0}{\text{emissivity} \cdot \text{Stefan Boltzmann} \cdot T_{\text{melting}}^4 \cdot a^2}$$

$$\text{or } (\tilde{p}_V \tilde{\delta}^2 / \tilde{h}_R)$$

The resulting dimensionless differential equations and boundary conditions are:

$$\text{a) } \frac{\partial^2 \theta}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial \theta}{\partial \xi} + \frac{\partial^2 \theta}{\partial \zeta^2} + \frac{\tilde{p}_v}{2} \Omega(\theta) |\Phi|^2 = 0$$

(for constant thermal conductivity) (2-19)

$$\text{b) } \frac{\partial \theta}{\partial \xi} = 0 \quad \text{at } \xi = 0$$

$$\text{c) } \frac{\partial \theta}{\partial \zeta} = 0 \quad \text{at } \zeta = 0$$

$$\text{d) } -\frac{\partial \theta}{\partial \xi} = \tilde{h}_R (\theta^4 - \tilde{T}_{\text{amb}}^4) + \tilde{h}_c (\theta - \tilde{T}_{\text{amb}}) \quad \text{at } \xi = 1$$

$$\text{e) } -\frac{\partial \theta}{\partial \zeta} = \tilde{h}_R (\theta^4 - \tilde{T}_{\text{amb}}^4) + \tilde{h}_c (\theta - \tilde{T}_{\text{amb}}) \quad \text{at } \zeta = \frac{Z_s}{2R_s}$$

where Z_s is the length of the sample

$$\text{a) } \frac{\partial^2 \Phi}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial \Phi}{\partial \xi} - \frac{\Phi}{\xi^2} + \frac{\partial^2 \Phi}{\partial \zeta^2} - \frac{2i}{\delta^2} \Omega(\theta) \Phi = 0 \quad (2-20)$$

$$\text{b) } \Phi = 0 \quad \text{at } \xi = 0$$

$$\text{c) } \frac{\partial \Phi}{\partial \zeta} = 0 \quad \text{at } \zeta = 0$$

$$\text{d) } \Phi \text{ and } \frac{\partial \Phi}{\partial \xi} \text{ are continuous at } \xi = 1$$

$$\text{e) } \Phi \text{ and } \frac{\partial \Phi}{\partial \zeta} \text{ are continuous at } \zeta = \frac{Z_s}{2R_s}$$

f) $\Phi = 0$ at the boundaries of the domain and at the susceptors

g) $\Phi = \text{constant}$ at each turn of the induction coil

h) at each turn of the induction coil,

$$\oint_{\Gamma} \text{curl } \Phi |_{\text{coil surface}} d\tilde{\ell} = \frac{ia}{R_s}$$

$$\text{where } \tilde{\ell} = \frac{\ell}{R_s}$$

ONE-DIMENSIONAL SOLUTIONS

Before attention is focused on methods to solve the fully coupled two-dimensional equations, it is valuable to consider one-dimensional versions of the partial differential equations. The two-dimensional equations simplify to one-dimensional equations when the electric and temperature fields are functions only of the radial variable. Such is the case when the sample and induction coil are very long compared to their diameters and the spacing of the turns of the induction coil are uniform and very small compared to the diameter of the coil. Although it is not possible to consider end losses and localized field effects with this model, the model is much easier to deal with mathematically and therefore can be used more effectively to investigate the effect of coupling between the differential equations, problems associated with melting, and transient behavior. There are even analytical solutions when the thermal and electrical conductivities are independent of temperature.

The one-dimensional versions of the temperature and electric field differential equations are given below:

$$\frac{d^2\theta}{d\xi^2} + \frac{1}{\xi} \frac{d\theta}{d\xi} + \frac{\bar{p}_v}{2} |\phi|^2 \Omega(\theta) = 0 \quad (3-1)$$

$$\frac{d^2\phi}{d\xi^2} + \frac{1}{\xi} \frac{d\phi}{d\xi} - \frac{\phi}{\xi^2} - \frac{2i}{\delta^2} \Omega(\theta)\phi = 0 \quad (3-2)$$

The surface radiation boundary condition is

$$-\frac{d\theta}{d\xi} = \tilde{h}_R (\theta^4 - \tilde{T}_{amb}^4) + \tilde{h}_C (\theta - \tilde{T}_{amb}) \quad (3-3)$$

and the surface electric field boundary condition is

$$\frac{d\phi}{d\xi} + \frac{\phi}{\xi} = -i \quad (3-4)$$

which is a direct consequence of one of Maxwell's equations, Eq. (1-1). Notice that the approximate boundary condition, developed for the two-dimensional problem, Eq. (2-20h), reduces to Eq. (3-4) in the one-dimensional limit. The field effects due to the charge distribution on the coil are significant only in the vicinity of the coil when the turns are closely spaced, analogous to the field leakage of two capacitor plates; as the distance between the plates is increased, the electric field due to charges extends further outside the space between the plates.

For a temperature dependent electrical conductivity, the two differential equations are coupled and nonlinear; specifically, the temperature equation is explicitly nonlinear in temperature through the electrical conductivity and implicitly nonlinear through the electric field. The nonlinear nature of the equations gives rise to multiple steady state solutions, a subject that is investigated in great depth in this chapter; but, first the coupled equations in linearized form are analyzed to gain some insight into the general form of the solutions.

