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GENERALIZATION OF INTERNAL CENTRIFUGAL
ZONE GROWTH OF METAL-CERAMIC COMPOSITES

Progress Report

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ABSTRACT

Our objectives are (1) to develop a realistic model of Internal Centrifugal Zone Growth (ICZG) and (2) to utilize the predictive capacities of this model to improve present ICZG systems and to extend the use of the ICZG concept in general. In a previous year, we developed models for hypothetical (infinitely long) samples heated by long RF induction coils. We established conditions for a large surface-to-center temperature difference and predicted an instability phenomenon (spontaneous coupling) to occur on RF heating. During the present year, we have developed models for finite samples heated by finite induction coils. These models enable calculation of two-dimensional temperature profiles in solid samples. Molten zone shapes can be calculated provided that simplified boundary conditions for the RF field are employed. Experiments to test these models have been conducted at Carnegie-Mellon University and at Oak Ridge National Laboratory. The heating instability phenomenon has possibly been observed but not quantified. Design data for future experiments is being provided to Oak Ridge.

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PROGRESS REPORT

A. Technical Progress

Briefly stated, the broad technical objectives of this contract are (1) to develop a realistic model of Internal Centrifugal Zone Growth (ICZG) and (2) to utilize the predictive capacities of this model to improve present ICZG and to extend the use of the ICZG concept in general. Both theoretical (mostly computer-aided modeling) and experimental work are carried out at Carnegie-Mellon University; whereas, some experimental work is carried out in cooperation with Dr. G. W. Clark at Oak Ridge National Laboratory.

During the first year of this contract, considerable progress was made on one-dimensional models (for very long samples heated by long induction coils) that display certain essential features of ICZG; this work is reported in Progress Report COO-2407-1 and Technical Report COO-2407-2.

During the first nine months of the current contract year, much more realistic two-dimensional models have been developed and some aspects of our modeling have been tested experimentally. In particular:

1. We have formulated a set of approximate boundary conditions for the electromagnetic fields produced by finite induction coils used to heat samples of finite length.
2. We have set up and solved by relaxation techniques the finite difference equations that describe the two-dimensional electromagnetic field and temperature profiles for finite solid samples heated by finite induction coils.
3. We have developed a computational algorithm for the calculation of liquid zone shapes for samples heated above their melting point.

4. We have assembled experimental equipment at Carnegie-Mellon University and have induction heated a number of poor conductors in an effort to observe the heating instability phenomenon predicted by our one-dimensional modeling.
5. We have communicated the main predictions of our models to Dr. Clark and his group at Oak Ridge (meeting at Oak Ridge on April 13-14, communication at several scientific meetings, continuous telephone contact). In particular, we have specialized our models to a prototype material (silicon) and provided design information for experiments that the Oak Ridge group has conducted to internally melt this material.

A detailed accounting of our theoretical work during this second period of the contract is contained in a technical report which will be available very soon.¹ A brief summary follows:

i) Boundary Conditions for Electromagnetic Fields

In order to calculate temperature profiles for finite samples heated by finite RF induction coils, suitable boundary conditions must be developed for the electromagnetic fields. For an actual induction coil in the form of a helical spiral complete with inlet and outlet leads, this is a formidable axially asymmetric problem. Approximate boundary conditions that preserve the major features of the induction heating process are therefore needed for modeling. We have explored two approximate procedures:

- a) For the part of the sample surrounded by the coil, we take the surface $\vec{E}$ field to be the same as for an infinite coil; for the part of the sample

¹ R. A. Hartzell and R. F. Sekerka, "Generalization of Internal Centrifugal Zone Growth of Metal-Ceramic Composites," Technical Report No. COO-2407-4.

outside the coil, we allow the $\vec{E}$ field to drop to zero (either abruptly or smoothly over the order of the coil length).

b) We admit the actual coil configuration and solve for an $\vec{E}$ field both inside and outside the sample subject to the boundary conditions that the tangential $\vec{E}$ field at the surface of the induction coil (treated as a nearly perfect conductor) is nearly zero.

Both of these approximate procedures allow the calculation of concomitant two-dimensional temperature profiles and zone shapes (as per 2 above and ii) below) but neither is really satisfactory because the errors involved cannot be estimated. Improved handling of the electromagnetic fields for finite coils will be one of the main thrusts of our future work; details are contained in the Proposed Technical Program.

ii) Two-Dimensional Temperature Profiles

Figures 1-3 display typical two-dimensional temperature profiles calculated by solving the appropriate finite difference equations numerically by means of overrelaxation techniques. The electromagnetic field is treated by approximation procedure b) above. Details are contained in our technical report COO-2407-4. Although no molten zones are allowed to form, it is evident that they will form in the regions where the dimensionless temperature is close to the dimensionless melting point (unity). Therefore, such modeling is capable of predicting those changes in zone configuration that accompany changes in sample size, material, and operating conditions. (See 5 above and v) below for an example of the practical utilization of these results.)

iii) Liquid Zones

Whenever power levels are high enough that liquid zones can form, the sample consists of two phases. Temperatures and electromagnetic fields must then be calculated in each phase and the results matched on a boundary

CAPTIONS - FIGURES 1 - 3

Two dimensional temperature profiles calculated numerically for inductively heated samples whose electrical conductivity is a strongly increasing function of temperature. RF induction coils are depicted by dashed lines and the curved lines are isotherms labelled by fraction of the melting point. The areas shown are one quarter of the longitudinal cross sections of samples in the shapes of circular cylinders with dimensionless radius unity and dimensionless length eight. Calculations are for silicon since its material properties are well known. Each figure is labelled with the following parameters:

- f = frequency of RF generator
- R = radius of sample
- δ = RF skin depth at the melting point
- h_r = heat transfer coefficient for radiative losses
- h_c = heat transfer coefficient for conductive and convective losses
- P = total power generated within sample

Fig.1 The induction coil is concentrated near the center of the sample but there is considerable axial loss. Melting is to be expected in a toroidal region at the surface within the 0.98 isotherm. This would not be a suitable arrangement for ICZG.