Constant Conductivity Model

An analytical solution to Eqs. (3-1) and (3-2) exists for sample materials that have constant electrical and thermal conductivities. The solution to the differential equation for the electric field is

$$\Phi(\xi) = - (1 + i) \frac{\gamma}{2} \frac{J_1[(1 + i) \xi/\gamma]}{J_0[(1 + i)/\gamma]} \quad (3-5)$$

where J_0 and J_1 are Bessel functions of orders zero and one. The electric field is zero at the axis of the sample; otherwise, curl E would be infinite. When the skin depth is much larger than the sample radius ($\gamma \gg 1$), the above expression simplifies to

$$\Phi(\xi) = -i \frac{\xi}{2} \quad (3-6)$$

which is independent of the skin depth. The electric field is 90° out of phase with the magnetic field and is therefore purely imaginary in this case. When the skin depth is much smaller than the sample radius ($\gamma \ll 1$),

$$\Phi(\xi) = -(1 + i) \frac{\gamma}{2} \exp [(1 + i)(\xi - 1)/\gamma] \quad (3-7)$$

In this limit, the electric field is appreciable only at the surface of the sample to a depth of approximately the skin depth. The magnitude of the two limiting cases of Eq. (3-5) are displayed in Figure (3-1) as a function of radial position.

The power density within the material being induction

heated is equal to

$$\frac{(P_V \delta^2)}{4} \frac{\text{ber}_1^2(\sqrt{2} \xi/\delta) + \text{bei}_1^2(\sqrt{2} \xi/\delta)}{\text{ber}_0^2(\sqrt{2}/\delta) + \text{bei}_0^2(\sqrt{2}/\delta)} *$$

The integral of the power density over the cross-sectional area of the sample equals the the total eddy current heating (power per unit sample length) within the sample. An analytical expression for this integral is given below and is shown in Figure (3-2) as a function of skin depth:

$$\frac{\sqrt{2} (P_V \delta^2)}{8} \delta \frac{\text{ber}_0(\sqrt{2}/\delta) \text{ber}_0'(\sqrt{2}/\delta) + \text{bei}_0(\sqrt{2}/\delta) \text{bei}_0'(\sqrt{2}/\delta)}{\text{ber}_0^2(\sqrt{2}/\delta) + \text{bei}_0^2(\sqrt{2}/\delta)} \quad (3-8)$$

From this expression for the eddy current heating it is possible to deduce the relative ease with which a particular material can be induction heated. An analysis of this curve shows that there is poor heating for skin depths much larger and much smaller than the sample radius. Heating is minimal for very small skin depth because the electric field is confined to a small area near the surface of the sample, and for very large skin depth, there is poor heating because the electrical conductivity is small. For a skin depth of roughly one-half the sample radius, heating is at its optimum.

The solutions to the temperature differential equation for the above two limiting cases are given below:

* Ber and bei are Kelvin functions. The dimensionless quantity $P_V \delta^2$ is independent of electrical conductivity.

Figure (3-1)

The variation of the electric field as a function of radial position for a large and small skin depth.

Figure (3-2)

Total eddy current heating as a function of skin depth for a material whose electrical conductivity is independent of temperature. The dimensionless quantity $P_v \gamma^2 \delta^2$ is independent of electrical conductivity.

Figure (3-3a) and (3-3b)

The power density and corresponding temperature distributions as functions of radial position for equal eddy current heating but for very different skin depths.

Figure (3-1)

Figure (3-2)

Figure (3-3a)

Figure (3-3b)

$$\theta(\xi) = \theta|_{\xi=1} + \frac{\tilde{p}_v}{128} (1 - \xi^4) \quad \tilde{\gamma} \gg 1 \quad (3-9)$$

$$\theta(\xi) = \theta|_{\xi=1} + \frac{\tilde{p}_v \tilde{\gamma}^4}{16} (1 - \exp[2(\xi-1)/\tilde{\gamma}]) \quad \tilde{\gamma} \ll 1 \quad (3-10)$$

The surface temperature, $\theta|_{\xi=1}$, is determined by surface heat losses. A closed form solution is possible if a linearized cooling law of the form,

$$-\frac{d\theta}{d\xi} = \tilde{h} (\theta - \tilde{T}_{amb}) ,$$

is used to account for both surface radiation and convection losses. With surface losses considered, the equations for the two limiting behaviors become:

$$\theta(\xi) = \tilde{T}_{amb} + \frac{\tilde{p}_v}{128} \left(\frac{4}{\tilde{h}} + 1 - \xi^4 \right) \quad \tilde{\gamma} \gg 1 \quad (3-11)$$

$$\theta(\xi) = \tilde{T}_{amb} + \frac{\tilde{p}_v \tilde{\gamma}^4}{16} \left(\frac{2}{\tilde{\gamma}\tilde{h}} + 1 - \exp[2(\xi-1)/\tilde{\gamma}] \right) \quad \tilde{\gamma} \ll 1 \quad (3-12)$$

These temperature solutions and their corresponding power density distributions are displayed in Figures (3-3b) and (3-3a) for equal surface temperatures but for very different skin depths.

Even though the total eddy current heating is the same for both temperature distributions in Figure (3-3b), the center to surface temperature difference is much greater for the large skin depth solution. If the term $\frac{4}{\tilde{h}}$ or $\frac{2}{\tilde{\gamma}\tilde{h}}$

(in dimensional form, $\frac{4k}{hR_s}$ or $\frac{2k}{h\delta}$) is large compared to 1, the center to surface temperature difference is small. This suggests that it would be difficult to centrally melt a sample that has a large thermal conductivity and/or a small radius or skin depth. This has, in fact, been demonstrated by Dr. Clark at Oak Ridge in his efforts to centrally melt silicon. For sample sizes on the order of 1 inch in diameter, the center to surface temperature difference was not sufficient to sustain a molten zone due to the large thermal conductivity of silicon. To centrally melt silicon successfully would probably require a diameter of at least 4 to 5 inches.

This constant conductivity model represents a reasonable approximation for the induction heating of metals whose thermal and electrical properties are not generally very sensitive to temperature, but it is a very poor approximation for the induction heating of oxides whose electrical conductivities are extremely sensitive to temperature.

Temperature Dependent Conductivity Model

There is no analytical set of solutions to the coupled differential equations when the electrical conductivity is temperature dependent. The differential equations are nonlinear and coupled and must be solved by some iterative process that effectively decouples the equations within each iteration. A suitable method of solution is to treat the

nonlinear and coupled terms as having known values which can be subsequently updated from solutions to the effectively decoupled equations. This iterative process is continued until a self-consistent solution is obtained.

In each iteration, the decoupled equations are separately solved by numerical integration via Euler's modified self-starting method.¹¹ The integration proceeds from a known value and slope at one boundary, either the center or the surface. This method of integration involves, in itself, an iterative process, because the boundary conditions for the temperature and electric field are such that only a value or a slope is known at either boundary. Therefore, it is necessary to integrate over the sample domain and adjust the starting value or slope and repeat this process until all boundary conditions are satisfied. In the case of the temperature equation, it is desirable to adjust the coil current rather than the center temperature.

The solutions obtained by numerical integration are approximations insofar as continuous quantities have been replaced by discrete quantities. Reasonable accuracy can be obtained, however, if the r axis is subdivided into a sufficiently large number of segments.

Figure (3-4) illustrates this multi-nested iteration scheme. Within the two inner iteration loops (indicated by superscript j) the two differential equations are numerically integrated as though they are decoupled. In the outer iteration loop (indicated by superscript k) the values of the nonlinear and coupled terms are updated with more

Figure (3-4)

Flow shart of a computer program that solves the coupled differential equations determining the electric field (Φ) and the temperature (Θ) within a sample that has a temperature dependent electrical conductivity (Ω).

recent values. A summary of the major steps are as follows:

1. The power density at each point is calculated from the most recent electrical conductivity and electric field values.

2. These power density values are then used to calculate the temperature distribution. The temperature differential equation is integrated and the coil current is adjusted to compensate for an unbalance in the heat flux at the surface. The whole process is repeated until the surface boundary condition is satisfied.

3. The electrical conductivity at each point is recalculated from the last temperature values.

4. With these newly-determined conductivity values, the electric field differential equation is integrated. The slope of the field at the center of the sample is readjusted according to how well the surface boundary condition is satisfied and the process is repeated until the surface condition is met.

5. The whole process is repeated until a self-consistent solution is obtained.

This method of solution has been used to solve the coupled differential equations for an electrical conductivity that varies as

$$\Omega(\theta) = \exp[\frac{\lambda}{T_{act}} (1. - 1/\theta)]$$

in the solid and is constant in the liquid, at some value exceeding the conductivity in the solid at melting. A typical set of solutions for the following parameters

$$\gamma = 1.25$$

$$\sigma_L = 30.0$$

$$\tau_{act} = 2.0$$

$$\tau_{amb} = .2$$

is illustrated in depth in the next ten figures. The relationship between the sample surface temperature and impressed field (specifically, coil current squared) is displayed in Figure (3-5). It can be seen that the surface temperature is a multi-valued function of coil current. In fact, for certain values of coil current, there are as many as five steady state solutions. For a particular heating history there is only one steady state solution; however, it is necessary to solve the time dependent differential equations in order to identify which of the multiple solutions is the correct steady state.

An S-shaped curve of the form seen in Figure (3-5) is common for solutions to nonlinear equations as exhibited in the literature on thermal switching.¹² Because the electrical conductivity increases with temperature in the solid and, in addition, increases discontinuously at melting, the curve in Figure (3-5) is actually two S curves; the top S associated with melting is degenerate at its lower knee which is attributable to the discontinuity in conductivity at melting.

The next ten figures represent a study of the steady state solutions corresponding to the following dimensionless parameters:

$$\gamma = 1.25$$

$$\tilde{\sigma}_L = 30.0$$

$$\tilde{T}_{act} = 2.0$$

$$\tilde{T}_{amb} = .2$$

Figure (3-5)

The surface temperature as a function of induction coil current squared. For coil current squared equal to 21.9, indicated by the dashed line, there are five multiple solutions to the coupled differential equations. The cusp at surface temperature equal to .79 corresponds to a center temperature equal to the melting point. For the highest surface temperature indicated in the figure, 90 percent of the sample is molten.

Figure (3-6)

The variation of the temperature distributions with radial position for the five solutions; melting has occurred for solutions 4 and 5.

Figure (3-7)

The variation of the magnitude of the electric field with radial position. Solutions 1 and 2 are not shown, because they are almost identical to solution 3 except they are even more linear.

Figure (3-8) thru (3-10)

The variation of the electrical conductivity with radial position. The electrical conductivity for solution 1 is not indicated, because it is so low in value. The discontinuous increase in conductivity for solutions 4 and 5 corresponds to the solid/liquid interface.

Figures (3-11) thru (3-13)

The variation of the power density with radial position. Similar to the electrical conductivity, the discontinuous increase for solutions 4 and 5 corresponds to the solid/liquid interface.

Figure (3-14)

Comparison of power generated with power radiated as a function of surface temperature for coil current squared equal to 21.9. The five intersections of the two curves correspond to the five solutions to the coupled equations. From this graph, it is possible to identify the unstable solution which, in this case, is solution 2.