Fig. 2 Same parameters as figure 1 except the induction coil is more widely spaced to offset the effect of axial heat losses. In this configuration, the molten zone would be expected to form within the 0.97 isotherm and extend through the surface of the sample; this might be a suitable configuration for crystal growth by a floating zone technique.

Fig. 3 Compared to figures 1 and 2 the radius has been increased by a factor of 5 and the frequency has been dropped by a factor of 45. Melting would be expected within the 0.97 isotherm and the resulting confined molten zone might be suitable for ICZG.

(the solid-liquid interface) whose shape must be determined as part of the problem. Provided that we treat the electromagnetic field according to approximate procedure "a" above, we can solve such problems under the following conditions:

a) The liquid in the zone is assumed to be so well mixed by convection that it is isothermal except in a thin boundary layer of invariant thickness adjacent to the solid.

b) Densities are such that no void is formed when the solid melts.

The mathematical technique consists of guessing a zone shape, solving the appropriate finite difference equations for such a zone shape, and then adjusting this guessed shape until the normal heat flux at each point along the solid-liquid interface is the same in both solid and liquid. Preliminary results have yielded some zone shapes that are convex and others that are reentrant (presumably because of axial losses). Conditions for the transition from one type of zone shape to the other along with computer-generated zone profiles will be one of our major objectives during the remaining three months of the current funding period.

iv) Experimental Work - Carnegie - Mellon University (CMU)

Experimental work at CMU has been directed toward the detection of the S curve heating instability phenomenon discovered during the first year of our work and detailed in reports COO-2407-1 and COO-2407-2. Up to now, we have tried samples such as Fe_2O_3 , PbO , TeO_2 , Cu_2O but have not been able to get sufficient heating with our 450 kHz - 10 kw generator. Figure 4 is a photograph of this and associated apparatus. We are currently setting up to conduct similar experiments with materials that have higher electrical conductivities (e.g., silicon and $\text{Cr}_2\text{O}_3 + 20$ mole % Mo) in order to couple with our available generator.

Fig. 4 Photograph of the central melting facilities being assembled at Carnegie-Mellon University. A 10 kw, 450 kHz induction generator supplies current to the induction coil which can be seen within the pyrex cylinder. Water-cooled metal plates form a seal against the cylinder so the sample can be heated in an appropriate protective atmosphere. Under the chamber are several motors which rotate and vertically translate the sample. This apparatus is, in some ways, similar to existing apparatus at Oak Ridge: however, it is more versatile and suited to testing the basic principles of ICZG.

v). Liaison with Oak Ridge

As mentioned under 5 above, we have been in constant communication with our colleagues at Oak Ridge. Particularly noteworthy are the following:

a) Scientists at Oak Ridge have observed sudden coupling and decoupling of Cr_2O_3 + 20 mole % Mo and Si samples heated by RF fields. This is believed to be related to our S-curve heating instability phenomenon but has not yet been quantified. (Incidentally, similar phenomena have been reported recently² by Dr. Wencus of Intermat Corporation for a related process called skull melting.)

b) Our calculated temperature profiles and zone shapes are in qualitative agreement with experiments on silicon at Oak Ridge. Rods of 1.5 cm diameter silicon heated at 400 kHz are both predicted and observed to melt either from the outside or in such a manner as to yield a toroidal zone shape. Calculations show that much larger rods heated at lower frequencies should have normal zone shapes; attempts will be made in the future to conduct crystal growth experiments under such conditions.

B. Expended and Contemplated Effort

This research is being conducted by the principal investigator, Professor Robert F. Sekerka, and one full-time graduate student, Mr. Robert A. Hartzell. Percentages of the expended and anticipated effort are given in tabular form below:

	Expended									Anticipated		
	1974			1975						1975		
	D	J	F	M	A	M	J	J	A	S	O	N *
Prof. Sekerka (Principal Investigator)	← 9% →			0%	← 38% →			← 50% →		← 9% →		
Mr. Hartzell (Graduate Student)	← 100% →											

*Note: This represents an average monthly charging rate of 16.8% for the 10 months of the academic year spanned by this contract period.

² Meeting of the American Association of Crystal Growth, Stanford University, July 1975.

C. Reports

Progress Report COO-2407-1	}	Submitted with first renewal
Technical Report COO-2407-2		

Progress Report COO-2407-3	}	Submitted with current renewal
Technical Report COO-2407-4		

Note: Some material from these reports (that on the S-curve heating instability phenomenon) is of archival nature and is being rewritten as an article for publication in the Journal of Crystal Growth.

D. Meetings

AEC/MIT Ceramic Science Meeting, January 13-14, 1975, report delivered by the principal investigator.

American Association of Crystal Growth, Stanford University, July 13-17, 1975, paper entitled "Modeling of Internal Centrifugal Zone Growth of Metal-Ceramic Composites" presented by principal investigator.