Figure (3-6)

Figure (3-7)

Figure (3-8)

Figure (3-9)

Figure (3-10)

Figure (3-11)

Figure (3-12)

Figure (3-13)

Figure (3-14)

Thermal switching is a very appropriate description for this type of instability in that the sample can change from a low temperature state to a high temperature state and vice versa with only a small change in coil current. It is possible to identify at least three ways to achieve thermal switching via Figure (3-5):

1. Steady state A, corresponding to coil current squared equal to 48, has a surface temperature of .24. A slight increase in coil current squared to 49 causes the sample to melt entirely. Thermal switching in an extreme limit:

2. Steady state B, with a surface temperature of .73 and coil current squared of 17, is changed to steady state C with a surface temperature of .21 when the coil current is reduced to 16.

3. Thermal switching associated with melting occurs at the cusp joining the two S curves (i.e., solution D). An increase in coil current squared from 27 to 28 results in formation of an internal zone and an appreciable increase in surface temperature as well.

The next eight figures, (3-6) thru (3-13), show the variation of the temperature, electric field, electrical conductivity, and power density with radial position within the sample corresponding to the five multiple solutions indicated in Figure (3-5). The five temperature and electric field solutions are displayed in Figures (3-6) and

(3-7) respectively; solutions 4 and 5 show the presence of a molten zone. The method of solution does not calculate the superheat in the melt so Figure (3-7) is drawn for zero superheat. The electric field for solutions 1, 2, 3, and 4 is nearly linear as a function of position, indicating that the effective skin depth * is large compared to the sample radius. The extreme variation of the electrical conductivity within the sample is well illustrated in Figures (3-8) thru (3-10). The power density, resulting from eddy currents, is shown in Figures (3-11) thru (3-13); the discontinuity at the interface is due to the discontinuity in the electrical conductivity. The unique phenomenon of central melting is certainly made more plausible when power is preferentially supplied to the interior of the sample.

The shape of the surface temperature/coil current curve can be significantly altered through small changes in any of the parameters of the problem; for example, in Figures (3-15) thru (3-21), the discontinuous increase in surface temperature at melting is progressively eliminated with

* The skin depth varies with position within a sample whose electrical conductivity is temperature dependent. For purposes of comparison, it is convenient to define an effective skin depth which is an approximate indicator of the over-all skin depth effect. One such definition is the following:

$$\int_0^{R_s} |E\{\delta(T), r\}|^2 \sigma(T) r dr =$$

$$\int_0^{R_s} |E\{\delta_{\text{eff}}, r\}|^2 \sigma_{\text{eff}} r dr$$

$$\text{where } \sigma_{\text{eff}} = \frac{2}{\delta_{\text{eff}}^2} \omega \mu$$

smaller δ (the dimensionless skin depth in the solid at the melting temperature). The explanation to this behavior is found in Figure (3-2). If the effective skin depth just before melting occurs is 2.0 and the effective skin depth after melting is .5, a sizable increase in power has been generated within the sample upon melting, most likely resulting in a large increase in surface temperature. On the other hand, if the effective skin depth after melting is .1, no change in power has occurred due to melting and the surface temperature probably does not exhibit a discontinuity.

The effect of activation temperature on the S curve can be observed in Figures (3-22) thru (3-24). As the activation temperature decreases, the lower knee of the S diminishes.

From the experimentalist's point of view, this analysis explains many mysteries and, better yet, suggests ways to avoid the problems associated with the melting instability.

Stability Analysis

When there are multiple steady state solutions to the coupled equations, all solutions can at best be metastable, but more likely, some are completely unstable. The solutions that are unstable can be identified through a comparison of the power generated with the power radiated, both expressed as functions of sample surface temperature. Such a comparison is illustrated in Figure (3-14) for a

Figures (3-15) thru (3-21)

Surface temperature as a function of coil current squared for different values of skin depth. The instability at melting is a sensitive function of this parameter. For skin depths that are large compared to sample size, the large increase in heating at melting can cause the entire sample to melt.

Figures (3-22) thru (3-24)

Surface temperature as a function of coil current squared for different values of activation temperature. The instability in the solid is a sensitive function of this parameter.

Figure (3-15)

Figure (3-17)

SKIN DEPTH 1.250
ACTIVATION TEMP. 3.000
ELECT. COND. IN MELT 30.00
RADIATION COOLING COEF. 1.800
CONVECTION COOLING COEF. 0.000

Figure (3-18)

Figure (3-19)

Figure (3-20)

Figure (3-21)

Figure (3-23)

Figure (3-24)

fixed value of coil current. The five intercepts of the two power curves correspond to the five steady states. An explanation of how Figure (3-14) is used to determine the stability of a particular solution is demonstrated below for solution 3:

If the surface temperature of the sample increases slightly to A' due to a momentary perturbation in the coil current, for example, the sample will lose heat because at temperature A' more power is radiated than is generated. The sample returns to its equilibrium temperature. If the sample should happen to decrease in temperature below its equilibrium temperature to A'' , more power is generated than radiated at this new temperature and the sample heats to its equilibrium value. Clearly solution 3 is stable for small perturbations. The same reasoning can be applied to solutions 1, 4, and 5 to prove their stability with respect to small perturbations. Solution 2, however, is quite unstable; any small deviation from equilibrium causes the sample to either heat to solution 3 or cool to solution 1.

This analysis generalizes to the rule that if the ratio of

$$\frac{d(\text{power radiated})}{d(\text{surface temperature})} / \frac{d(\text{power generated})}{d(\text{surface temperature})} ,$$

evaluated at the intercepts, is greater than 1, the solution is metastable (or, stable if there is only one intercept); if less than 1, it is unstable. In terms of Figure (3-5), the portion of the curve for which

$$\frac{d\theta}{dl^2}\bigg|_{\xi=1}$$

is less than zero represents unstable solutions.

Transient Solution

Of the four metastable solutions, the steady state that is appropriate for a particular heating history is determined by the time dependent differential equations. The time dependent equations for the temperature, electric field, and interface velocity (if there is an internal melt) are given below in dimensional form:

$$k \frac{\partial^2 T}{\partial r^2} + \frac{k}{r} \frac{\partial T}{\partial r} + |E|^2 \frac{\sigma}{2} = \rho_d \frac{\partial H}{\partial t}^* \quad (3-13)$$

$$\frac{\partial^2 E}{\partial r^2} + \frac{1}{r} \frac{\partial E}{\partial r} - \frac{E}{r^2} - \sigma \mu \frac{\partial E}{\partial t} = 0 \quad (3-14)$$

$$\rho_d L_f \frac{dR}{dt} = k_{\text{solid}} \frac{\partial T}{\partial r}\bigg|_{\text{solid}} - k_{\text{liquid}} \frac{\partial T}{\partial r}\bigg|_{\text{liquid}} \quad (3-15)$$

The time derivative of the electric field in Eq. (3-14) reflects two different time dependencies--the $e^{i\omega t}$ harmonic

-
- *
 L_f = latent heat of fusion
 H = enthalpy
 $H_{T_m}^S$ = enthalpy in solid at melting
 $H_{T_m}^L$ = enthalpy in liquid at melting
 R = interface position
 ρ_d = density

variation and the transient change in the electric field.

If the material being induction heated has considerable thermal inertia, it is expected that the temperature changes rather slowly with time, and this restraining influence of the temperature governs the transient change of the electric field. The time derivative of the field associated with the transient change in the field is many orders of magnitude smaller than the time derivative of the harmonic change of the field at frequencies common to induction heating. Therefore, Eq. (3-14) simplifies to a quasi-steady state equation for the field. In dimensionless form, the coupled equations are

$$\frac{\partial^2 \Theta}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial \Theta}{\partial \xi} + \tilde{p}_v |\Phi|^2 r(\Theta) = \frac{\partial H}{\partial \tau} \quad (3-16)$$

$$\frac{\partial^2 \Phi}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial \Phi}{\partial \xi} - \frac{\Phi}{\xi^2} - \frac{2i}{\gamma^2} \Omega(\Theta) \Phi = 0 \quad (3-17)$$

$$\tilde{L}_f \frac{dR}{d\tau} = \frac{\partial \Theta}{\partial \xi} \Big|_{\text{solid}} - \tilde{k}_L \frac{\partial \Theta}{\partial \xi} \Big|_{\text{liquid}} \quad (3-18)$$

An explicit Schmidt recurrence formula¹³ is used to solve the time dependent temperature equation:

$$H_i^{k+1} = H_i^k + \tilde{p}_v |\Phi_i|^2 \Omega(\Theta_i^k) \Delta \tau \\ + \frac{\Delta \tau}{\Delta^2 \xi} [\Theta_{i+1}^k (1 + \frac{1}{2i}) + \Theta_{i-1}^k (1 - \frac{1}{2i}) - 2\Theta_i^k]$$

$$* \tilde{L}_f = \frac{L_f}{H_{T_m}^s}$$

$$H = \frac{H}{H_{T_m}^s}$$

$$\tau = \frac{t k T_m}{\rho_d H_{T_m}^s R_s^2}$$

where k is the time interval and i is the radial position. The value of $\Delta\tau$ is limited by the stability criterion for this method of integration to be less than or equal to $\Delta^2\xi/2$. The electric field is calculated as before by Euler's method.

The transient temperature profiles associated with four different uniform pre-heat temperatures, used as initial conditions, are obtained for the same parameters as before. Figures (3-25) thru (3-28) show that the steady state corresponding to a pre-heat temperature of .2 is solution 1; .3, solution 1; .5, solution 3; .7, solution 3.

When the sample is being internally heated and the solid finally reaches the melting temperature, melting does not occur until the eddy current heating supplies the latent heat of fusion. In the central region of the sample, uniformly at the melting temperature, $\nabla^2\theta$ is equal to zero, simplifying the time dependent differential equation to

$$\frac{dH}{d\tau} = \frac{\tilde{p}_v}{2} |\Phi|^2 \Omega(\theta) \quad (3-19)$$

This equation is only valid in the region where the temperature is uniformly at the melting temperature before melting occurs. If the electrical conductivity is independent of temperature, Eq. (3-19) is easily integrated to yield

$$H(\xi, \tau) = 1. + \frac{\tilde{p}_v}{2} |\Phi|^2 \tau$$

Melting first occurs at the point where the enthalpy reaches its value at melting, $H_{T_m}^L$, (illustrated in Figure (3-29))

and, clearly, this occurs away from the center where the power density is zero.

At this stage of the modeling, the transient profiles with a moving interface are still under development.

Figures (3-25) thru (3-28)

Transient temperature profiles for equally spaced time intervals (but different time intervals for each figure). Parameters are identical to those of Figure (3-5). A preheat temperature of .2 heats to solution 1. A preheat temperature of .3 cools to solution 1. A preheat temperature of .5 heats to solution 3. A preheat temperature of .7 cools to solution 3. Solution 2 is unstable to any preheat temperature.

Figure (3-29)

Temperature profiles for equally spaced time intervals for the same parameters as in the previous figure except the coil current squared is 34. The first liquid forms at ξ equal to .4. The preheat temperature in this case is .7 and the enthalpy of the liquid at melting is 1.2.

Figure (3-25)

Figure (3-26)

Figure (3-27)

Figure (3-28)

Figure (3-29)

TWO-DIMENSIONAL SOLUTIONS

From the one-dimensional version of the coupled differential equations, the instability in the solid and at melting were discovered and studied; from the two-dimensional differential equations, end losses and localized field effects can be investigated. Partial differential equations are considerably more difficult to solve, especially when coupled, than ordinary differential equations, and methods of solution are fewer. The same type of iteration scheme is used to solve the coupled partial differential equations as was used to solve the ordinary differential equations. Euler's method is not possible in two-dimensions; instead, a relaxation method is used.

Discretization of the Domain Space

The continuous domain, bounded by the radiation/splash shield, bottom (or top) metal base plate, and the midplanes, is replaced by a network of discrete points which define a mosaic of rectangles. The rectangles in the sample are made smaller than those outside the sample, because the curvature of the electric field is greater in the sample. Discretization of the governing differential equations and their boundary conditions is accomplished by the replacement of the partial derivatives by finite difference approximations, and solutions to the finite difference

versions of the differential equations are obtained only at these discrete points within the domain.

There are many different forms of finite difference approximations for derivatives of any order. The most common and usually the most convenient are the central difference formulas:

$$\left. \frac{\partial \phi}{\partial \xi} \right|_{\xi=m \cdot \Delta \xi} \approx \frac{\phi_{m+1} - \phi_{m-1}}{2 \cdot \Delta \xi}$$

$$\left. \frac{\partial^2 \phi}{\partial \xi^2} \right|_{\xi=m \cdot \Delta \xi} \approx \frac{\phi_{m+1} - 2 \phi_m + \phi_{m-1}}{\Delta \xi^2}$$

The inherent error (i.e., truncation error) made in the replacement of the partial derivative by a central difference formula is proportional to $\Delta \xi^2$ if $\Delta \xi$ is small. As $\Delta \xi$ approaches zero, the difference formula approaches in value the derivative it is approximating.

The two formulas above are used to convert the coupled differential equations into difference equations. The difference equations * for the temperature are:

for $m = 0$

$$4 (\theta_{1,n} - \theta_{0,n}) \frac{1}{\Delta \xi^2} + (\theta_{0,n+1} - 2 \theta_{0,n} + \theta_{0,n-1}) \frac{1}{\Delta \zeta^2} = 0 \quad (4-1a)$$

* The grid network is numbered so the vertex of each rectangle is uniquely identifiable; for example, the point $m=1, n=3$ is located at ξ equal to $1 \cdot \Delta \xi$ and ζ equal to $3 \cdot \Delta \zeta$. $\theta_{m,n}$ is shorthand notation for the value of θ at the point $\xi=m \cdot \Delta \xi, \zeta=n \cdot \Delta \zeta$.

for $m \neq 0$,

$$\begin{aligned}
 & (\theta_{m+1,n} - 2\theta_{m,n} + \theta_{m-1,n}) \frac{1}{\Delta^2 \xi} + \\
 & (\theta_{m+1,n} - \theta_{m-1,n}) \frac{1}{2m\Delta^2 \xi} + \\
 & (\theta_{m,n+1} - 2\theta_{m,n} + \theta_{m,n-1}) \frac{1}{\Delta^2 \zeta} + \\
 & \frac{\rho_v}{2} |\phi_{m,n}|^2 \Omega_{m,n} = 0
 \end{aligned} \tag{4-1b}$$

The reason that there is a special form for the difference equation at r equal to zero is due to the special geometry of the cylindrical coordinate system that introduces the term $\frac{1}{r} \frac{\partial T}{\partial r}$ into the heat equation. At r equal to zero, $\frac{1}{r} \frac{\partial T}{\partial r}$ must be carefully evaluated (as r approaches zero, $1/r$ approaches infinity whereas $\frac{\partial T}{\partial r}$ approaches zero).

The difference equation determining the electric field is

$$\begin{aligned}
 & (\phi_{m+1,n} - 2\phi_{m,n} + \phi_{m-1,n}) \frac{1}{\Delta^2 \xi} + \\
 & (\phi_{m+1,n} - \phi_{m-1,n}) \frac{1}{2m\Delta^2 \xi} - \frac{\phi_{m,n}}{m\Delta^2 \xi} + \\
 & (\phi_{m,n+1} - 2\phi_{m,n} + \phi_{m,n-1}) \frac{1}{\Delta^2 \zeta} - \\
 & \frac{2i}{\gamma^2 \delta} \phi_{m,n} \Omega_{m,n} = 0
 \end{aligned} \tag{4-2}$$

The solution of these difference equations is accomplished by a relaxation method called successive over-relaxation.

The computer time required for convergence of the solution places a limitation on the number of grid points that can be used to represent the sample cross-section and the area outside the sample. Consequently, the radial and axial increments (i.e., $\Delta\xi$ and $\Delta\zeta$) are too large for an accurate evaluation of the electric field in the melt if the skin depth is small. An alternative method to calculate the electric field is to use an analytical solution for the electric field in the melt and join it to the numerical solution in the solid. The electric field for small skin depths (see Eq. (3-7)) is simply an exponential function of the form

$$\Phi(\ell, \eta) = \Phi_\ell \exp\left[-(1+i) \frac{\eta}{\delta_L}\right] \quad (4-3)$$

where ℓ is a point on the interface and η is the normal distance into the melt as measured from the interface.

The melt zone area is best represented in terms of spherical coordinates—a radial distance, ρ , and the angle, θ , measured from the axis. See Figure (4-1). The center of the coordinate system is chosen such that the interface positions can be defined as a single-valued function of θ . The equation for the interface can be represented as a set of parametric equations, $\rho(\ell)$ and $\theta(\ell)$, where ℓ as in Eq. (4-3) corresponds to a point on the interface. Moreover, θ can be set proportional to ℓ : $\theta = \alpha\ell$ where α is a constant of

Figure (4-1)

The melt zone is represented in terms of spherical coordinates, ρ and θ , in relationship to a center ξ_0 which is chosen such that the interface positions, $\rho(l)$, can be defined as a single-valued function of θ . η is the normal distance into the melt as measured from the interface.

proportionality that reflects the number of points defining the interface. The points on the interface do not necessarily correspond to grid points; in fact, interface points should be more closely spaced than grid points, because the interface separates two regions of very different conductivity.

An analytical expression for the total power generated within the melt is

$$4\pi \tilde{P}_{\sigma_L} \int_0^{\rho(l)} \rho (\rho \cos\{\theta(l)\} + \xi_0) |\phi_l|^2 \exp\left(\frac{-2\eta}{\delta_L}\right) d\rho \alpha \quad (4-4)$$

where η in terms of the ρ and θ coordinate system is

$$\eta = \rho(l) \hat{\rho}(l) \cdot \hat{\eta}(l) - \sqrt{\rho^2 - \rho^2(l) [1 - (\hat{\rho}(l) \cdot \hat{\eta}(l))^2]}$$

and $\hat{\rho}(l) \cdot \hat{\eta}(l)$ is a dot product between unit vectors.

It is also convenient to express the temperature gradient normal to the interface, $\frac{\partial \theta}{\partial \eta}|_{\text{solid}}$ *, as a function of l . The difference between

$$\tilde{k}_L \frac{\partial \theta}{\partial \eta}|_{\text{liquid}} - \frac{\partial \theta}{\partial \eta}|_{\text{solid}},$$

in any particular iteration, is an indication of the heat flow unbalance at that interface position.

$$-\tilde{k}_L \frac{\partial \theta}{\partial \eta}|_{\text{liquid}}$$

from Eq. (2-17) equals the power generated in the melt

* $\frac{\partial \theta}{\partial \eta}|_{\text{solid}}$ is calculated by a forward (or backwards) difference formula from neighboring temperatures in the solid.

divided by the zone area. The eddy current heating in the melt is calculated from Eq. (4-4), and the area of the melt zone is equal to

$$4\pi \sum_l [\rho(l) \cos\{\theta(l)\} + \xi_0] \propto \rho(l)$$

If the quantity

$$k_L \frac{\partial \theta}{\partial \eta} \Big|_{\text{liquid}} - \frac{\partial \theta}{\partial \eta} \Big|_{\text{solid}}$$

is equal to zero at position l , that interface position is in thermal equilibrium as established by Eq. (2-16b); if less than 0, point l on the interface should be repositioned in what is currently the solid region; if greater than 0, the point should be relocated in the liquid region. In this way, the interface gradually assumes its correct shape through the iteration process. There is no way to estimate precisely how far into the solid or liquid region the point should be moved; such considerations must be based exclusively on convergence and stability factors pertaining to the iteration process.

To maintain the stability of the iteration process, the sum of all the changes in interface positions should not be so large as to cause an appreciable change in the shape and size of the zone from one iteration to the next. If such were the case, the sudden change in the effective conductivity of the sample would disturb the convergence of the iteration process which is programmed to proceed in a gradual manner in order to insure stability. Yet, the

change in interface positions should not be so conservative that convergence is unnecessarily slowed. The change in the size of the zone from one iteration to the next is determined by the net heat unbalance at the interface. If the net heat flow is out of the zone, the zone is made to contract; if the net heat flow is into the zone, the zone is made to expand. An effective way to stabilize the iteration process is to require that the area of the interface cannot change by more than some pre-established amount from one iteration to the next; the relative movement of each individual point on the interface is calculated to equal the heat unbalance at that point times some constant factor, the same for all points, whose value is such that the newly positioned interface points yield the desired zone area.

Evaluation of the Electric Field outside the Sample

In the one-dimensional problem, evaluation of the electric field outside the sample is not necessary, because the electric field at the surface of the sample is calculated from the magnetic field which is constant between the induction coil and the sample. In the two-dimensional case, however, relating the surface electric field to the current through the induction coil can involve considerable computation and computer storage space. Modeling the area outside the sample which represents approximately 100 times the cross-sectional area of the sample is a problem without an easy solution.

It is believed that at positions fairly distant from the sample, the electric field is not appreciably influenced by the eddy currents in the sample. A fictitious boundary is created between the sample and the outer boundary formed by the metal end plates and radiation shield, and the electric field at this fictitious boundary is set equal to the electric field at the same position for an induction heating situation in which there is no sample (just an induction coil inside a metal chamber). The fictitious boundary is gradually enlarged, until at some distance from the sample, the electric field in the sample is insensitive to the position of the fictitious boundary. In this way, repeated calculation of the electric field within the whole domain for each iteration can be avoided.

Currently, no solutions have been obtained for the two-dimensional equations, but the method of solution is promising.

CONCLUSIONS

The internal heating phenomenon of ICZG has been investigated in this report through models of varying degrees of sophistication. The differential equations governing the heat transport and penetration of the electric field within the sample being induction heated have been developed and solved via analytical and numerical methods. Results so far have been obtained only for one-dimensional heating. Even so, much has been learned about the interrelationship between material and process parameters and experimental observables such as sample surface temperature, melt zone size, and stability of the steady state. A concise summary of the results of the one-dimensional modeling are stated below:

1. To achieve a sufficient center-to-surface temperature difference conducive to central melting, the parameter k/hD (where D is the smaller of either the skin depth or the sample radius) must be less than about 5. It's possible to infer from the magnitude of k/hD if internal melting is plausible for a particular heating situation.

2. The temperature dependence of the electrical conductivity is responsible for heating instabilities in the solid and at melting, but these instabilities are controllable through proper choice of operating

frequency, sample size, and sample alloying. Usually the instabilities can be eliminated through a decrease in the skin depth.

3. Solution of the time dependent differential equations shows that melting generally first occurs away from the center of the sample which, in two-dimensions, would be a zone, torroidal in cross-section.

Generalization of One-Dimensional Modeling

One dimensional modeling has accounted for the basic causes responsible for heating instabilities in the induction heating of temperature dependent materials. In actual crystal growth situations, two-dimensional effects can be expected to influence the instability phenomenon. The anticipated effects of internal susceptors, end losses, and coil geometry on steady state stability are enumerated below:

1. Internal susceptors increase the effective conductivity of the sample particularly at low temperatures, and, as a result, the instability in the solid is greatly mitigated and possibly even eliminated in some instances.

2. Radiation, convection, and conduction through sample ends amount to increased heat losses--a

stabilizing effect. If the ratio of the length of the sample to its diameter is too small, the zone shape and translation of the zone through the sample will be strongly influenced by end effects.

3. The effective skin depth of the electric field within the sample is partially determined by the geometry of the induction coil. As the ratio of the length of the coil to its diameter decreases, the penetration of the electric field within the sample also decreases. Consequently, a short coil results in a smaller effective skin depth than a long coil. Shortening a coil may eliminate an instability problem but instead may adversely affect the shape of the melt zone.

A factor that largely affects the heating instability which would be most difficult to incorporate into the modeling is the interaction of the electronics of the RF generator with the complex impedance of the sample. It is an observable fact that the current through the induction coil is influenced by the coupling efficiency between sample and coil. This is particularly true at points of instability where the interaction of the generator can tend to either stabilize or accenuate the instability depending primarily on the effective skin depth of the sample. For example, when the effective skin depth is large compared to sample radius, a sudden increase in sample temperature (and therefore an increase in conductivity), caused by an

instability in heating, likewise increases the coil current because of more efficient coupling to the sample. The increased coil current accenuates the severity of the instability. On the other hand, when the effective skin depth is small, a sudden decrease in sample temperature due to a cooling instability results in an increase in coil current, slowing down the cooling instability. A more complete understanding of the instability necessitates a modeling of the coupling between sample and generator.

Catastrophe Theory

The subject of instability phenomena has lately attracted the attention of mathematicians on account of the immergence of a new branch of mathematics called catastrophe theory¹⁵ which is exclusively devoted to the study of instabilities such as those observed in ICZG. Instability is an integral part of all facets of existence from the behavior of angry dogs to the gyrations of the stock market. Quite often the differential equations governing the instability are unknown and the knowledge concerning the instability must be generalized from what observable information is present. Fortunately in the case of ICZG, the differential equations are well known, and the instability can be studied through solutions to the equations. In catastrophe theory, the stability of a solution is related to its potential energy function, an approach which might possibly be used in the future to

determine under what conditions metastable solutions become unstable in the case of ICZG.

The theoretical discovery of the heating instability in induction-heated samples is indeed an important contribution toward a better understanding of ICZG. Although the instability probably has no value in itself and at the present is merely an annoying consequence of the heating process, an understanding of its origins is essential to being able to control and hopefully mitigate its influence. During the history of ICZG, the instability has often confounded the experimentalists, frequently leading them to remedies that only served to increase its destructive influence. With the predictive capabilities of the modeling, however, proper diagnosis of the causes can lead to elimination of the instability through intelligent engineering of material and process parameters.

REFERENCES

1. J. Duchene et al., Phys. Stat. Sol. (a) 8 (1971) 459-462.
2. A. T. Chapman, G. W. Clark, and D. E. Hendrix, J. Amer. Ceram. Soc. 53 (1970) 60-61.
3. A. T. Chapman et al., Melt-Grown Oxide-Metal Composites, Reports 1-5, ARPA Order No. 1637, School of Ceramic Engineering, Georgia Institute of Technology, December 1972.
4. J. D. Holder and G. W. Clark, Proc. Conf. on In-Situ Composites-II, to be published.
5. J. F. Wenckus et al., Study, Design and Fabricate a Cold Crucible System, AF CRL-TR-75-0213, Arthur D. Little, March 1975.
6. S. O Hara, L. A. Tarshis, R. Viskanta, J. Crystal Growth 3,4 (1968) 583-593.
7. E. Fromm and H. Jehn, British J. Appl. Physics 16 (1965) 653-663.
8. J. D. Hunt, J. Crystal Growth 3,4 (1968) 82-91.
9. R. F. Sekerka, J. Crystal Growth 3,4 (1968) 71-81.
10. R. M. Fano, L. J. Chu, and R. B. Adler, Electromagnetic Fields, Energy, and Forces, New York, Wiley, 1960.
11. M. L. James, G. M. Smith, and J. C. Wolford, Applied Numerical Methods for Digital Computation with Fortran, Scranton, International Textbook Co., 1967.

12. J. L. Jackson and M. P. Shaw, Applied Physics Letters
25 (1974) 666-668.

13. S. H. Crandall, J. Assoc. for Computing Machines 2
(1955) 42-48.

14. W. F. Ames, Numerical Methods for Partial
Differential Equations, New York, Barnes and Noble, 1969.

15. E. C. Zeeman, Scientific American (April 1976)
65-83.